SECOND PERIODIC REPORT ON THE STATE OF ACID DEPOSITION IN EAST ASIA

Part I Regional Assessment

Cambodia
China
Indonesia
Japan
Lao P.D.R
Malaysia
Mongolia
Myanmar
Philippines
R. of Korea
Russia
Thailand
Vietnam

Acid Deposition Monitoring Network in EAST ASIA
(EANET)

The Second Periodic Report on the State of Acid Deposition in East Asia

Part I: Regional Assessment

***Edited by*:**

Hajime Akimoto	ACAP, Japan
Jesada Luangjame	ACAP, Japan
Hiroshi Hara	Tokyo University of Agriculture and Technology, Japan
Sergey Gromov	Institute of Global Climate and Ecology, Roshydromet and RAS, Russia
Pojanie Khummongkol	King Mongkut's University of Technology, Thailand
Willie Carandang	University of the Philippines Los Banos, College, Laguna , Philippines

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Table of Contents

List of Lead authors and contributors to the Second Periodic Report Part I

Chapter	Authors and contributors	E-mail address
1	Jesada Luangjame*	
	R. Tjang Mushadji Sutamihardja	
	Nik Muhamad Majid	
	Jiro Sato	
	Ken Yamashita	
2	Sergey Gromov*	
	Takuya Shiozaki	
	Jesada Luangjame	
3	Hiroshi Hara*	
	Takuya Shiozaki	
4	Pojanie Khummongkol*	
	Keiichi Sato	
5	Wilfredo M. Carandang*	
	Apolonio M. Ocampo	
	Hiroyuki Sase	
	Naoyuki Yamashita	
	Elizabeth M.P. Philip	
	Ryo Kobayashi	
	Simon S. Brahmana	
	Van Tuan Vu	
6	Hajime Akimoto*	
	Hiroyuki Sase	
	Keiichi Sato	
	Toshimasa Ohara	
	Duong Hong Son	
	Ken Yamashita	
	Naoyuki Yamashita	
	Takeshi Izuta	
	Junko Shindo	
	Masamichi Takahashi	
7	Hajime Akimoto*	
	Tsuyoshi Ohizumi	
	Ken Yamashita	
	Hiroyuki Sase	
	Keiichi Sato	
	Jyunichi Kurokawa	

Note * = Lead author

Drafting Committee members in Scientific Advisory Committee

Name	**Country**	**E-mail address**
Siv Kung	Cambodia	
Wang Rubin	China	
R. Tjang Mushadji Sutamihardja	Indonesia	
Hiroshi Hara	Japan	
Darounny Vilaythong	Lao PDR	
Nik Muhamad Majid	Malaysia	
Elizabeth M.P. Philip	Malaysia	
Azzaya Dolgorsuren	Mongolia	
Htwe Htwe Win	Myanmar	
Wilfredo M. Carandang	Philippines	
Cho Seog-Yeon	R. of Korea	
Sergey Gromov	Russia	
Pojanie Khummongkol	Thailand	
Duong Hong Son	Vietnam	
Nguyen Dinh Luong	Vietnam	
Adelaida Bonquin Roman	(Secretariat)	

Forward

This report is the Regional Assessment of the Second Periodic Report on the State of Acid Deposition in East Asia (PRSAD2) which is focused on EANET activities (2005-2009). However, the analyzed monitoring data during 2004-2005 is also welcomed for trend analysis. Other national and international monitoring results are also used as references for additional assessment for interpretation of EANET data.

This report included the future directions and possible impacts of air pollution on atmospheric environment and ecosystems e.g., soils and vegetation, inland aquatic systems, etc. Also highlighted are the quality assurance and quality control (QA/QC) activities, designed to ensure reliable data and consistent research activities, which were implemented jointly by EANET community to improve understanding of the acid deposition processes in the region.

There is still a large uncertainty for estimation of the regional distributions of O_3 and $PM_{2.5}$ which is harmful to human and agricultural imparts. Therefore, more extensive epidemiological studies are necessary in each EANET counties since most of previous studies are based on the data obtained in Western countries. Hence, monitoring data of these species are necessary for further development. Fortunately, some of these activities are contained in the next five-year Strategy Development Plan (2011-2015). It is hoped that this Second Periodic Report will promote better understanding of acid deposition in our region and important for further development of EANET.

Dr. Duong Hong Son

Chairperson, Scientific Advisory Committee of the EANET

Overview of the report

EANET has made progress and important achievements in the monitoring, data acquisition and management, research, and other technical issues. This is the second edition of the *Periodic Report on the state of acid deposition in East Asia* that continued to present the result of second period (2005-2009) of a scientific evaluation of the information that obtained through the regular phase activities after the first period (2000-2004) to date and provides some conclusions. This report covered the main goals and objectives of the network. The five-year data assessment is able to draw more precise and definite conclusions on temporal and spatial variations of atmospheric deposition. Most achievements were obtained under the *Strategy on EANET development (2006-2010)* which were covered almost all network activities to be coordinated among the participating countries, the Secretariat and the Network Center.

The *Second Periodic Report* was comprised of 7 chapters as follows:

- Chapter 1: Introduction
- Chapter 2: Data Quality
- Chapter 3: Wet deposition in East Asia
- Chapter 4: Dry and overall deposition in East Asia
- Chapter 5: Impacts on Ecosystems in East Asia
- Chapter 6: Other Related Studies for EANET Region
- Chapter 7: Summary and recommendations for future activities.

The report was prepared as a scientific assessment of the state of acid deposition in East Asia based on the data accumulated from the network from 2005 to 2009 (five years). The decided title is the "Second Periodic Report on the State of Acid Deposition in East Asia" (PRSAD2). The report also used some data of 2000-2004 for trend analysis and as appropriate.

Data quality of chemical analysis and evaluation is emphasized on the Inter-Laboratory Comparison which has been done annually since 1998 during the preparatory phase. The Inter-laboratory Comparison Projects are one of the activities under the quality assurance and quality control (QA/QC) programs with all EANET laboratories be participated it. Through this project, each laboratory can take advantage of the opportunity to assess their performance in established analytical procedures and identify problems in order to improve data quality. The ratio of flagged data has decreased from the start of projects in spite of an increase in the number of laboratories participating. These results prove that most laboratories satisfied the requirements for certain levels of data quality. The QA/QC was started on wet deposition. Now, there are different activities on the Inter-Laboratory comparison projects, i.e., wet deposition, dry deposition, soil, and inland aquatic environment. In 2009, they were the 12th, 5th, 11th, and 10th Inter-laboratory comparison projects on wet deposition, dry deposition, soil, and inland aquatic environment, respectively. The wet results are showing the percentage of flagged data and percentage of data that satisfied the data quality objectives (DQOs).

Wet deposition is an analytical chemistry on an acidic compound group that will release its hydrogen atom(s) as hydrogen ion(s) when dissolved into water, which suggests acid is intrinsically water soluble and therefore is deeply involved in atmospheric precipitation processes. Most of the acids in the atmosphere are not emitted directly from sources but chemically formed during the course of the transport. Major acids include sulphuric and nitric acids derived from sulphur and nitrogen oxides. Atmospheric acids thus formed acidify atmospheric water droplets including rain, where the acidity is commonly expressed in pH. The acidity of the solution will be decreased more or less, which would allow us to estimate acidification of atmospheric water by pH. Another role of atmospheric acids is to transport basic compounds by incorporating the bases through acid-base reactions. For example, during atmospheric transport of sulphuric and nitric acids, gaseous ammonia is collected to form ammonium salts, which enables the ammonia to be transported over longer distances. These precipitation acids eventually deposit onto earth's surface, which could impact ecological elements like water, soil, vegetation, and materials by acidification, and at least perturbation of acid-base balance of the elements.

The present analysis was applied to volume-weighted annual mean concentrations of the analytical ion suite in order to understand the general regional picture of acid deposition. For this purpose, all available annual means since 2000 were subjected to some data screening only to include the annual mean sets meeting the data completeness criteria for the nine ions (NH_4^+, Ca^{2+}(nss-Ca^{2+}), K^+, Mg^{2+}, Na^+, NO_3^-, SO_4^{2-}, (nss-SO_4^{2-}) and , Cl^-) including H^+ as pH.

The state of acid deposition is assessed in terms of pH in order that the discussion of “acid rain” as pH of rainwater is the indicator for focusing among the general public. A description of pH is provided as the starting point of understanding of the wet deposition. Although some more chemistry should be explored for more detailed discussion, it suffices here to mention that precipitation in East Asia is significantly acidic.

Although the EANET has accumulated ten-year record at most, the length of period does not suffice for temporal trend analysis because the wet deposition has a number of factors most of which are quite variable with time, the existing time trend could be concealed unlike gas and aerosol species. This would stress the significance of a long-term high-quality monitoring is indispensable to detect some symptoms at all for elimination and mitigation of potential impacts.

Dry deposition is the transport of gaseous and particulate species from the atmosphere onto surfaces in the absence of precipitations. The relative amount of dry deposition, as compared with wet deposition principally depends on the amount of precipitation in the region. EANET monitoring scheme has facilitated to collect both the wet and dry concentrations and to determine the amount of wet and dry depositions. Within the dry deposition study, the priority gases to be monitored include SO_2, O_3, NO, NO_2 (urban), HNO_3, HCl and NH_3 and the particulate components include SO_4^{2-}, NO_3^-, Cl^-, NH_4^+ and PM_{10}. The second priority chemicals which are not a mandatory include NO_2 (rural and remote) and $PM_{2.5}$.

The primary gases: SO_2, NO_x (NO and NO_2) and NH_3 are important in relation to atmospheric deposition. Once emitted to the atmosphere, SO_2 and NO_x become oxidized to sulphate and nitrate through both gas and aqueous –phase processes. The result of these reactions is the formation of acids in the gas phase (HNO_3, HCl, HCOOH, CH_3COOH, etc.), in the aerosol phase (sulphate, nitrate, chloride, organic acids, etc.), and in the aqueous phase.

Impacts on Ecosystems on acid deposition are definite progress in monitoring in the East Asian region. In ecological monitoring sites where more than a single observation have been made already, temporal analyses in some cases have already revealed certain trends in the changes in the soil, forest and inland water monitoring parameters. The data trend generated over the years of monitoring in the ecological stations across the EANET participating countries suggests possible acidification or nitrogen saturation in several sites. There is, however, a need to isolate other existing environmental conditions that may have contributed to such an observation.

An increase in the nitrogen flux in the soil in several ecological monitoring sites has been observed. This report has advanced a number of reasons for the occurrence of such mostly related to forest growth and/or degradation, litter production and decomposition in stands and how these processes contribute to the nitrogen dynamics in the soil and inland waters. However, as pointed out earlier in this report, the amount and kind of data generated thus far in the ecological monitoring stations are not yet sufficient to attribute fully the nitrogen flux to atmospheric deposition.

There were also evidences of decline in certain forest monitoring stations in Japan, China, and Mongolia. In the first two countries such decline symptoms were not directly and solely attributed to air pollution effects. However, in a recent study of Sase *et al.*, (2005), in Mongolia, the possible direct effects of pollutants in tree decline has been well documented. There were no evidences of decline in other East Asian countries. The decrease in biometric figures of trees in certain monitoring plots in the region were attributed to natural phenomena like typhoons, pest infestations, the occurence of infectious tree diseases, and even anthropogenic factors.

The pH of water in five inland aquatic systems significantly decreased from 2000 to 2009. The accumulated data for the last years suggested the possible acidification or nitrogen saturation of the inland ecosystems in several EANET sites. But just like in the cases of the forest soil and vegetation, some other factors that can bring about nitrogen saturation in inland waters need further amplification.

The availability of data covering more than a single measurement over a period of time has led to the establishment of trends in certain parameters that are being investigated in the EANET monitoring framework. With the kind of data that is already available at the end of 2009, attempts have been made to identify the possible relationships that exist among the different phenomena that describe the dynamics of nitrogen and sulphur loading in both the terrestrial and inland aquatic environments. As the monitoring progresses through the coming years, there is a growing need to carefully discuss the relationship among components of the ecosystems, including soil, vegetation, inland water, and atmosphere.

The promotion of catchment scale analysis is an approach to clarify such relationship. This analysis takes the form of an integrated monitoring including atmosphere, vegetation, soil, and inland water to evaluate the effects of atmospheric deposition on forest ecosystems qualitatively and quantitatively. The EANET has been implementing a number of case studies in the East Asian region covering three sites representing three different forest types. The experiences in such studies have led to the formulation of the Guideline for Catchment-scale Monitoring in East Asia by the Task Force on Soil and Vegetation Monitoring of EANET. The Guideline was endorsed by Scientific Advisory Committee at its Tenth Session (SAC10) in October 2010. With this development the monitoring initiatives of the EANET takes on a new dimension to better explain the phenomenon of acid deposition and its concomitant effects in the East Asian Region.

For other Related Studies for EANET Region, numbers of the peer reviewed journal papers were reported and published in the last few years to elucidate the causes and responses of regional acid deposition, air pollution and their environmental impacts in East Asia. They were based on observational and modeling studies. Most of those studies were reviewed in deeply viewpoints that related the EANET monitoring data. The results were also obtained from the EANET additional activities by collaborated scientists of the participating countries. These kinds of scientific information are crucial for EANET for passing messages of environmental implication of the monitoring data to policy makers and public.

Moreover, Ozone monitoring data of acid deposition and atmospheric concentration have also been widely used in the international science communities which meant that the data quality of EANET has been accepted to be high enough to be used for scientific research. Therefore, more active integration of emission inventory, model simulation and monitoring would be a key for the EANET in the future successful operation.

Finally, the last chapter on summary and recommendations for future activities, it is started reported from introduction of the 10 year anniversary celebration (2010) of the EANET and 5 years (2005-2009) of the monitoring data assessment. Quality assurance and quality control was continued and followed with the wet and dry deposition, ecological impacts on soil, forest vegetation and inland aquatic environment as well as catchment analysis which those impacts were affected the human health. Emission inventories and modeling also included in this report. Moreover, the risk assessments of acidification and eutrophication, O_3 and PM were viewed. Because O_3 causes the reductions of crop yield, tree growth and carbon sequestration and chemical species composition modification as well. Monitoring data of O_3 and $PM_{2.5}$ are necessary for estimates of their human and agricultural impacts more accurately.

1. Introduction

1.1 Background

Acid deposition is a general term that includes more than simply acid rain. Acid deposition is primarily the result of emissions of sulphur dioxide (SO_2) and nitrogen oxides (NOx) that can be transformed into dry or moist secondary pollutants such as sulphuric acid (H_2SO_4), ammonium nitrate (NH_4NO_3) and nitric acid (HNO_3) as they are transported in the atmosphere over distances of hundreds to thousands of kilometres.

Acidic particies and vapours are deposited in two processes – wet and dry deposition. Wet deposition is what is called acid rain, the process by which acids are removed from the atmosphere in rain, snow, sleet or hail. Dry deposition takes place when particles such as fly ash, sulphates, nitrates, and gases (such as SO_2 and NOx), are deposited on, or absorbed onto deposition surfaces. The gases can then be converted into acids when contact water. Damage caused by acid depostion affects lakes, rivers, forest, soils, fish and wildlife populations and building. Prior to failing to the earth, acid-causing gases and aerosols contribute to visibility degradation and impact public health.

The main sources of emissions contributing to acid rain are :
For sulphur dioxide (SO_2) :

- Base metal smelters;
- Fossil-fuelled power plants;
- Industrial fossil-fuel conbustion sources (natural gas plants, pulp and paper mills, petroleum refineries, cement plants, upstream oil and gas, etc.).

For nitrous oxide (NOx) :

- The transportation sector (automobiles, truck, off-road engines);
- Fossil-fuelled power plants;
- Industrial fossil-fuel combustion sources (natural gas plants, pulp and paper mills, petroleum refineries, cement plants, upstream oil and gas, etc).

The concept of long range transboundary pollution was born in the 1970s when scientists were able to link ecological damage to the depostion of acidifying pollutants that were transported long distances from their sources by prevailing winds. The solution to this complex problem required action locally, provincially, nationally and internationally.

Acid deposition is related to smog and visibility/regional haze through common emissions and precursors, production pathways, and meteorological processes. Emissions of sulphur dioxide (SO_2) and nitrogen oxides (NOx) from coal-and oil-fired power platns, smelters, motor vehicles , and other human-related sources are the principal contributors to acid depostion, ground- level ozone, and particulates matter. Fine particulate matter is the main cause of poor visibility and regional haze pollution. Reduction in SO_2 and NOx will result in improvements in acid depostion, atmospheric concentration, particulate matters (PM) and ozone.

There are also linkages between acid rain and climate change. The interactions between acid rain and climate change can magnify the impacts of acid rain. Climate change can affect acid levels in lakes, because hot, dry conditions convert harmless sulphur compounds that have accumulated in wetlands into acid-forming sulphates. When it rains, these sulphates are flushed into surrounding lakes, boosting their acid levels.

As a result of rapid industrialization in the East Asian region, it faces the increasing risks of problems related to excess deposition of acidic substances. Thus the adverse effects of acid deposition in East Asia could become a critical problem in the near future. With this background , expert meetings were held four times since 1993 to discuss the state of acid deposition in the region, effects on ecosystems and future moves toward regional cooperation on this issue. The experts recommended that acid deposition monitoring should be improved and strengthened. Thus, the expert meetings contributed the necessary knowledge for establishing a regional collaborative monitoring network. On these results, the experts developed a preliminary outline for the design of such a network as well as guidelines for monitoring methods.

Acid Deposition Monitoring Network in East Asia (EANET), therefore, was established as a regional cooperative initiative to promote efforts for environmental sustainability. The First Session of the Intergovernmental Meeting (IG1) on the EANET was held in March 1998 in Yokohama, Japan. Based on the agreement at the IG1, EANET was started the preparatory phase activities in April 1998. On an interim basis, there were useful inputs to provide to the Second Session of the Intergovernmental Meeting (IG2) which was held in 2000 in Niigata, Japan with participation of ten countries in East Asia to cover both the atmospheric and ecological environments. These countries were China, Indonesia, Japan, Malaysia, Mongolia, Philippines, Republic of Korea, Russia, Thailand, and Vietnam for the formal establishment of the Network.

The IG2 concluded that the preparatory phase activities of EANET had been successful and decided to start the EANET activities as a regular basis from January 2001 based on the " Joint Announcement (JA) on the Implementation of EANET" and the "Tentative Design (TD) of EANET". The Meeting also designated the United Nations Environment Programme (UNEP) as the Secretariat and the Acid Deposition and Oxidant Research Center (ADORC) in Japan as the Network Center for EANET respectively. The Third Session of the Intergovernmental Meeting (IG3) was held in November 2001 in Chiang Mai, Thailand and the “Rules of Procedure for EANET” were adopted. Since then, Sessions of the Intergovernmental Meetings have been taken place annually following the Rules of Procedures. In 2001, 2002, and 2005, Cambodia, Lao PDR, and Myanmar have joined the EANET respectively , which now covers the thirteen countries in East Asia.

The cooperative activities of EANET participating countries based on developed national monitoring plans include the implementation of monitoring of wet deposition, dry deposition, inland aquatic environment, and soil and vegetation in line with the guidelines and other technical documents with the conducting of quality assurance/quality control (QA/QC) programs as an important part of the monitoring activities. Collected data are compiled by the Network Center, so that periodic reports on the state of acid deposition in the East Asian region are produced. It is expected that the participating countries create a common understanding on the status of the acid deposition problems through EANET activities, which will become a scientific basis for taking further steps to tackle the problems.

In November 2010, the “Instrument for the Strengthening the Acid Deposition Monitoring Network in East Asia (EANET)” was adopted at the Twelfth Session of the Intergovernmental Meeting (IG12). At the IG12, it was decided that the Instrument will become operational on the date on which all participating countries have signed it, or on 1st January 2012, whichever is earlier;

1.1.1 Experiences from the past for the future

EANET has made progress and important achievements in monitoring, data acquisition and management, research, and other technical issues. This is the second edition of the *Periodic Report on the State of Acid Deposition in East Asia* that continued to present the result of second period (2005-2009) of a scientific evaluation of the information that obtained through the regular phase activities after the first period (2000-2004) to date and provides some conclusions. This EANET development covered the main goals and objectives of the network. The measurements collected for ten years are able to draw more precise and definite conclusions on temporal and spatial variations of atmospheric deposition. Even some data assessments up to date have identified weaknesses and some measures that should be taken to detect their potential impact on the environment. However, there is general agreement that improvements on monitoring and data quality are among the most important directions for EANET to focus on. Progress in these areas should provide a more solid basis for advanced assessment of acid deposition and related environmental problems. In this regard, the number of coordinated research projects and studies support both the elaboration of technical issues and the use of other approaches to investigate the state of the atmospheric environment in EANET region.

Many discussions were held among the scientists, experts, and representatives from the participating countries at the meetings of EANET bodies to approach for further development of the network as well as new directions and topics. The discussions highlighted the necessity of more comprehensive and wider application of data obtained using the experience of other international programs. They also emphasized the need to provide fact-finding basis for ensuring healthy conditions for humans and a sustainable environment in East Asia. Most achievements were obtained under the *Strategy on EANET development (2006-2010)* which were covered almost all network activities to be coordinated among the participating countries, the Secretariat and the Network Center. A new *Strategy on EANET development* which is called *Medium Term Plan for EANET (2011-2015)* was also prepared to cover more activities for strengthening cooperation for our participating countries.

The *First Periodic Report* recommended the scientists and experts on the research and studies to view the need for future projects all activities to take into account that not only the necessity to eliminate weaknesses in current monitoring and data evaluation, but also the challenges of prospective directions in environmental assessments and new issues of the network. The *Second Period Report* has been continued on this perspective. In recent years, there is a new trend to view regional air pollution problems from a global perspective. In particular, ozone and fine particulate matter such as $PM_{2.5}$ have become a noticeable issue of hemispheric air pollution, including inter-continental transport. The impacts on climate change and global warming caused by regional air pollution are also notable issue. The Network Center aims to accomplish the mission of researching acid deposition and air pollution problems in the East Asian region, including these new issues.

1.1.2 Preparation of the second periodic report

The Ninth Session of the Scientific Advisory Committee (SAC9) approved the establishment of the Drafting Committee for the Second Periodic Report on State of Acid Deposition in East Asia (PRSAD2) to prepare the implementation plan and other related works. Subsequently, Dr. Doung Hong Son (Vietnam) was designated as the Chairperson of the Drafting Committee as the Chairperson of the Scientific Advisory Committee (SAC). The Tenth Session of the Scientific Advisory Committee (SAC10) also reviewed the progress report of the Drafting Committee for the PRSAD2.

The DC developed the format and contents of PRSAD2. There were three parts: Part I: Regional Assessment, Part II: National Assessments and Part III: Executive Summary. However, this report is emphasized on Part I. The Drafting Committee also nominated the Lead Authors and Contributors to draft the report. The Drafting Committee and Lead Authors considered following manuscript as the previous format. Finally, the Drafting Committee and the Lead Authors decided the contents of PRSAD2 as follows:

- Chapter 1: Introduction
- Chapter 2: Data Quality
- Chapter 3: Wet deposition in East Asia

- Chapter 4: Dry and overall deposition in East Asia
- Chapter 5: Impacts on Ecosystems in East Asia
- Chapter 6: Other Related Studies for EANET Region
- Chapter 7: Summary and recommendations for future activities.

The report was prepared as a scientific assessment of the state of acid deposition in East Asia based on the data accumulated from the network from 2005 to 2009 (five years). The decided title is the "Second Periodic Report on the State of Acid Deposition in East Asia" (PRSAD2). The report also used some data in 2000-2004 for trend analysis and as appropriate.

1.2 Objectives

The objectives of EANET stated in the Joint Announcement of EANET (EANET/IG 2/5/2 rev.) adopted at the IG2 are described as follows:

1. To create a common understanding of the state of the acid deposition problems in East Asia;
2. To provide useful inputs for decision-making at local, national and regional levels aiming at preventing or reducing adverse impacts on the environment caused by acid deposition; and
3. To contribute to cooperation among the participating countries the issues related to acid deposition.

To achieve the above objectives, many activities have been conducted. Major activities of EANET are as follows:

a. Acid deposition monitoring in the participating countries using common methodologies. The monitoring covers four environmental fields: wet deposition, dry deposition, soil/vegetation and inland aquatic environment;
b. Compilation, evaluation, storage and provision of data obtained through the monitoring;
c. Promotion of QA/QC activities to obtain high quality monitoring data;
d. Capacity building of monitoring in participating countries including implementation of training programs;
e. Promotion of research, studies and public awareness related to acid deposition problems.

1.3 Institutional arrangement

The Intergovernmental Meeting (IG), Scientific Advisory Committee (SAC), Secretariat and Network Center (NC) are described in the "Tentative Design (TD) of the Acid Deposition Monitoring Network in East Asia (EANET)" (EANET/IG 2/5/3) as institutional bodies for implementing of the EANET activities on a regular basis.

The Intergovernmental Meeting, composed from the representatives of the participating countries, is the decision making body of EANET, and deals with the matters related to the management of the Network and implementation of the work program.

The Scientific Advisory Committee incorporated scientific and technical experts from the participating countries and should advise and assist the Intergovernmental Meeting with various scientific and technical matters related to the network activities. Four Task Forces of the Monitoring for Dry Deposition, Soil and Vegetation Monitoring, Monitoring Instrumentation, and Research Coordination were established under the SAC supervising to assist in the development of a strategy for vital directions of monitoring as well as four Expert Groups of the Dry Deposition Flux Estimation, Technical Manual on Wet Deposition Monitoring, Technical Manual on Air Concentration Monitoring, and Technical Manual on Inland Aquatic Environment Monitoring, (Figure 1.3.1).

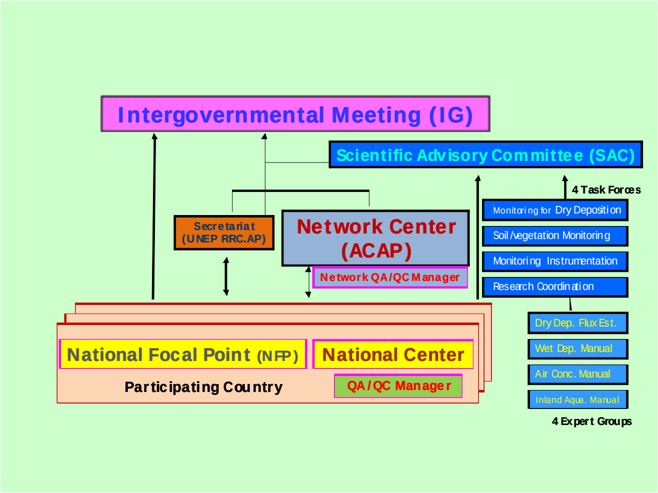

Figure 1.3.1. Institutional Framework for EANET.

UNEP Regional Resource Center for Asia and the Pacific (RRC.AP) was designated as the Secretariat for EANET where is located in Pathumthani, Thailand and Acid Deposition and Oxidant Research Center (ADORC) was designated as the Network Center for EANET that is located in Niigata, Japan. The ADORC was renamed to Asia Center for Air Pollution Research (ACAP) on 30 June 2010 in order to promote broader scope of air pollution research including ozone, aerosols, acidification, eutrophication, and climate change related issue and so on as a research institute.

ADORC (ACAP) was designated as the Network Center for EANET to carry out the following tasks under the guidance of the Intergovernmental Meeting:

1) Central compilation, evaluation and storage of monitoring data and related information;
2) Preparation of data reports on acid deposition in East Asia;
3) Dissemination of monitoring data and other relevant information;
4) Provision of technical assistance to the participating countries in implementing the network activities;
5) Implementation and coordination of QA/QC activities;
6) Development and implementation of education/training programs for those engaged in the network activities;
7) Implementation of research activities on acid deposition;
8) Provision of scientific and technical support for the Intergovernmental Meeting, Scientific Advisory Committee and other subsidiary bodies; and
9) Other tasks as requested by the Intergovernmental Meeting.

1.3.1 Progress of EANET on adoption of the "Instrument for Strengthening the EANET"

The Seventh Session of the Intergovernmental Meeting (IG7) held in 2005 adopted the Decision 1/IG7 (Niigata Decision) which decided that the participating countries of EANET should begin a process to discuss an appropriate Instrument and its legal status to provide a sound basis for financial contribution to EANET and reported the results of the discussion to the Tenth Session of the Intergovernmental Meeting (IG10) for its consideration.

Discussion on an Appropriate Instrument to Provide a Sound Basis for Contribution to EANET was implemented from May 2006. Many time discussions at the Sessions of the Working Group on Future Development of EANET (WGFD) and the Sessions of the Intergovernmental Meeting (IG) were carried out in order to adopt the Instrument. The main points of the discussion include the following:

- Legal status: Legally binding or non-legally binding, and with/without signature;
- Scope of the instrument: Regional air pollution or acid deposition, and air quality management or only monitoring;
- Title of the Instrument;
- Definition of "Acid Deposition", etc.;
- Objectives: If the Instrument includes "regional air pollution", "management", etc.; and
- Detailed stipulation of the "Financial arrangement" and so on.

As a result, the Twelfth Session of the Intergovernmental Meeting on EANET (IG12) held in November 2010 in Niigata, Japan adopted the Decision 1/IG12 on Instrument for the Strengthening the Acid Deposition Monitoring Network in East Asia (EANET) after 5 years discussion among the participating since the Niigata Decision. The following were stipulated in the Decision:

- Adopted the "Instrument for the Strengthening the Acid Deposition Monitoring Network in East Asia (EANET)" for signing in the High Level Segment during IG12 and thereafter;
- Decided that the Instrument will become operational on the date on which all participating countries have signed it, or on the 1st of January 2012, whichever is earlier;
- Encourages participating countries that will not be able to sign the Instrument by the specified date to communicate in writing with the EANET Secretariat before IG13 expressing their willingness to continue their EANET activities; and
- Decides that IG13 will consider the nature of involvement of the countries that are unable to sign by the specified date.

Item 1 of the Instrument, "Definition" was decided as follows:

"Acid deposition" in this Instrument means deposition of major acidifying species and related chemical substances.

It is common understanding that "related chemical species" in the Item 1 include ozone and particulate matter (PM).

Item 2 of the Instrument "Objectives and Scope" stipulated as follows:

The objectives and scope of this Instrument are:
- to create a common understanding of the state of acid deposition problems in East Asia;
- to provide useful inputs for decision-making at local, national and regional levels aimed at preventing or reducing adverse impacts on the environment caused by acid deposition; and
- to contribute and cooperation on the issues related to acid deposition among the Participating Countries.

The scope of this Instrument may be extended, as decided by the IG.

1.3.2 National Centers of EANET participating countries

Each participating country in EANET is expected to submit a national monitoring plan to the Network Center in the proposed data reporting formats and is may be requested to revise its plan where appropriate, after feedback from the Network Center based on the experience obtained in previous years and scientific advice from the Scientific Advisory Committee.

Each national monitoring plan should include the following information:

1.) Information on the National Center and the contact person;
2.) National monitoring plan
  - Responsible agency
  - Number of monitoring sites
  - Measurement parameters and their intervals
  - Participating laboratories for each monitoring activity;
3.) Information on respective monitoring sites
  - Outline of each monitoring site, including name, address, and geographical description
  - Sample collection
  - Meteorological observation
  - Situation around the site, including descriptions and sketch maps of the topography, land use, vegetation, source of air pollutants, etc.

In EANET, each National Center is responsible for preparation and implementation of the participating country's national monitoring plan (Table 1.3.1).

Table 1.3.1. National Focal Points and National Centers of EANET.

Participating countries	National Focal Points (Organizations)	National Centers
Cambodia	Department of Environmental Pollution Control, Ministry of Environment (MOE)	Department of Environmental Pollution Control, Ministry of Environment (MOE)
China	International Cooperation Department, Ministry of Environmental Protection (MEP)	China National Environmental Monitoring Centre (CNEMC)
Indonesia	Deputy Minister for Environmental Degradation Control and Climate Change, Ministry of Environment (MOE)	Environmental Impact Control Facility (SARPEDAL)
Japan	Environmental Management Bureau, Ministry of the Environment (MOE)	Asia Center for Air Pollution Research (ACAP)
Lao PDR	Water Resources and Environment Research Institute, Ministry of Natural Resources and Environment (MONRE)	Water Resources and Environment Research Institute, Ministry of Natural Resources and Environment (MONRE)
Malaysia	Environmental Conservation Department, Ministry of Natural Resources and Environment (NRE).	Malaysian Meteorological Department (MMD), Ministry of Science, Technology and Innovation (MOSTI)
Mongolia	National Agency for Meteorology and Environment Monitoring (NAMEM)	Central Laboratory of Environment and Metrology (CLEM)
Myanmar	Department of Meteorology and Hydrology (DMH), Ministry of Transport	Department of Meteorology and Hydrology (DMH), Ministry of Transport
Philippines	Environmental Management Bureau (EMB), Department of Environment and Natural Resources (DENR)	Environmental Management Bureau (EMB), Department of Environment and Natural Resources (DENR)
R. of Korea	Climate and Air Quality Management Office, Ministry of Environment (MOE)	National Institute of Environmental Research (NIER), Ministry of Environment (MOE).
Russia	Institute of Global Climate and Ecology, Roshydromet and RAS	- Institute of Global Climate and Ecology, Roshydromet and RAS, - Limnological Institute, Russian Academy of Sciences/Siberian Branch (RAS/SB)

Thailand	Pollution Control Department (PCD), Ministry of Natural Resources and Environment (MNRE)	Pollution Control Department (PCD), Ministry of Natural Resources and Environment (MNRE).
Vietnam	Institute of Meteorology, Hydrology and Environment (IMHEN), Ministry of Natural Resources and Environment (MONRE)	Institute of Meteorology, Hydrology and Environment (IMHEN), Ministry of Natural Resources and Environment (MONRE)

1.4. EANET Activities in 2005-2009

EANET activities are summarized as follows:

1) Each participating country developed and implements their national monitoring plans, which has reviewed and revised by taking into account the experience since the preparatory-phase. Acid deposition monitoring is implemented in accordance with the monitoring guidelines, technical manuals and other technical documents adopted by the Network. Each participating country prepares and reports its monitoring data and other relevant information in accordance with specified procedures. Each participating country designates a national center, which is responsible for implementation of the monitoring activities for the Network in the country and for reporting monitoring data.
2) The monitoring data and other information submitted by participating countries is compiled, evaluated and stored by the Network Center. Any participating country is able to obtain the data and information submitted to the Network Center by other participants (Figure 1.4.1).
3) In order to obtain monitoring data of high quality, the quality assurance/quality control (QA/QC) programs are implemented in full collaboration among the participating countries.
4) Periodic reports on the state of acid deposition in the East Asian region are produced and made available to the participating countries and scientific reviews.
5) The data and information compiled through the network activities may be provided to individuals, organizations and non-member countries, in accordance with specified procedures.
6) The participating countries will promote studies of related scientific issues in order to improve understanding of the risks of acid deposition.
7) Other activities necessary to achieve the objectives of the Network can be implemented.

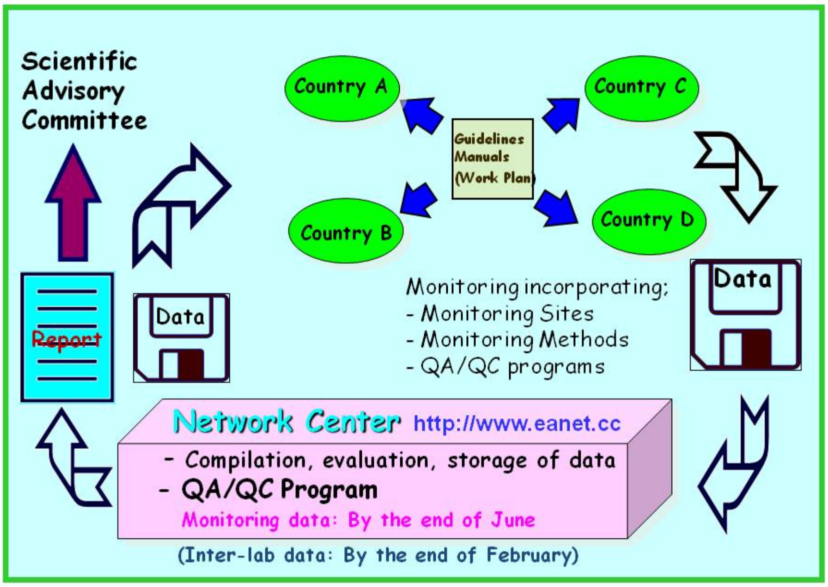

Figure 1.4.1. Principal scheme of data exchange in EANET.

1.4.1 Strategy on EANET Development (2006-2010)

The Strategy on EANET Development (2006-2010) which was approved by the Eighth Session of the Intergovernmental Meeting (IG8) in 2006 focused on the whole activities of EANET with clearly stated targets, activities to be undertaken and expected results at the end of 2010. The implementation of the Strategy activities since 2006, and the regular activities of the Secretariat and the Network Center since the start of EANET, has brought EANET closer towards achieving its objectives. It also includes the activities relevant to the modeling and emission inventories.

The Strategy on EANET Development (2006-2010) was fulfilled and recognized the significant achievements during the implementation. The Secretariat and the Network Center made their best efforts to carry out and implement all the activities within the manpower and funding resources available, according to the priorities as decided by the IG.

The implementation of the Strategy activities since 2006 and the regular activities has brought EANET closer towards achieving its objectives. Major results and outputs of the Strategy are as follows:

- Report on activities to improve data completeness;
- Set of Standard Operating Procedures (SOPs);
- New monitoring sites in EANET;
- Annual Data Reports;
- Updated Technical Manuals, Strategy Papers and Guidelines for EANET monitoring;
- First and Second Periodic Reports on the State of Acid Deposition in East Asia;
- Research Projects;
- Fellowship Research
- EANET Science Bulletin;
- Second Report for Policy Makers (RPM2);
- Bi-annual issues/publications of the EANET Newsletter;
- Report of the Working Group on Future Development of EANET (WGFD) on the Instrument to Provide a Sound Basis for Contribution to EANET;
- Capacity building workshops for policy makers, technical trainings and national workshops on public awareness in the participating countries;
- CDs, videos and brochures on acid deposition and other public awareness materials; and
- Development of the Medium Term Plan for EANET (2011-2015), etc.

1.4.2 Monitoring activities

The EANET monitoring sites consist of wet deposition, dry deposition, soil and vegetation and inland aquatic environment, and the monitoring was carried out in accordance with the monitoring guidelines, technical manuals and other technical documents, which were approved since the Second Interim Scientific Advisory Group Meeting (ISAG2). Participating countries are noted to be urged to endeavor to improve their activities in acid deposition monitoring by making effective use of these documents with adjustment, if appropriate, taking into account specific conditions or circumstances of them

EANET monitoring sites were classified into two categories under their purposes: Acid Deposition Monitoring Site and Ecological Survey Site following the criteria presented in Table 1.4.1.

Table 1.4.1. Classification of Monitoring Sites.

Site Category	Site Classification	Main Purposes and Siting Criteria
Monitoring Site for wet and dry deposition	Urban Site	- Assessment of the state of acid deposition in urban areas - Urbanized and industrial areas, and the areas immediately outside the urban area - Data can be used for evaluation of acid deposition effects on buildings and historical monuments or human health
	Rural Site	- Assessment of the state of acid deposition in rural areas and/or hinterlands - Data can be used for the evaluation of acid deposition on agricultural crops, forests and other environmental elements. - More than 20 km apart from large pollution sources like cities, power plants and highways - More than 500 m apart from main roads (more than 500 vehicles per day).
	Remote Site	- Assessment of the state of acid deposition in background areas - Data can be used for evaluation of long-range transport and deposition models - More than 50 km apart from large pollution sources like cities, power plants or highways - More than 500 m apart from main roads (less than 500 vehicles per day)
Ecological Survey Site for soil and vegetation and inland aquatic environment	Basic Survey Site	- Accumulation of basic data on soil, forest, and inland aquatic environment and disclose trends in their properties - In the vicinity of the acid deposition monitoring site
	Ecosystem Analysis Site	- Assessment of acid deposition impacts on the whole ecosystem through application of terrestrial ecosystem analysis and/or catchment analysis - Sensitive Areas to changes in atmospheric acidity and ecologically conserved area

In 2009, wet deposition monitoring was conducted at 54 sites (remote: 20, rural: 13, urban: 21) in thirteen participating countries (Figure 1.4.2, Table 1.4.2).

Instrumental monitoring methods and the filter pack method are mainly used to implement the dry deposition (air concentration) monitoring. Automatic or manual gas monitor for NO_x and SO_2 are operated at 24 sites, and 23 sites are equipped with automatic instrument including monitor of ozone concentration. Concentration of particulate matters (PM) as PM_{10} was measured by automatic or manual instruments at 21 sites. Table 1.4.3 provides a list of the monitoring sites and sampling methods for dry deposition at each monitoring site.

Ecological survey sites established for soil and vegetation monitoring and inland aquatic environment monitoring are basically located in the vicinity of the acid deposition monitoring sites. According to the National Monitoring Plans of each participating country, nineteen areas have been selected as the soil and vegetation monitoring sites. Basically, two forest areas, whose soils have different sensitivities to acid deposition, are recommended to be selected in the area. Summary of the individual monitoring sites is listed.

Thirteen EANET countries, namely, China, Japan, Mongolia, Republic of Korea and Russian Federation of the North-East Asian region, and Cambodia, Indonesia, Malaysia, Myanmar, Lao PDR, Philippines, Thailand and Viet nam participated in monitoring of acid deposition in 2005-2009. Monitoring data were submitted from a total of 54 monitoring sites, including 20 remote, 13 rural and 21 urban sites in 2009. A map showing the location of these sites is provided in Figures 1.4.2 and 1.4.3. The details on the locations and outlines of the monitoring sites are presented in Tables 1.4.2 and 1.4.3.

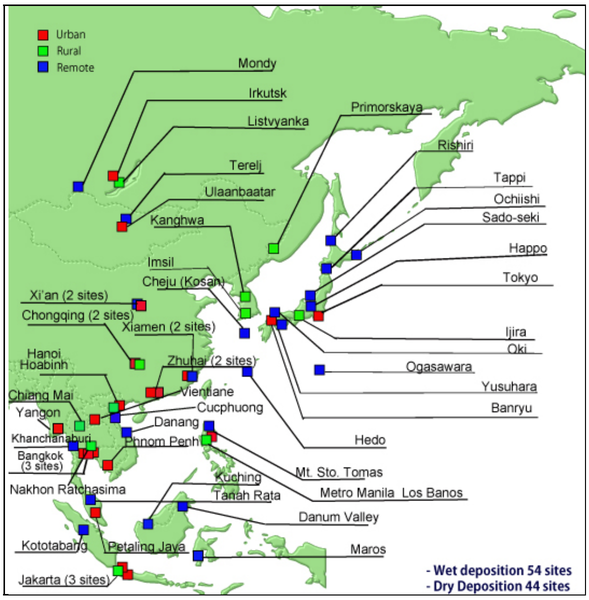

Figure 1.4.2. EANET deposition monitoring sites (2009)

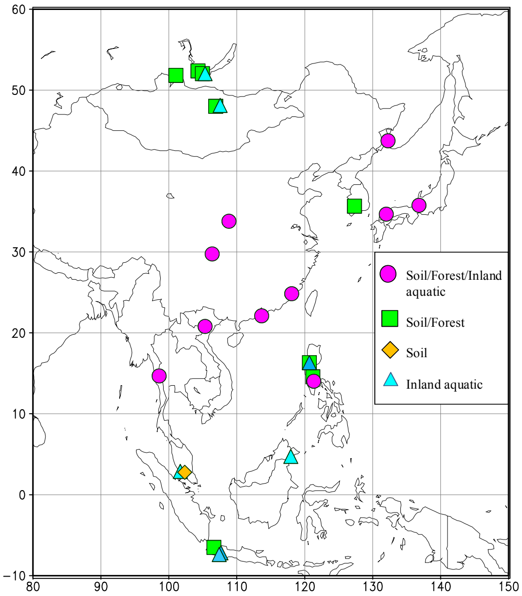

Figure 1.4.3. EANET ecological impact monitoring sites (2009)

- *Soil: 19 areas (27 forests)*
- *Forest vegetation: 18 areas (26 forests)*
- *Inland aquatic environments: 17 lakes/rivers.*

Table 1.4.2. Locations of EANET Deposition Monitoring Sites.

Country	Name of sites	Characteristics of sites	Latitude	Longitude	Ht. above sea level
Cambodia	Phnom Penh	Urban	11°33'N	104°50'E	10 m
China	Chongqing-Haifu	Urban	29°37'N	106°30'E	317 m
	- Jinyunshan	Rural	29°49'N	106°22'E	800 m
	Xi'an - Shizhan	Urban	34°14'N	108°57'E	400 m
	- Jiwozi	Remote	33°50'N	108°48'E	1,800 m
	Xiamen - Hongwen	Urban	24°28' N	118°08' E	50 m
	- Xiaoping	Remote	24°51' N	118°02' E	686 m
	Zhuhai - Xiang Zhou	Urban	22°16'N	113°34'E	40 m
	- Zhuxiandong	Urban	22°12'N	113°31'E	45 m

Table 1.4.2. *(continued)*

Country	Name of sites	Characteristics of sites	Latitude	Longitude	Ht. above sea level
Indonesia	Jakarta	Urban	6°11’ S	106°50’ E	7 m
	Serpong	Rural	6°15’ S	106°34’ E	46 m
	Kototabang	Remote	0°12’ S	100°19’ E	864 m
	Bandung	Urban	6°54’ S	107°35’ E	743 m
	Maros	Rural	4°55’ S	119°34’ E	11 m
Japan	Rishiri	Remote	45°07’11” N	141°12’33” E	40 m
	Ochiishi	Remote	43°09’43” N	145°29’50” E	49 m
	Tappi	Remote	41°15’06” N	140°20’59” E	106 m
	Sado-seki	Remote	38°14’59” N	138°24’00” E	136 m
	Happo	Remote	36°41’48” N	137°47’53” E	1,850 m
	Ijira	Rural	35°34’20” N	136°41’38” E	140 m
	Oki	Remote	36°17’19” N	133°11’06” E	90 m
	Banryu	Urban	34°40’54” N	131°47’59” E	53 m
	Yusuhara	Remote	33°22’45” N	132°56’05” E	790 m
	Hedo	Remote	26°51’58” N	128°14’55” E	60 m
	Ogasawara	Remote	27°05’30” N	142°12’58” E	230 m
	Tokyo	Urban	35°41’18” N	139°45’22” E	26 m
Lao PDR	Vientiane	Urban	17° N	102° E	
Malaysia	Tanah Rata	Remote	04°28’ N	101°23’ E	1,470 m
	Petaling Jaya	Urban	03°06’ N	101°39’ E	87 m
	Danum Valley	Remote	04°59’ N	117°51’ E	427 m
	Kuching	Urban	01°29’ N	110°28’ E	22 m
Mongolia	Ulaanbaatar	Urban	47°54’ N	106°49’ E	1,282 m
	Terelj	Remote	47°59’ N	107°29’ E	1,540 m
Myanmar	Yangon	Urban	16°30’ N	96°07’ E	22 m
Philippines	Metro Manila	Urban	14°38’ N	121°04’ E	54 m
	Los Banos	Rural	14°11’ N	121°15’ E	35 m
	Mt. Sto. Tomas	Remote	16°25’ N	120°36’ E	1,500 m
Republic of Korea	Ganghwa	Rural	37°42’ N	126°17’ E	150 m
	Jeju (Kosan)	Remote	33°18’ N	126°10’ E	72 m
	Imsil	Rural	35°36’ N	127°11’ E	
Russia	Mondy	Remote	51° 40’ N	101°00’ E	2,000 m
	Listvyanka	Rural	51° 51’ N	104°54’ E	700 m
	Irkutsk	Urban	52° 14’ N	104°15’ E	400 m
	Primorskaya	Rural	43° 42’ N	132°07’ E	84 m
Thailand	Bangkok	Urban	13° 46’ N	100°32’ E	2 m
	Samutprakan	Urban	13° 44’ N	100°34’ E	2 m
	Pathumthani	Rural	14° 02’ N	100°46’ E	2 m
	Kanchanaburi (Vachiralongkorn Dam)	Remote	14° 46’ N	98°35’ E	170 m
	Chiang Mai (Mae Hia)	Rural	18° 46’ N	98°56’ E	350 m
	Nakhon Ratchasima	Rural	14° 27’ N	101°53’ E	418 m
Vietnam	Hanoi	Urban	21° 01’ N	105°51’ E	5 m
	Hoa Binh	Rural	20° 49’ N	105°20’ E	23 m
	Da Nang	Urban	16°02’34” N	108°12’23” E	60 m
	Cuc Phuong	Remote	20°15’ N	105°43’ E	155 m

Table 1.4.3. Outlines of Deposition Monitoring.

Country	Name of sites	Characteristics of sites	Wet	Dry			
				Automatic			Filter Pack/ PS
				SO_2, NO_x	O_3	PM	
Cambodia	Phnom Penh	Urban	✓	None	None	None	None
China	Chongqing-Haifu	Urban	✓	None	None	None	None
	- Jinyunshan	Rural	✓	✓	None	✓	None
	Xi'an - Shizhan	Urban	✓	None	None	None	None
	- Jiwozi	Remote	✓	None	None	None	None
	Xiamen - Hongwen	Urban	✓	✓	None	✓	✓
	- Xiaoping	Remote	✓	None	None	None	None
	Zhuhai - Xiang Zhou	Urban	✓	✓	None	✓	None
	- Zhuxiandong	Urban	✓	None	None	None	None
Indonesia	Jakarta	Urban	✓	None	None	None	PS
	Serpong	Rural	✓	None	None	None	✓
	Kototabang	Remote	✓	None	None	None	PS
	Bandung	Urban	✓	None	None	None	PS
	Maros	Rural	✓	None	None	None	None
Japan	Rishiri	Remote	✓	✓	✓	✓	✓
	Ochiishi	Remote	✓	✓	✓	✓	✓
	Tappi	Remote	✓	✓	✓	✓	✓
	Sado-seki	Remote	✓	✓	✓	✓	✓
	Happo	Remote	✓	✓	✓	✓	✓
	Ijira	Rural	✓	✓	✓	✓	✓
	Oki	Remote	✓	✓	✓	✓	✓
	Banryu	Urban	✓	✓	✓	✓	✓
	Yusuhara	Remote	✓	✓	✓	✓	✓
	Hedo	Remote	✓	✓	✓	✓	✓
	Ogasawara	Remote	✓	✓	✓	✓	✓
	Tokyo	Urban	✓	None	None	None	✓
Lao PDR	Vientiane	Urban	✓	None	None	None	None
Malaysia	Tanah Rata	Remote	✓	✓	✓	None	✓
	Petaling Jaya	Urban	✓	None	None	None	✓
	Danum Valley	Remote	✓	None	None	None	✓
	Kuching	Urban	✓	None	None	None	None
Mongolia	Ulaanbaatar	Urban	✓	None	None	None	✓
	Terelj	Remote	✓	None	None	None	✓
Myanmar	Yangon	Urban	✓	None	None	None	None
Philippines	Metro Manila	Urban	✓	None	None	None	✓
	Los Banos	Rural	✓	None	None	None	✓
	Mt. Sto. Tomas	Remote	✓	None	None	None	✓
Republic of Korea	Ganghwa	Rural	✓	✓	✓	✓	✓
	Jeju (Kosan)	Remote	✓	✓	✓	✓	✓
	Imsil	Rural	✓	✓	✓	✓	✓
Russia	Mondy	Remote	✓	None	✓	None	✓
	Listvyanka	Rural	✓	None	None	None	✓
	Irkutsk	Urban	✓	None	None	None	✓
	Primorskaya	Rural	✓	None	None	None	✓

Thailand	Bangkok	Urban	✓	✓	✓	✓	✓
	Samutprakan	Urban	✓	✓	✓	✓	None
	Pathumthani	Rural	✓	None	None	None	✓
	Kanchanaburi (Vachiralongkorn Dam)	Remote	✓	✓	✓	✓	✓
	Chiang Mai (Mae Hia)	Rural	✓	✓	✓	✓	✓
	Nakhon Ratchasima	Rural	✓	None	None	None	✓
Vietnam	Hanoi	Urban	✓	None	None	None	✓
	Hoa Binh	Rural	✓	None	None	None	✓
	Da Nang	Urban	✓	None	None	None	None
	Cuc Phuong	Remote	✓	None	None	None	None

PS: Passive sampler

1.4.3 Wet deposition

Each participating country is required to carry out acid deposition monitoring using common methodologies as specified in the *Guidelines for Acid Deposition Monitoring in East Asia*, *Technical Documents on Wet Deposition Monitoring in East Asia* and related QA/QC documents. The wet-only sampler is recommended to collect the precipitation samples. This instrument is designed to collect precipitation samples during the period of rainfall which is detected by equipped rain sensor by opening the lid on the collecting bucket or funnel. The lid of this instrument closed to avoid any collection into the collecting bucket or funnel, when the rainfall is not detected by the sensor. On the other hand, when a manual sampler is used to collect the precipitation since an automatic instrument is not available, the sampling shall be done with ensuring the sample collection only during rainfall period.

The collected wet deposition samples in the tropical region have to be prevented from any microbiological degradation. Biocides such as thymol should be used to prevent the microbiological degradation when the storage during sampling, shipping and storage of the samples under the refrigeration is not possible to be used. Samples without biocides have to be shipped to the laboratories in charge of chemical analysis using a cooling box to keep the sample temperature low enough to preserve the sample chemistry. On the other hand, the special care for the sampling of wet deposition in the higher latitude region during winter seasons should be done.

1.4.4 Dry deposition

Automatic monitoring methods and filter pack method were mainly used to implement the dry deposition (air concentration) monitoring. The monitoring items were selected, taking into account priority of the chemical species defined in the "Strategy Paper for Future Direction of Dry Deposition Monitoring of EANET (Second Edition, 2005)".

1.4.4.1 Automatic monitoring method

In the "Strategy Paper for Future Direction of Dry Deposition Monitoring of EANET (Second Edition) (EANET/SAC 5/10/1 Appendix 1)" endorsed at the Fifth Session of the Scientific Advisory Committee (SAC5) in October 2005, it was mentioned that the first priority chemical species for dry deposition monitoring, namely SO_2, NO, NO_2 (urban), O_3, and PM could be monitored by use of automatic monitoring methods. The quality assurance and quality control (QA/QC) program for monitoring of SO_2, NO, NO_2 (urban), O_3, and PM using automatic monitors is described in the "QA/QC Program for the Air Concentration Monitoring in East Asia (2001)".

1.4.4.2 Filter pack method

The filter pack method could be used to determine gaseous substances (SO_2, HNO_3, HCl, NH_3) and particulate matter components (SO_4^{2-}, NO_3^-, Cl^-, NH_4^+, Na^+, K^+, Mg^{2+}, Ca^{2+}).

The “Technical Document of Filter Pack Monitoring in East Asia (EANET/SAC 3/8/1)” was endorsed at the Third Session of the Science Advisory Committee (SAC3) held in November 2003. The recommended monitoring method by means of the four-stage filter pack was explained in the technical document.

The filter pack methods were used by China, Indonesia, Japan, Malaysia, Mongolia, Philippines, Republic of Korea, Russia, Thailand and Vietnam in 2009.

The measurement methods and parameters in each monitoring site for dry deposition (air concentration) monitoring in 2009 are shown in Table 1.4.4. Automatic monitors used by China, Japan, Republic of Korea, Russia and Thailand are shown in Table 1.4.5.

Table 1.4.4. Sampling Method and Parameter for Dry Deposition (Air Concentration) Monitoring in 2009.

Country	Name of sites	Characteristics of sites	Method	Parameter
China	Chongqing - Jinyunshan	Rural	AT	SO_2, NO, NO_x*, PM_{10}
	Xiamen - Hongwen	Urban	AT FP	SO_2, NO_2, PM_{10} HNO_3, HCl, NH_3, PMC
	Zhuhai - Xiang Zhou	Urban	AT	SO_2, NO_2, PM_{10}
Indonesia	Jakarta	Urban	PS	SO_2, NO_2
	Serpong	Rural	FP	SO_2, HNO_3, HCl, NH_3, PMC
	Kototabang	Remote	PS	SO_2, NO_2
	Bandung	Urban	PS	SO_2, NO_2
Japan	Rishiri	Remote	AT FP	SO_2, NO, NO_x*, O_3, $PM_{10}/_{2.5}$ HNO_3, HCl, NH_3, PMC
	Ochiichi	Remote	AT FP	SO_2, NO, NO_x*, O_3, $PM_{10}/_{2.5}$ HNO_3, HCl, NH_3, PMC
	Tappi	Remote	AT FP	SO_2, NO, NO_x*, O_3, PM_{10} HNO_3, HCl, NH_3, PMC
	Sado-seki	Remote	AT FP	SO_2, NO, NO_x*, O_3, PM_{10} HNO_3, HCl, NH_3, PMC
	Happo	Remote	AT FP	SO_2, NO, NO_x*, O_3, PM_{10} HNO_3, HCl, NH_3, PMC
	Ijira	Rural	AT FP	SO_2, NO, NO_x*, O_3, PM_{10} HNO_3, HCl, NH_3, PMC
	Oki	Remote	AT FP	SO_2, NO, NO_x*, O_3, $PM_{10/2.5}$ HNO_3, HCl, NH_3, PMC
	Banryu	Urban	AT FP	SO_2, NO, NO_2, NO_x, O_3, PM_{10} HNO_3, HCl, NH_3, PMC
	Yusuhara	Remote	AT FP	SO_2, NO, NO_x*, O_3, PM_{10} HNO_3, HCl, NH_3, PMC

	Hedo	Remote	AT FP	SO_2, NO, NO_x*, O_3, PM_{10} HNO_3, HCl, NH_3, PMC
	Ogasawara	Remote	AT FP	SO_2, NO, NO_x*, O_3, PM_{10} HNO_3, HCl, NH_3, PMC
	Tokyo	Urban	FP	SO_2, HNO_3, HCl, NH_3, PMC
Malaysia	Tanah Rata	Remote	FP	SO_2, HNO_3, HCl, NH_3, PMC
	Petaling Jaya	Urban	FP	SO_2, HNO_3, HCl, NH_3, PMC
	Danum Valley	Remote	FP	SO_2, HNO_3, HCl, NH_3, PMC
Mongolia	Ulaanbaatar	Urban	FP	SO_2, HNO_3, HCl, NH_3, PMC
	Terelj	Remote	FP	SO_2, HNO_3, HCl, NH_3, PMC
Philippines	Mt. Sto. Tomas	Remote	FP	SO_2, HNO_3, HCl, NH_3, PMC
Republic of Korea	Ganghwa	Rural	AT FP	SO_2, NO_2, O_3, PM_{10} HNO_3, HCl, NH_3, PMC in $PM_{2.5}$
	Jeju (Kosan)	Remote	AT FP	SO_2, NO_2, O_3, PM_{10} HNO_3, HCl, NH_3, PMC in $PM_{2.5}$
	Imsil	Rural	AT FP	SO_2, NO_2, O_3, PM_{10} HNO_3, HCl, NH_3, PMC in $PM_{2.5}$
Russia	Mondy	Remote	AT FP	O_3 SO_2, HNO_3, HCl, NH_3, PMC
	Listvyanka	Rural	FP	SO_2, HNO_3, HCl, NH_3, PMC
	Irkutsk	Urban	FP	SO_2, HNO_3, HCl, NH_3, PMC
	Primorskaya	Rural	FP	SO_2, HNO_3, HCl, NH_3, PMC
Thailand	Bangkok	Urban	AT FP	SO_2, NO, NO_2, NO_x, O_3, PM_{10} HNO_3, HCl, NH_3, PMC
	Samutprakan	Urban	AT	SO_2, NO, NO_2, NO_x, O_3, PM_{10}
	Pathumthani	Rural	FP	SO_2, HNO_3, HCl, NH_3, PMC
	Kanchanaburi (Vachiralongkorn Dam)	Remote	AT FP	SO_2, NO, NO_x*, O_3, PM_{10} HNO_3, HCl, NH_3, PMC
	Chiang Mai (Mae Hia)	Rural	AT FP	SO_2, NO, NO_x*, O_3, PM_{10} HNO_3, HCl, NH_3, PMC
	Nakhon Ratchasima	Remote	FP	SO_2, HNO_3, HCl, NH_3, PMC
Vietnam	Hanoi	Urban	FP	SO_2, HNO_3, HCl, NH_3, PMC
	Hoa Binh	Rural	FP	SO_2, HNO_3, HCl, NH_3, PMC

(Note 1) AT: Automatic monitor, PS: Passive sampler, FP: Filter pack, PMC: Particulate matter components.

(Note 2) NOx: NOx measured by Chemiluminescence detection method (CLD) in remote or rural sites.*

(Note 3) Purposely monitor for 2 weeks and 4 times per year at Kanchanaburi (Vachiralongkorn Dam) due to the site located in remote area.

Table 1.4.5. Methods of automatic monitors in 2009.

Country	Sites	Parameter			
		SO_2	NO_x	O_3	PM_{10} ($PM_{2.5}$)
China	Jinyunshan	UVF	CLD	–	β–ray
	Hongwen	DOAS	DOAS	–	TEOM
	Xianzhou	DOAS	DOAS	–	TEOM
Japan	Rishiri	UVF	CLD	UVP	β–ray (TEOM)
	Ochiishi	UVF	CLD	UVP	β–ray (β–ray)
	Tappi	UVF	CLD	UVP	TEOM
	Sado-seki	UVF	CLD	UVP	TEOM
	Happo	UVF	CLD	UVP	β–ray
	Ijira	UVF	CLD	UVP	TEOM
	Oki	UVF	CLD	UVP	TEOM (TEOM)
	Banryu	UVF	CLD	UVP	TEOM
	Yusuhara	UVF	CLD	UVP	β–ray
	Hedo	UVF	CLD	UVP	β–ray
	Ogasawara	UVF	CLD	UVP	β–ray
Republic of Korea	Ganghwa	UVF	CLD	UVP	β–ray
	Jeju (Kosan)	UVF	CLD	UVP	β–ray
	Imsil	UVF	CLD	UVP	β–ray
Russia	Mondy	–	–	UVP	–
Thailand	Bangkok	UVF	CLD	UVP	TEOM
	Samutprakan	UVF	CLD	UVP	–
	Kanchanaburi	UVF	CLD	UVP	β–ray
	Chiang Mai	UVF	CLD	UVP	TEOM

UVF: Ultraviolet fluorescent method, CLD: Chemiluminescence detection method, UVP: Ultraviolet photometric method, β-ray: β-ray absorption method, TEOM: Tapered element oscillating microbalance method, DOAS: Differential optical absorption spectroscopy method.

1.4.5 Soil and vegetation

Soil and vegetation data principally needed to carry out to detect the possible impact. The Technical Manual on Soil and Vegetation Monitoring in East Asia is prepared for the participating countries as described in the Table 1.4.6, however , actual implementation of item sets is depended on the respective site.

Table 1.4.6. Basic survey for soil and forest.

Item	Parameters
Soil	- pH (H_2O), pH (KCl), Exchangeable base cations (Na, K, Ca, and Mg), Exchangeable acidity, Effective cation exchange capacity (ECEC), Carbonate content (if pH > 7) - Exchangeable Al and H, Total C content, Total N content (optional) - Available P, Sulfate (voluntary) - Physical properties (Fine earth bulk density, and Penetration resistance) (optional)
Forest	- General description of the forest (Description of trees, and Understory vegetation) - Observation of tree decline - Photographic record of tree decline, Estimation of decline causes (optional)

Analytical methods recommended in the Manual are presented in Table 1.4.7.

Table 1.4.7. Analytical equipment and methods for soil monitoring.

Parameters	Equipment/methods
1. Chemical Properties of Soil	
a) Moisture Content b) pH (H_2O) and pH (KCl) c) Exchangeable Base Cations (Ca, Mg, K and Na) d) Exchangeable Acidity e) Exchangeable Al , H f) Effective Cation Exchange Capacity (ECEC) g) Carbonate Content (for calcareous soil) h) Total Carbon Content i)Total Nitrogen Content j) Available Phosphate k) Sulfate	Drying oven, Balance Glass electrode AAS, ICP-AES or ICP-MS (CH_3COONH_4-Extraction) Titration (KCl-Extraction) ibid. Calculation (as sum of exchangeable cations) Volumetric calcimeter Titration (Walkley-Black method) or CN-analyzer Titration (Kjeldahl method) or CN- analyzer Spectrophotometry (Bray-1 test) Turbidimetry, IC, ICP-AES or ICP-MS
2. Physical Properties of Soil	
a) Fine Earth Bulk Density b) Penetration Resistance (in the fieldwork)	Metal sampling cylinder, Drying oven, Balance Pocket penetrometer

Soil and forest data in 2009 were submitted and shown in Table 1. 4.8. Observation of the tree decline was also done annually in the most plots according to suggestion of the Sub-Manual on Forest Vegetation Monitoring (2006).

Table 1.4.8. Outline of the Monitoring Sites in 2009.

Country	Nearest deposition monitoring site	Site: Name of forests	Soil type	Items[*1]
China	Jinyunshan (Chongquing)	Jinyunshan (2 plots for soil and forest)	(Acidi-Udic Argosols) [*2]	S, F
	Jiwozi (Xi'an)	Jiwozi (2 plots for soil; 1 plot for forest)	(Brown soil) [*2]	S, F
	Xiaoping (Xiamen)	Xiaoping (1 plot for soil and forest)	(Red soil)[*2]	S, F
	Zhuxiandong (Zhuhai)	Zhuxiandong (3 plots for soil and forest)	(Ochinic Udic Ferrosols) [*2]	S, F
Indonesia	Serpong	Research Forest Station of Center and Development of Forestry Research	Typic Dystrudepts	S
Japan	Banryu	Banryu-2	Cambisols	F[*3]
		Iwami "rinku" Factory Park	Acrisols	F[*3]
	Ijira	Lake Ijira	Dystric Cambisol	F[*3]
		Yamato	Andosols	F[*3]
Philippines	Mt. Sto. Tomas	Boneco Long Term Ecological Research Site		S[*4], F[*5]
Russia	Irkutsk	Irkutsk	Eutric Regosols/ Calcaric Luvisols	F[*4]
Thailand	Vachiralongkorn	Dam	Ferric Acrisols	F[*6]
		Puye	Luvisols	F[*6]

[*1] *S, Soil monitoring; F, Forest monitoring;*
[*2] *Soil type has not been classified by the FAO/UNESCO or relevant taxonomies;*
[*3] *Observation of tree decline was only conducted;*

*[*4] The data in 2008 is shown, which was not included in the Data Report 2008 because of the editorial mistake;*
*[*5] Correction table for the Data Report 2008 was shown;*
*[*6] Observation of tree decline and understory vegetation was not conducted.*

1.4.6 Inland aquatic environment

In general, inland bodies of water with low alkalinity and low electric conductivities are prone to be sensitive to acidification by acid deposition. Therefore, it is important to conduct continuous monitoring of water bodies and aquatic fauna and so on.

The participating countries of EANET are expected to carry out the monitoring of water temperature, pH, electric conductivity (EC), alkalinity and concentration of SO_4^{2-}, NO_3^-, Cl^-, NH_4^+, Na^+, K^+, Ca^{2+} and Mg^{2+} of targeted lakes/rivers at least four times a year (seasonally), and, transparency, water color, dissolved organic carbon (DOC) (if impossible, Chemical Oxygen Demand (COD)), NO_2^-, PO_4^{3-} more than once a year while some other items are specified as optional parameters to be monitored.

According to the Manual for Monitoring Inland Aquatic Environment, the lakes chosen for monitoring should be harmonic type lakes, preferably with depths of approximately 10 m or less, a water residence time of 1 year or less, an area of 1 ha or more, with low alkalinity and electric conductivity, minimal anthropogenic water pollution and no coverage of the surface with aquatic organisms. For the monitoring sites, it is desirable to locate in nature protection areas, and a minimum of human activities such as deforestation, and cultivation should be conducted or planned in the upstream area. Collected samples are analyzed by analytical methods specified in Table 1.4.9 immediately; otherwise they are stored in a refrigerator.

Table 1.4.9. Parameters and recommended analytical methods.

Parameter	Analytical method
pH	pH meter (glass electrode)
Electric Conductivity	Conductivity meter
Alkalinity	Titration by burette or digital burette with pH meter
NH_4^+, NO_3^-, NO_2^-, PO_4^{3-}	Ion Chromatography or spectrometry
K^+, Mg^{2+}, Ca^{2+}, Na^+	Ion Chromatography or atomic absorption spectrometry
SO_4^{2-}	Ion Chromatography or turbidimetry
DOC	Combustion- infrared method or wet-oxidation method

1.4.7 Research activities

Research is to improve air concentration monitoring methodologies, deposition estimations, building capacity in development of emission inventories and will also promote efforts to develop and use appropriate models to assess and analyze the trend of acid deposition and other relevant air pollutants at all scales.

Promotion of research activities related to acid deposition problems as follows:

- Promotion of research studies particularly on the applicability of various methodologies for measurement of air concentrations in East Asia;
- Promotion of studies on the effects of acid deposition and other priority chemical species on the ecosystem, human health, and other social aspects from the viewpoint of the socio-economics;
- Promotion of studies on proposed models to assess and analyze the trend of national and regional acid deposition and other air pollutants in East Asia by evaluation of existing

models and providing a suitable one, and promotion of atmospheric simulation model through workshops, training courses, etc.;
- Promotion of emission inventories through workshops, training course, pilot studies, preparation of reference materials, etc.

Joint research projects are implemented as well. A number of joint scientific research projects on acid deposition and its effects were conducted by EANET participating countries. Joint projects that were conducted are:

- Joint study with Japan, Malaysia and Thailand on catchment analysis;
- Joint study with Mongolia on plant sensitivity;
- Joint study with Thailand on dry deposition flux and gas concentration monitoring;
- Joint study with Republic of Korea on dry deposition (aerosol concentration) monitoring methodology;
- Joint study with Russia on evaluation of East Siberian atmospheric environment;
- Model Inter-comparison Study in Asia (a multilateral research activity).

Fellowship research program is also introduced annually. It has been promoted since 2005 which is an effective mechanism for encouraging young researchers from across the region to participate in air pollution research activities.

1.4.8 Capacity building activities

The technical capabilities and skills of the participating countries for acid deposition monitoring and assessment were significantly enhanced through a number of EANET activities including EANET individual training courses at the Network Center, and through the Network Center technical missions annually to all participating countries, to assist them in monitoring performance, laboratory operations, data management, and other procedures. Other activities to enhance the skills and knowledge of personnel included national workshops, annual expert meetings as well as scientific workshops on ecological impacts and other topics related to acid deposition. The Japan International Cooperation Agency (JICA), in cooperation with EANET, conducted the JICA Third Country Training Program in Thailand and the JICA Training Course on EANET in Japan to provide training on acid deposition and air quality management. Numerous EANET publications (Technical Manuals and Guidelines, data publications, reports on QA/QC projects, training materials, etc.) have been produced for use by technical staff, specialists and researchers involved in monitoring, data quality, and data management. All of these materials are available on the EANET website (http://www.eanet.cc/product/index.html).

1.4.9 Raising public awareness

EANET has promoted public awareness activities on acid deposition and other priority chemical species, including their effects, control and mitigation measures to various levels of the community which includes policy makers, scientists, general public, youth, school children and others to share common understanding on atmospheric environmental issues among the scientific community and policy makers by exchanging information through a network of experts, particularly for school children and teachers through a number of activities.

Two reports for policy makers were published titled **“**Goals, Achievements and Way Forward” and “Clean Air for Sustainable Future”. EANET has undertaken joint projects with participating countries to develop brochures and videotapes on acid deposition in the national language, and several times has held “Workshop on Public Awareness on Acid Deposition Problems” in the participating countries. An e-learning program on acid deposition problems was issued on the EANET website (http://www.eanet.cc/index.html) as one of the public awareness materials for environmental education and other public awareness activities. The capacity building workshops for policy makers were held in EANET to raise awareness of policy makers of participating countries to the adverse impacts on the environment caused by acid deposition. The Factsheets titled "Country efforts and achievements in combating acid deposition" were developed by all participating countries of EANET through collaboration and coordination with the Network Center and the Secretariat. The published public awareness materials were also issued on the EANET website (http://www.eanet.cc/product/index.html).

1.5 References

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Website (http://www.eanet.cc/product/index.html).

Chapter 2. Data Quality

2.1. Introduction

Data quality for acid deposition monitoring of EANET covers four environmental items – wet deposition, dry deposition (air concentration), soil and vegetation, and inland aquatic environment. Monitoring of wet and dry deposition was implemented in order to measure atmospheric concentrations and to evaluate fluxes of acidic substances to the land surface, while monitoring for soil/vegetation and inland aquatic environment has been carried out to assess adverse impacts on terrestrial and aquatic ecosystems. The monitoring data were used to evaluate the state of acid deposition as well as impacts on ecosystems.

Quality assurance and quality control (QA/QC) plays an important role in acid deposition monitoring to ensure and assure the measurement to be accurate, comparable and quantifiable. The QA/QC program is composed of four procedures: i) siting, ii) sampling and sample handling, iii) chemical analysis, and iv) QA/QC for the measurements at each site. Each National Center needs to execute various QA/QC activities, including development of national QA/QC programs, standard operating procedures (SOPs) and satisfaction of the data quality objectives (DQOs).

The Inter-laboratory Comparison project is also provided for all analytical laboratories involving in the EANET monitoring. The purposes of this project are to evaluate the analytical systems through the evaluation of analytical results, analytical instruments and their operating condition and other relevant and appropriate practices.

The QA/QC plays an indispensable role in acid deposition monitoring as well as other environmental measurements. QA/QC activities are also seen as an essential part of EANET monitoring to ensure that meaningful data are obtained. Through QA/QC activities, it is especially important that measured data satisfy specified levels of reliability and are accompanied by required information on the measurements themselves.

Several documents on QA/QC programs have been developed by EANET to guide receiving reliable data that can be comparable among the participating countries as well as with other monitoring networks outside of the East Asian region. The QA/QC programs cover activities for the components of the measurement/analysis system in a general way, i.e., in the field (sampling sites), laboratory, data management, and data reporting processes. According to EANET documents, QA/QC activities are documented by each relevant entity.

2.2. General procedures of QA/QC

The objectives of QA/QC programs are to obtain reliable and comparable monitoring data among the participating countries in East Asian region, as well as with other networks by ensuring data accuracy, precision, representativeness and completeness in the acid deposition monitoring.

2.2.1 Preparation of National Monitoring Plan

Every participating country of EANET has been required to prepare its National Monitoring Plan in accordance with the “Data Reporting Procedures and Formats for Acid Deposition Monitoring in East Asia”. The National Monitoring Plan of each participating countries were prepared after starting formal phase. The National Monitoring Plan should be reviewed every year and revised, if necessary.

The National Monitoring Plan consists following items:
1) Number of monitoring sites and their arrangement;
2) Measurement parameters and monitoring interval;

3) Participating laboratories for each monitoring activities, and;
4) Information on the respective monitoring sites with their scale maps.

2.2.2 Siting

EANET monitoring sites are classified into two basic categories, namely acid deposition monitoring sites and ecological survey sites. Acid deposition monitoring sites are sites to collect fundamental data on the temporal and spatial distribution of acid deposition, and they are further classified into 3 sub-categories: remote sites, rural sites and urban sites for the objectives of the monitoring. Ecological survey sites are those that provide basic data for assessing the effects of acidification on terrestrial ecosystems, and they are further classified into 2 sub-categories: survey sites and ecosystem analysis sites. The criteria used for classification of the sites are shown in Table 1.4.1 in Chapter 1.

2.2.3 Sampling and sample handling

The sampling has been carried out according to the monitoring manuals for each monitoring categories. Especially for the collected wet deposition samples and dry monitoring samples using filter pack method are stored in the refrigerator or added a biocide such as thymol to prevent or at least minimize possible conversion of the chemical species in the collected sample before chemical analysis.

2.2.4 Chemical analysis

The major ions, hydronium ion (H^+, as pH), sulphate ion (SO_4^{2-}), nitrate ion (NO_3^-), chloride ion (Cl^-), ammonium ion (NH_4^+), calcium ion (Ca^{2+}), potassium ion (K^+), magnesium ion (Mg^{2+}) and sodium ion (Na^+) in the collected samples are analyzed. The operation manuals for each monitoring categories provide several acceptable analytical techniques to be applied in the laboratories of the participating countries.

2.2.5 QA/QC procedure prior to the data submission to the National and Network Center

The quality of the chemical analysis was evaluated in terms of ion balance and conductivity checks. The evaluation was quantified as R_1 and R_2 as defined below.

$$R_1 = \frac{(C-A)}{(C+A)} \qquad R_2 = \frac{(\Lambda_{obs} - \Lambda_{calc})}{(\Lambda_{obs} + \Lambda_{calc})}$$

where the symbols are defined as below and the symbols of the ion denote their concentration on an equivalent basis.

$C = [H^+] + [NH_4^+] + [Ca^{2+}] + [K^+] + [Mg^{2+}] + [Na^+]$

$A = [SO_4^{2-}] + [NO_3^-] + [Cl^-]$

$$\Lambda = \lambda_{H+}[H^+] + \lambda_{NH4+}[NH_4^+] + \lambda_{Ca2+}[Ca^{2+}] + \lambda_{K+}[K^+] + \lambda_{Mg2+}[Mg^{2+}] + \lambda_{Na+}[Na^+] + \lambda_{SO42-}[SO_4^{2-}] + \lambda_{NO3-}[NO_3^-] + \lambda_{Cl-}[Cl^-]$$

The acceptable ranges for R_1 and R_2 are defined as a function of the concentration sums of the analytical suits, details of which are available in the operation manual. When either R_1 or R_2 does not meet the criteria, the samples are supposed to be subjected to reanalysis as well as detailed examination of all analytical procedures involved.

The analytical accomplishments do not meet the R_1 and R_2 criteria, it is strongly requires that some ionic species such as hydrogen carbonate (HCO_3^-) should be added to the analytical suits or the criteria should be modified as a function of pH. The acceptable ranges are encouraged to be revised in consideration of the ionic composition and concentration levels as well as the present state of arts of the analytical laboratories.

The data quality in terms of R_1 and R_2 are generally well within the acceptable ranges. When each measurement is evaluated for its validity, the chemistry of the sample should be explored from the viewpoint of the possible contribution of HCO_3^- and other ionic species. The performance of the round robin tests of the Inter-laboratory Comparison projects is quite helpful for the exploration.

2.2.6 Inter-laboratory Comparison Projects

Chemical analysis and evaluation of the Inter-laboratory Comparison are very important for the data quality of EANET. For Inter-laboratory Comparison, it has been done annually since 1998 during the preparatory phase of EANET in order to improve the quality assurance and quality control (QA/QC) by starting on wet deposition. There are different activities on the Inter-laboratory Comparison projects, i.e., wet deposition, dry deposition, soil, and inland aquatic environment. In 2009, they were the 12th, 5th, 11th, and 10th Inter-laboratory Comparison projects on wet deposition, dry deposition, soil, and inland aquatic environment, respectively.

The Inter-laboratory Comparison project is a round-robin test of common lots of artificial rainwater, which involves almost all analytical laboratories for the EANET monitoring. The purpose of this project is to evaluate the analytical systems through the evaluation of analytical accomplishments, analytical instruments and their operating condition and other relevant and appropriate practices. Particularly emphasis is placed on the issues: (i) to recognize the analytical precision and accuracy of the measurement of each laboratory, and provide an opportunity for the laboratories to improve the quality of the chemical analysis of wet deposition samples, and (ii) to improve reliability of analytical measurements through assessing the relevant techniques.

Throughout the Inter-laboratory Comparison project, the outlier data and the actual procedure in each participating laboratory have been evaluated using artificial rainwater sample for wet deposition monitoring, artificial filter sample for the dry deposition monitoring with filter pack method, actual soil sample for soil monitoring and artificial inland water sample for the inland aquatic environment monitoring.

2.3. Evaluation of Inter-laboratory Comparison Projects

2.3.1 Wet deposition

2.3.1.1. Participating countries

The Inter-laboratory Comparison surveys on wet deposition were carried out 12 times from 1998 to 2009 with participation of 22 to 31 laboratories in the thirteen countries: Cambodia, China, Indonesia, Japan, Lao PDR, Malaysia, Mongolia, Myanmar, Philippines, Republic of Korea, Russia, Thailand and Vietnam. Table 2.3.1 shows the number of participating laboratories in each country.

Table 2.3.1. Number of participating laboratories and countries in the Inter-laboratory Comparison Project on wet deposition.

Attempt Year	1st 1998	2nd 1999	3rd 2000	4th 2001	5th 2002	6th 2003	7th 2004	8th 2005	9th 2006	10th 2007	11th 2008	12th 2009
Cambodia						1	1	1	1	1	1	1
China	5	4	4	4	4	4	4	4	4	4	4	4
Indonesia	2	2	3	2	2	2	2	3	3	3	3	3
Japan	8	8	8	8	8	8	8	7	7	7	7	7
Lao PDR						1	1	1	1	1	1	1
Malaysia	1	1	1	1	1	1	1	1	1	1	1	1

Mongolia	1	1	1	1	1	1	1	1	1	1	1	1
Myanmar								1	1	1	1	1
Philippines	1	1	1	1	1	1	1	1	2	2	2	2
R. of Korea	1	1	1	1	1	1	1	1	1	1	1	1
Russia	1	1	1	1	2	2	2	2	2	2	2	2
Thailand	3	2	3	3	3	4	5	5	5	5	5	5
Vietnam	1	1	1	1	1	1	1	2	2	3	3	5
Numbers of laboratories	24	22	24	23	24	27	28	30	31	31	31	31
Numbers of countries	10	10	10	10	10	12	12	13	13	13	13	13

2.3.1.2. Preparation of artificial rainwater samples and their procedures

Two kinds of artificial rainwater of different concentration levels, high and low concentration samples were prepared by the Network Center to include all ions of analytical suites and distributed these samples to the participating laboratories in October/November in each year.

The samples required to be diluted 100 times prior to analyze. Measurement of pH and electronic conductivity (EC), and determination of the concentration of SO_4^{2-}, NO_3^-, Cl^-, Na^+, K^+, Ca^{2+}, Mg^{2+} and NH_4^+. Analytical results were submitted to the Network Center to summarize and review the results by comparing to the prepared values and further by carrying out some statistical analysis. The annual "Report of the Inter-laboratory Comparison Project 2009" includes the lists of the participating laboratories and countries together with their analytical results, laboratory codes and statistical descriptions.

The prepared values of the artificial rainwater samples from the 1st attempt (1998) to the 12th attempt (2009) were summarized in Table 2.3.2.

Table 2.3.2. The prepared values of each parameter in artificial rainwater for Inter-laboratory Comparison project of EANET.

year/sample #		pH	EC (mS.m^{-1})	SO_4^{2-}	NO_3^-	Cl^-	Na^+	K^+	Ca^{2+}	Mg^{2+}	NH_4^+
				(μmol.l^{-1})							
1998	No.1	4.05	7.94	83.5	93.3	129	95.8	11.1	41.1	13.1	84.8
	No.2	4.51	2.82	29.1	36.1	45.1	33.5	7.42	14.3	4.6	29.5
1999	No.1	4.14	6.38	67	75.0	104	77.0	8.9	33.0	11.0	68.0
	No.2	4.59	2.30	24.0	27.0	38.0	28.0	3.2	12.0	3.8	25.0
2000	No.1	4.10	6.23	59.7	63.3	101.3	51.3	9.9	29.4	11.7	60.5
	No.2	4.85	1.55	20.1	27.5	15.5	8.7	4.9	11.0	7.8	18.2
2001	No.1	4.10	7.45	85.0	93.3	108.4	68.4	15.8	41.1	18.7	87.8
	No.2	4.82	1.76	21.5	19.4	34.4	27.4	4.00	13.2	3.7	16.7
2002	No.1	4.30	3.75	40.3	51.0	33.7	13.7	6.92	19.1	7.02	42.4
	No.2	5.15	0.69	8.88	8.49	9.13	5.13	1.98	6.6	1.75	4.54
2003	No.031	4.52	3.44	44.7	30.9	66.0	46.1	6.9	20.5	7.0	48.3
	No.032	4.80	1.48	12.0	21.3	29.6	25.6	2.5	4.4	3.4	15.1
2004	No.041	4.60	3.93	58.5	41.4	76.6	66.6	7.0	38.9	9.7	39.3
	No.042	5.00	1.33	17.5	18.4	22.4	20.5	5.0	10.0	2.7	15.1
2005	No.051	4.66	3.32	43.7	40.3	68.5	56.5	6.9	23.2	11.7	40.9
	No.052	5.05	1.05	14.4	13.2	15.3	10.3	3.0	7.6	3.1	13.6
2006	No.061	4.72	3.10	45.8	36.3	57.5	44.5	6.9	23.8	11.7	43.9
	No.062	5.15	1.21	16.9	15.0	24.5	20.5	4.9	9.3	3.5	15.1
2007	No.071	4.64	3.72	54.9	43.6	69.0	53.4	8.3	28.6	14.0	52.7
	No.072	5.00	1.47	14.0	21.0	38.8	30.8	4.9	6.4	7.0	15.1
2008	No.081	4.49	3.97	49.5	47.4	75.8	56.8	9.1	25.7	12.6	47.5
	No.082	5.10	1.18	14.7	16.2	23.4	15.4	3.8	7.6	6.2	14.2
2009	No.091	4.52	4.23	57.8	46.7	81.9	59.9	9.1	29.0	14.8	57.5
	No.092	4.92	1.66	19.9	22.0	34.0	24.0	5.9	9.3	7.0	21.2

2.3.1.3. Results

The EANET QA/QC program has prescribed the Data Quality Objectives (DQOs) as a value of ±15% for each analytical suit. The results of the present project were evaluated in terms of the excess of the DQOs value. The flag "E" was added to the data that exceeded DQOs by a factor of 2 (±15%~±30%), and the flag "X" to the data that exceeded DQOs by a factor of over 2 (<-30% or >30%). A set of the data for each sample was evaluated with the data checking procedures.

The Inter-laboratory Comparison surveys were carried out 12 times, and the overall percentages of flagged data, “E” and “X” respectively, are shown in Figure 2.3.1. The percentage of data within the data quality objectives (DQOs) increased from 75-78% (in 1998) to 89-96% (in 2009) . The percentage of the flagged data, which may be related to the concentration levels of the artificial rainwater, has been gradually decreasing toward the 12th attempt.

Predominant decreases of the relative standard deviations (R.S.D.) have been reported in the 1st “Periodic Report on the State of Acid Deposition in East Asia”. After 5 years of previous summarization, the R.S.D. of project data sets during from 2005 to 2009 was also shown in Figure 2.3.2. As shown in this figure, the relative standard deviations were transited as almost flat, and it is suggested that any further improvement of quality management system might be required to transcend this convergent trend

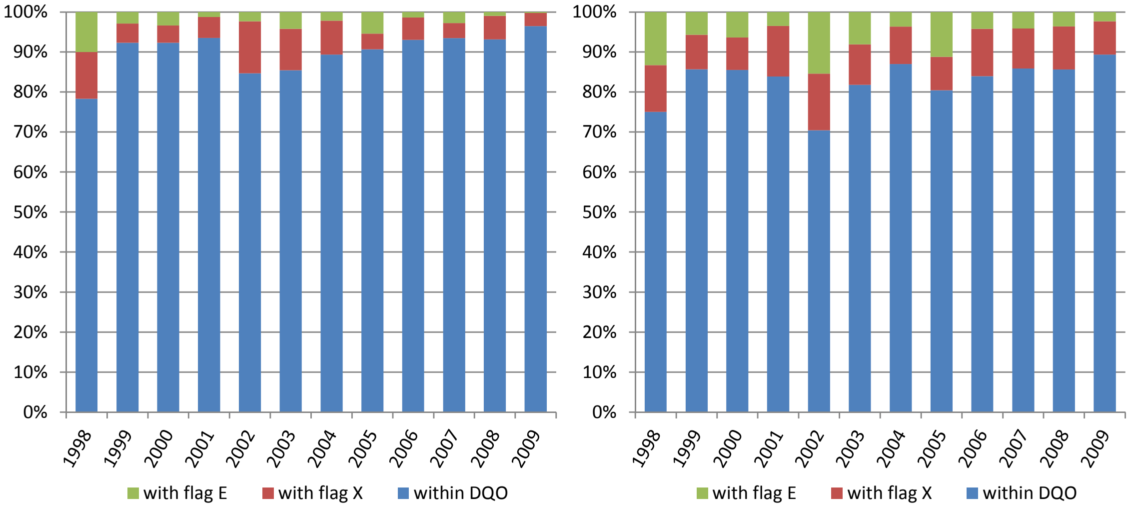


Figure 2.3.1. Overall comparisons of 1st to 12th Inter-laboratory Comparison projects.

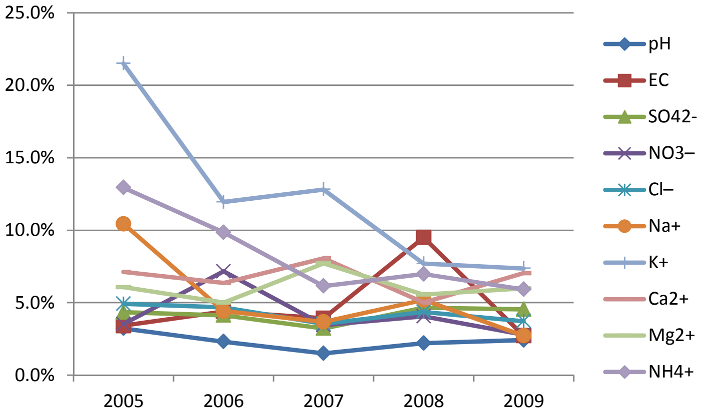


Figure 2.3.2. Relative standard deviation of each constituent data (2005-2009) for high concentration sample.

2.3.2 Dry deposition

2.3.2.1. Participating countries

The Inter-laboratory Comparison surveys were carried out 5 times from 2005 to 2009 with participation of 18 to 23 laboratories in the eleven countries: Cambodia, China, Indonesia, Japan, Malaysia, Mongolia, Philippines, Republic of Korea, Russia, Thailand and Vietnam. Table 2.3.3 shows the number of participating laboratories in each country.

Table 2.3.3. Number of participating laboratories and countries in the Inter-laboratory Comparison Project on dry deposition.

Attempt Year	1st 2005	2nd 2006	3rd 2007	4th 2008	5th 2009
Cambodia					1
China		1	1	1	1
Indonesia	1	1	1	1	3
Japan	8	7	7	7	7
Lao PDR					
Malaysia	1	1	1	1	1
Mongolia	1	1		1	1
Myanmar					
Philippines	1	1	1	1	1
R. of Korea	1	1	1	1	1
Russia	1	1	1	1	1
Thailand	3	4	4	4	4
Viet Nam	2	2	2	3	3
Numbers of laboratories	19	19	18	20	23
Numbers of countries	9	10	9	10	11

2.3.2.2. Preparation of artificial filter samples and their procedures

Two kinds of filter samples, one contained two ions ($SO_4^{2-}$and Cl^-), the other contained one ion (NH_4^+), were prepared. The blank filters, which were impregnated with K_2CO_3 or H_3PO_4 but did not contain any SO_4^{2-}, Cl^-, or NH_4^+, were also prepared These samples were distributed to the participating laboratories in October/November in each year.

The recommended procedure for sampling and analyzing by the filter pack method is described in EANET Document, "Technical Document for Filter Pack Method in EAST Asia" (EANET, 2010a). Each filter sample was put in a centrifuge tube, and solvent was directly poured into the tube for extraction.

The annual "Report of the Inter-laboratory Comparison Project 2009" includes the lists of the participating laboratories and countries together with their analytical results, laboratory codes and statistical descriptions.

The amount of ions in the artificial filter samples from the 1st attempt (2005) to the 5th attempt (2009) were summarized in Table 2.3.4.

Table 2.3.4. The prepared values of each parameter in artificial filter for Inter-laboratory Comparison project of EANET.

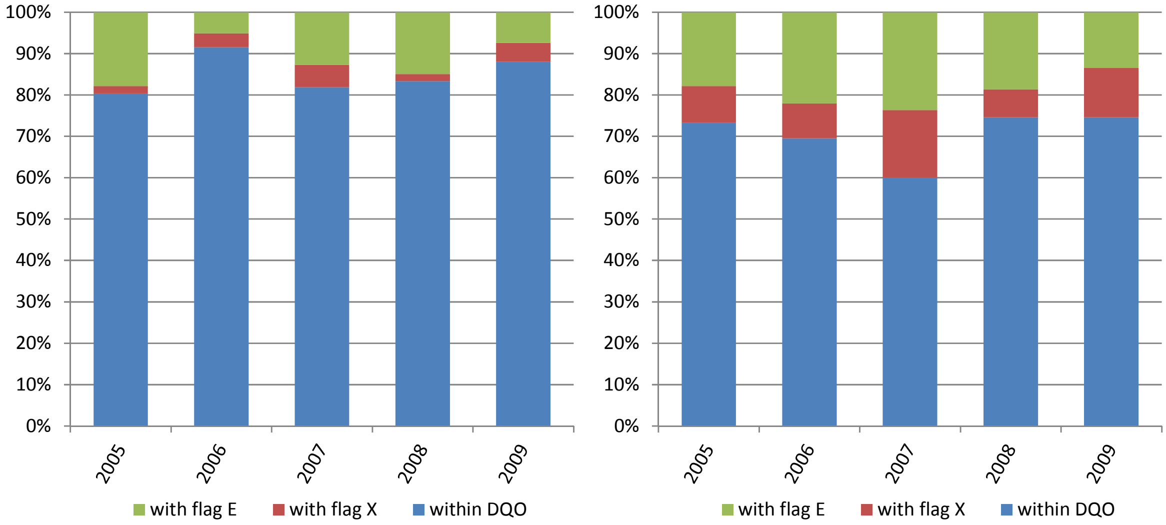
year/sample No.		SO_4^{2-} (μg)	Cl^- (μg)	NH_4^+ (μg)
2005	No.051	25.0	2.80	5.30
	No.052	70.0	30.0	20.0
2006	No.061	40.0	3.50	6.50
	No.062	140	40.0	50.0
2007	No.071	20.0	1.60	4.60
	No.072	80.0	25.0	30.0
2008	No.081	30.0	2.50	5.60
	No.082	110	32.0	35.0
2009	No.091	35.0	3.00	5.30
	No.092	120	38.0	40.0

2.3.2.3. Results

The EANET QA/QC program has prescribed the Data Quality Objectives (DQOs) as a value of ±15% for each analytical suit. The results of the present project were evaluated in terms of the excess of the DQOs value. The flag "E" was added to the data that exceeded DQOs by a factor of 2 (±15%~±30%), and the flag "X" to the data that exceeded DQOs by a factor of over 2 (<-30% or >30%). A set of the data for each sample was evaluated with the data checking procedures.

The Inter-laboratory Comparison surveys were carried out 5 times, and the overall percentages of flagged data, “E” and “X” respectively, are shown in Figure 2.3.3. The percentage of data within the DQOs increased from 75-78% to 89-96%. The percentage of the flagged data, which may be related to the amount levels of the artificial filter, has been gradually decreasing toward 5th attempt.

Predominant decreases of the relative standard deviations (R.S.D.) have been reported in the 1st “Periodic Report on the State of Acid Deposition in East Asia”. After 5 years of previous summarization, the R.S.D. of project data sets during from 2005 to 2009 was also shown in Figure 2.3.4. As shown in this figure, the relative standard deviations were varied. This variation was considered that the difference of the extraction procedure caused the variation of the results among the participating laboratories. It is suggested that any further improvement of quality management system might be required to transcend this convergent trend



Higher concentration sample *Lower concentration sample*

Figure 2.3.3. Overall comparisons of the 1st to 5th Inter-laboratory Comparison projects.

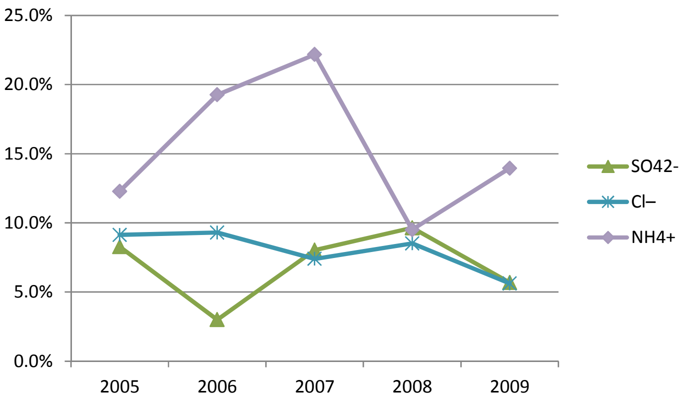


Figure 2.3.4. Relative standard deviation of each constituent data (2005-2009) for higher concentration sample.

2.3.3 Soil

2.3.3.1. Outline

The project on soil sample analysis started in 1999 as one of the activities within the QA/QC programs. 14 to 16 laboratories participated from the following ten countries: China, Indonesia, Japan, Malaysia, Mongolia, Philippines, Republic of Korea, Russia, Thailand, and Vietnam, participated in the projects. Air-dried soil samples, which were prepared by the Network Center, were distributed to the laboratories every year except 2001. The laboratories carried out the whole procedures of the soil analysis including extraction, instrumental analysis or titration, and reporting. Performance at each step would be responsible for the inter-laboratory variations. In order to eliminate the differences in the sample preparation, therefore, samples of soil extract were also distributed in 2001 and 2002 to evaluate the procedures of respective steps. Distributed samples of the respective years and mandatory parameters to be measured for soil samples were listed in Tables 2.3.5 and 2.3.6, respectively.

Table 2.3.5. Distributed samples and their characteristics.

Year	Sample 1	Sample 2
1999 (1st)	No. 991: Acrisols (red soil)	No. 992: Gleysols (grey soil)
2000 (2nd)	No. 001: Cambisols (brown forest soil)	No. 002: Andosols (black soil: used for the 3rd and 4th Project)
2001 (3rd)	No. 011: Soil extract by CH_3COONH_4 solution (for exchangeable base cations)	No. 012: Soil extract by KCl solution (for exchangeable acidity, Al, and H)
2002 (4th)	No. 021: Andosols (black soil: soil sample)	No. 022: Soil extract by CH_3COONH_4 solution (for exchangeable base cations)
2003 (5th)	No. 031: Cambisols (brown forest soil)	No. 032: Arenosols (red soil)
2004 (6th)	No. 041: Andosols (black soil)	No. 042: Cambisols (brown forest soil)
2005 (7th)	No. 051: *Quercus* serrate (secondary forest)	No. 052: *Pinus densiflora* (secondary forest)
2006 (8th)	No. 061: Eutric Cambisols (brown forest soil)	No. 062: Dystric Cambisols (brown forest soil)
2007 (9th)	No. 071: Acrisols (red soil)	No. 072: Combisols (brown forest soil)
2008 (10th)	No. 081: Combisols (brown forest soil)	No. 082: Acrisols (red soil)
2009 (11th)	No. 091: Combisols (brown forest soil)	No. 092: Acrisols (red soil)

Note: Soils were collected in Akita, Aichi, Niigata, and Tottori prefectures of Japan in 1999, 2000, 2003, and 2004, respectively.

Table 2.3.6. Mandatory parameters to be measured for the soil samples.

Parameters	Unit
a) Moisture Content	wt %
b) pH (H_2O), pH (KCl)	
c) Ex- base cations, such as Ca^{2+}, Mg^{2+}, K^+, and Na^+	cmol(+) kg^{-1}
d) Ex- Acidity, Al^{3+}, and H^+	cmol(+) kg^{-1}

Note: Ex-, exchangeable

2.3.3.2. Major outcomes

Inter-laboratory variations in pH (H_2O) and pH (KCl) were very small during 2002 to 2009, as shown by CVs less than 10%. The ranges from upper to lower quartile, which are indicated by box, are kept within 0.2 pH units in almost all samples (Figures 2.3.5 and 2.3.6). We speculate from the figure that participating countries can detect the difference of around 0.5 pH unit. Meanwhile, there were a few extreme outliers in each sample. This outlier might be critical for the temporal variation within the same plot.

Inter-laboratory variation in Ex-Ca and Ex-Acidity were shown on the scales of high concentration and low concentration (Figures 2.3.5 and 2.3.6). Large variation in Ex-Ca has been observed for 8 years, as shown by CVs more than 50%. On a large scale (0 to 15 $cmol_c$ kg^{-1}) we can detect the difference between the sample more than 2 or 3 $cmol_c$ kg^{-1} (high concentration sample) and the others (low concentration sample). However, at the area of low concentration, inter-laboratory variation might be well over the difference between the samples. Because exchangeable base cations do not drastically change in time at the same site, accuracy of analysis should be improved in the participating country covered with poor-nutrient soils (e.g. tropical soil or undeveloped soil like a desert). In addition, the minimum values of Ex-Ca were very low in almost all samples, even if the median value were very high. It is proposed that the laboratory providing the minimum value has a fatal fault for measuring the exchangeable base cations (e.g. background of extraction liquid, maintenance of Atomic Absorption Spectrometer (AAS) and so on). Inter-laboratory variations in Ex-Acidity were relatively small in comparison with Ex-Ca, except the samples of very low concentration (e.g. No.052 and No.092). From each range as shown by boxes, we assume that participating country can enough detect the temporal variation in soil acidity of not only strong acidic soil but also moderately or weakly acidic soil.

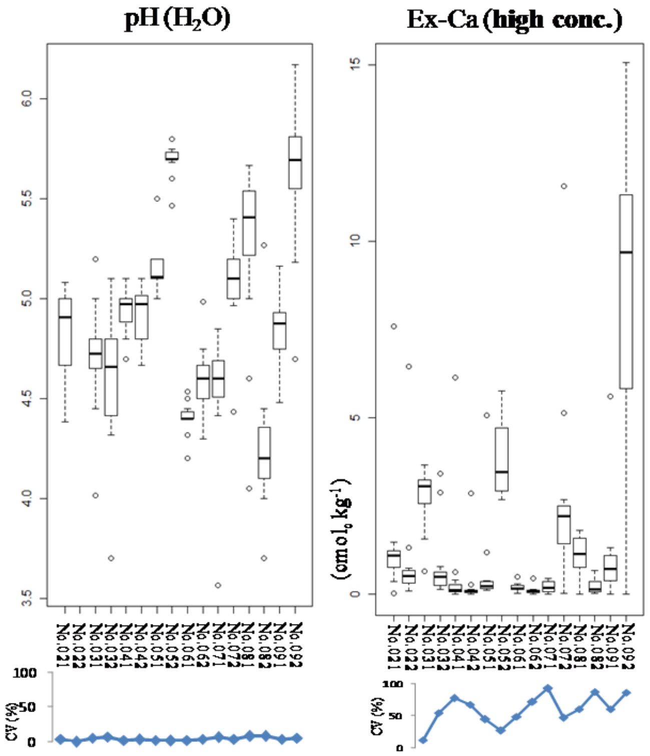


Figure 2.3.5. Box-and-whisker plots of inter-laboratory variations in pH, Ex-Ca and Ex-Acidity of 16 soil samples from 2002 to 2009, and the variation in CVs (entire dataset).

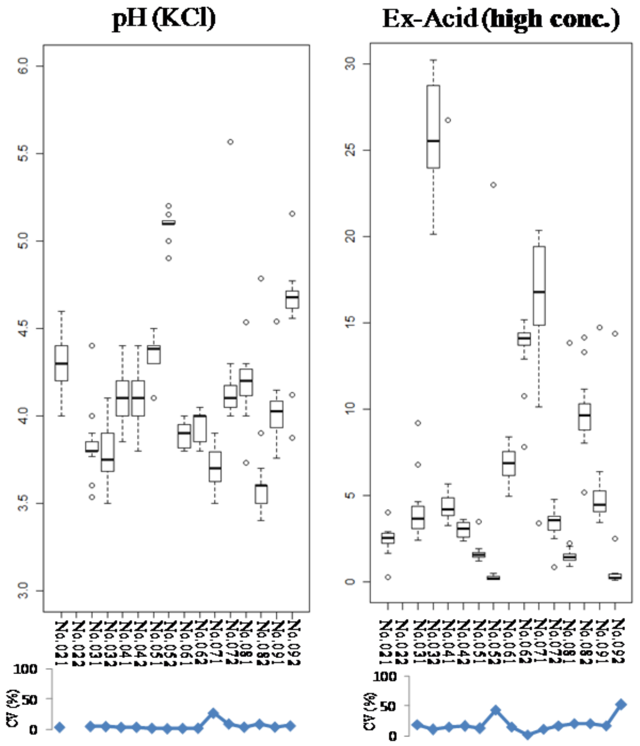


Figure 2.3.6. Box-and-whisker plots of inter-laboratory variations in pH, Ex-Ca and Ex-Acidity of 16 soil samples from 2002 to 2009, and the variation in CVs (entire dataset).

2.3.4 Inland aquatic environment

2.3.4.1. Outline

The project on inland aquatic environment sample analysis started in 2000 as one of the activities within the QA/QC programs. 13 to 22 laboratories participated from the following ten countries: China, Indonesia, Japan, Lao PDR, Malaysia, Mongolia, Philippines, Russia, Thailand, and Vietnam, participated in the projects. One artificial inland water sample, which contained all analyzed ions and was prepared by the Network Center, was distributed to the laboratories every year. The laboratories carried out the whole procedures of the inland water analysis including instrumental analysis or titration, and reporting. Performance at each step would be responsible for the inter-laboratory variations. Distributed samples of the respective years and mandatory parameters to be measured for inland aquatic environment samples were listed in Table 2.3.7.

Table 2.3.7. Mandatory parameters and their designed concentrations of artificial inland aquatic environment samples.

Parameter		2000 1st	2001 2nd	2002 3rd	2003 4th	2004 5th	2005 6th	2006 7th	2007 8th	2008 9th	2009 10th
pH		5.52	6.10	6.69	7.31	7.00	7.18	7.01	6.88	7.41	7.00
EC	(mS.m^{-1})	3.64	7.45	7.76	7.41	4.00	6.06	4.91	5.64	6.57	3.75
Alkalinity	(meq.l^{-1})			0.214	0.238	0.119	0.179	0.159	0.206	0.238	0.149
SO_4^{2-}	(meq.l^{-1})	6.37	15.7	10.6	8.29	4.48	8.00	6.04	7.96	6.72	2.99
NO_3^-	(meq.l^{-1})	1.64	3.19	4.13	5.70	3.16	3.33	0.74	0.82	0.82	0.54
Cl^-	(meq.l^{-1})	3.71	5.47	4.87	4.28	2.46	3.67	3.65	3.16	5.80	3.62
Na^+	(meq.l^{-1})	2.36	3.54	8.07	8.22	4.31	6.48	3.85	5.05	5.78	3.47
K^+	(meq.l^{-1})	0.39	0.77	0.77	1.16	0.77	0.71	1.02	0.86	1.21	1.00
Ca^{2+}	(meq.l^{-1})	2.36	3.53	4.12	3.23	1.63	3.01	2.48	3.1	2.48	1.02
Mg^{2+}	(meq.l^{-1})	0.19	0.38	0.57	0.76	0.38	0.43	0.96	0.80	1.59	1.01
NH_4^+	(meq.l^{-1})	0.27	2.73	0.27	0.20	0.22	0.29	0.16	0.20	0.29	0.21

2.3.4.2. Major outcomes

The Inter-laboratory Comparison projects of EANET have been carried out 10 times, and the results showing the percentage of flagged data and the percentage of data that satisfied the Data Quality Objectives are shown in Figure 2.3.7.

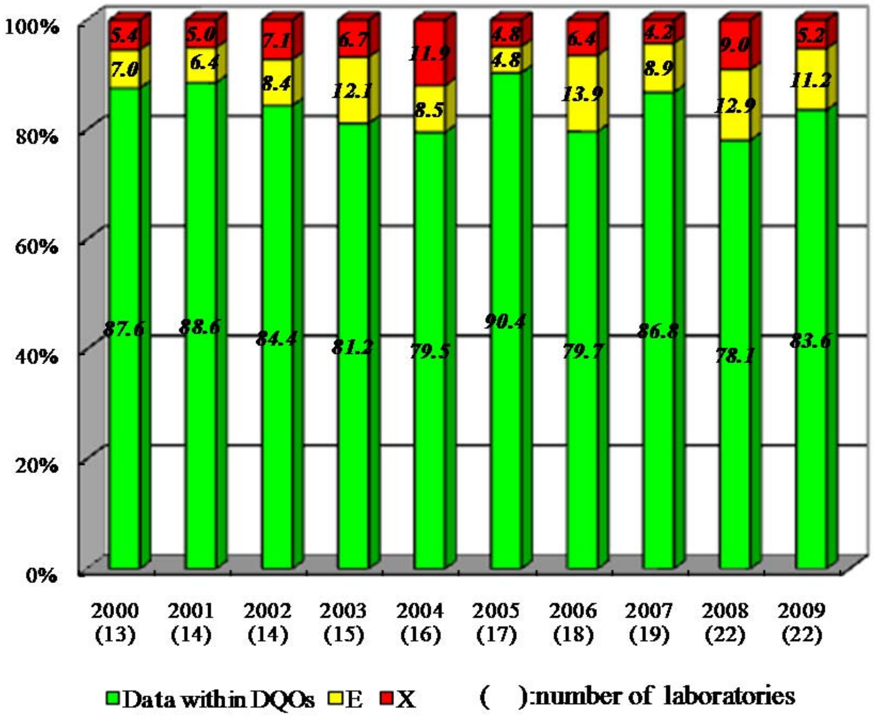


Figure 2.3.7. Comparison of the results from the Inter-laboratory Comparison projects.

The graph shows the number of laboratories increase and data within Data Quality Objectives decrease. But it was relatively good results in this attempt.

The comparison for each parameter from the 1st to 10th project with the percentage of flagged data is shown in Figure 2.3.8.

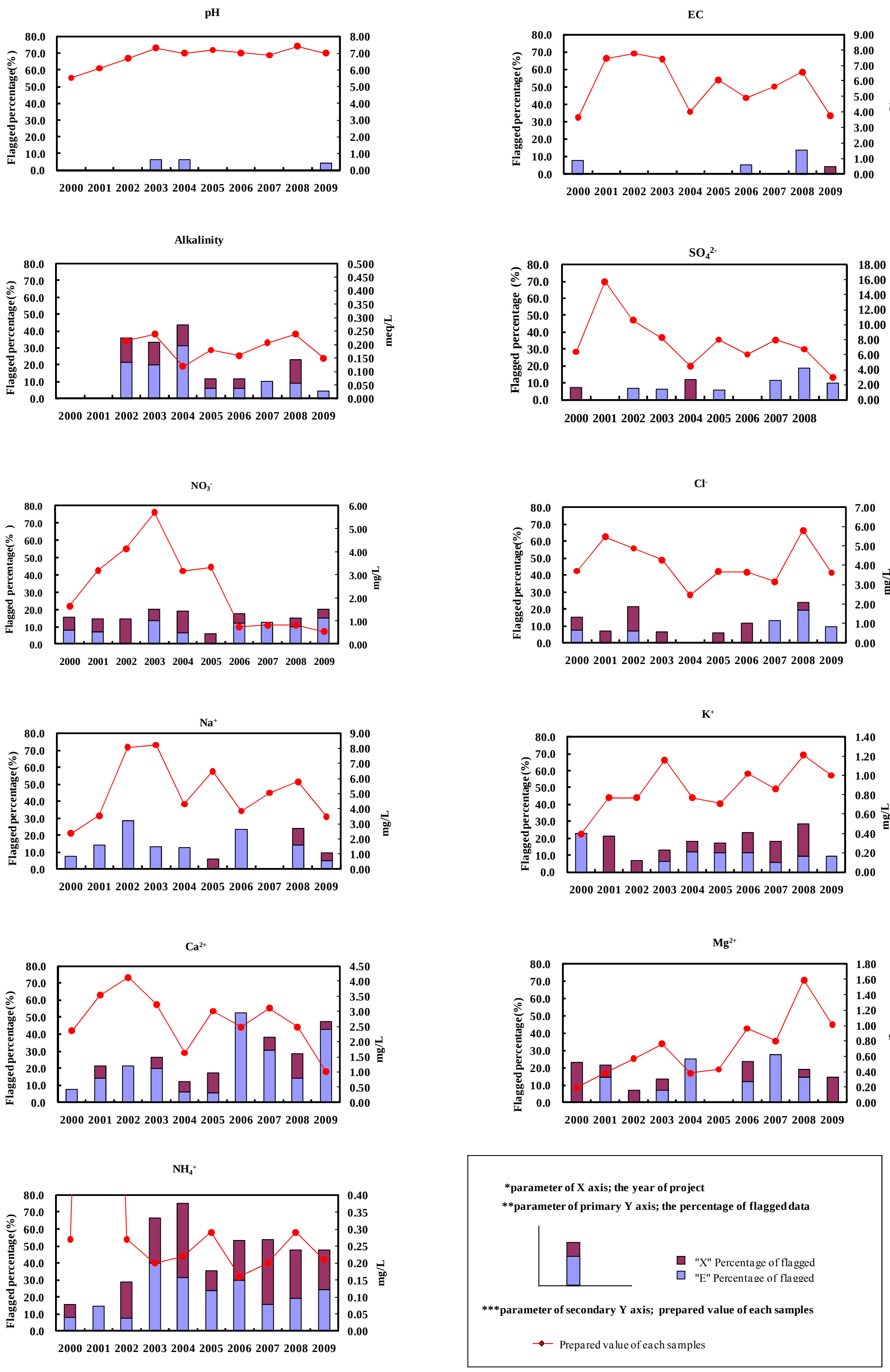


Figure 2.3.8. Comparison of the percentage of flagged data for each parameter in the Inter-laboratory Comparison projects.

The percentage of flagged data of NH_4^+ increased when the concentration of NH_4^+ was low. Concerning other parameters, there were no clear relationship between the concentration and the percentage of flagged data.

The percentages of flagged cations were larger than those of anions in this project. Especially the percentages of flagged Ca^{2+} and NH_4^+ were more than 25%. The percentage of flagged NH_4^+ was larger than other parameters in every survey except the 1st- 3rd projects. The percentage of flagged Ca^{2+} of the 7th - 10th projects was relatively high. It was not clear why the number of flagged data for Ca^{2+} increased. In this project the percentage of flagged K^{+} was lower than last attempt. But it seems that there is necessity to pay attention to the analysis of Ca^{2+} and NH_4^{+} in inland aquatic water.

2.4 Evaluation of the measurements

One of the objectives of the QA/QC program is to obtain reliable data that can be comparable among the countries of the East Asian region, as well as with other networks by ensuring data accuracy, precision, representativeness and completeness in acid deposition monitoring. All activities of data management are directed to ensure acceptable quality assurance levels.

2.4.1 Data flow for the reporting and qualification

The annual measurements in a participating country are subjected to the data qualification by its national committee of data evaluation. Some suspected datasets will be feedback to the analytical laboratories or appropriate organizations. The principal flow chart of data submission and verification is presented in Figure 2.4.1.

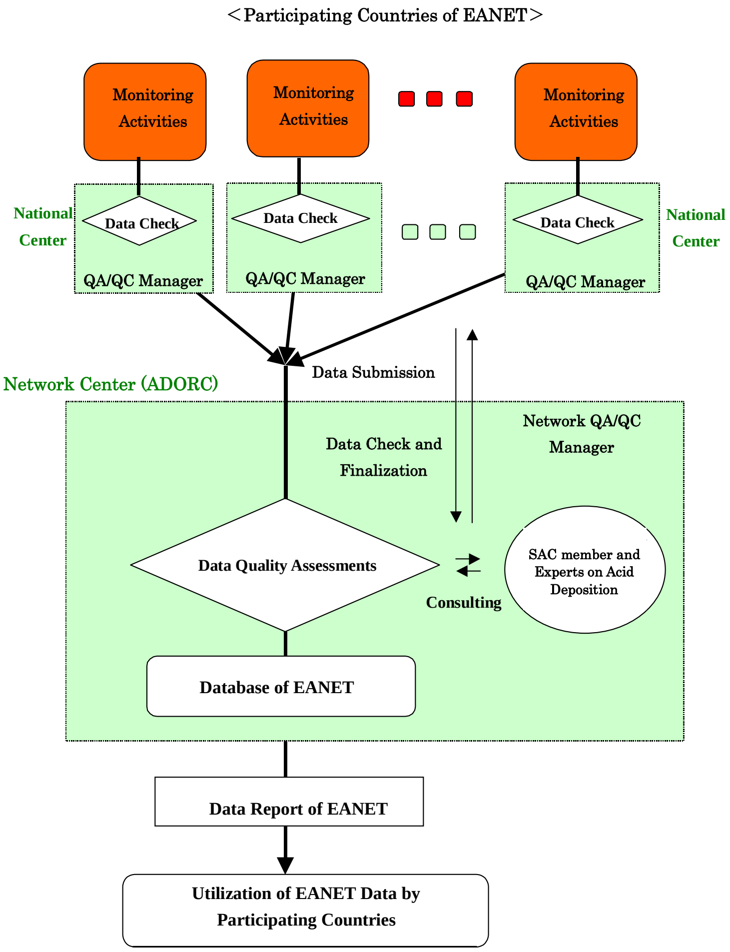


Figure 2.4.1. Flow Chart of EANET Monitoring Data.

All datasets are eventually collected by the Network Center (NC) where the datasets are further examined and qualified through communications with national centers. The datasets qualified so far are submitted to the international data verification group for the final checking. The international experts of the monitoring activities qualify each of the datasets in a careful and detailed manner. After discussion between the Network Center and national centers, the qualified datasets are submitted to the Scientific Advisory Committee to be scientifically and technically approved.

2.4.2 Validation of the data

All the data were checked in terms of ion balance (R_1) and the conductivity agreement (R_2) as presented in 2.2.5. If a sample or individual datum has problems including "insufficient sample volume" or "low precision", the flags corresponding to these problems were attached to the data. Details of data management are described in the "Quality Assurance/Quality Control (QA/QC) Program for Wet Deposition Monitoring in East Asia" adopted at the Second Interim Scientific Advisory Group Meeting in March 2000.

2.4.3 Data completeness

Periodic reporting of mean concentrations and deposition should be based on the full operation measurements without any missing data, which would be actually inevitable in the long-term regional monitoring network activities. The quality of the annual and monthly means was, therefore, evaluated by setting two criteria of the data completeness. When the fraction of the missing data exceeds the first criterion but meets the second criterion, the annual or monthly mean was shadowed in the summary tables of the annual data reports. Of course, if the fraction even exceeds the second criterion, the mean was not approved as an official figure. In this way , the means are qualified in the tables for further analysis and interpretations.

2.4.4 Site representativeness

The characteristics of the individual site were reported by answers on a series of questions and photos from four or more directions. And the nature of the sites (e.g. urban, rural, remote, or ecological) was kept in mind for the analyses.

The specific features of EANET monitoring are to combine the measurements in different environmental media such as the atmosphere or surface water and more complicated soils or vegetation. The first ones are characterized by concentration levels of air or water pollutants while for latest we need also to take ecosystem peculiarities and their "annual life cycles" into account. The possible impact of anthropogenic sources (like emission, sludge discharge or land use) on level of pollution could be concerned during the site establishing. The environmental conditions have to be taken into account also in a view of possibility to trace ecosystem changes under influence of long range transport of air pollutants.

EANET set up three categories for monitoring sites of atmospheric deposition presented in the Guidelines and Technical Manuals (EANET, 2000b) with respect to the level of impact on surrounded area: urban, rural and remote stations. The site criteria define the distance of possible air pollution sources with different intensity, land use in the vicinity of different spatial scale, etc. The specific information are required and collected on site condition in accordance with formats of National Monitoring Plan for the surrounded areas in a distance to 150 m, 0.15-10 km and 10-50 km from the monitoring sites.

For the rural and remote sites, the applied requirements were established based on the experience gained from activities of other networks in the world. These criteria specifications are closely correspondent to the similar requirements of well-operated monitoring networks such as EMEP or regional GAW-PC network (EMEP, 2001; WMO, 2004). However, mentioned networks are not concerned urban atmospheric pollution which became a serious problem in East Asian megacities and other big cities especially in the tropics. Those kinds of measurements are particularly related to a number of national networks as well as to specific international initiatives directed to urban pollution.

Taking into account the differences in atmospheric processes and emission impact in urban and non-urban conditions and evaluation of regional pollution has to be done separately for mentioned areas. And the nature of the sites (e.g. urban, rural, remote, or ecological) was kept in mind for the analyses. The characteristics of the individual site were reported by answers on series of questions and photos from four or more directions. For the practical purposes, the brief overview of site conditions could be observed at EANET site (EANET, 2010a).

The site is evaluated from a perspective of available potential sources of sample contamination which could violate its representativeness of the area of interest on the three different spatial scales: on-site, local, and regional. Detailed examinations of the site by inspection and the related documents will evaluate the type of the site. Any changes of the site conditions on three different scales (on-site, local and regional) and relocation of the sampler may require a reevaluation of the type of the site (WMO, 2004).

The sites of fourth category have a focus on ecological monitoring of natural ecosystems which is also, might be referred to relevant conditions for the atmospheric deposition measurement. They are commonly stated as ecological sites being correspondent to one of these three mentioned above in terms of the location conditions, but its focus of measurement programs is on the data acquisition of the input to the ecological elements of interest.

Guideline for Acid Deposition Monitoring in East Asia (EANET, 2000b) recommended that ecological sites for soil and vegetation monitoring (basic survey program) and monitoring on inland aquatic environment have to be preferably established near the deposition monitoring sites or in their vicinity. In particular for soil and vegetation monitoring, more detailed criterion was proposed for the distance from the atmospheric deposition monitoring site as “within a radius of approximately 50 km”. Moreover, it was suggested that two forest plots with different soil types be selected in each area within 50 km from the deposition site.

The current situation on co-location of basic survey sites for ecological monitoring and the nearest deposition monitoring sites was presented in Table 2.4.1. Soil and vegetation monitoring and monitoring on inland aquatic environment are conducted at 28 plots in 10 countries and 18 lakes/rivers in 10 countries, respectively. Most of ecological monitoring sites are corresponded to the deposition monitoring sites, however, their classification could be different. Totally, both soil and vegetation monitoring and monitoring on inland aquatic environment are conducted in the vicinity of 4 deposition monitoring sites in China, 2 sites in Japan, 1 site in Malaysia, 2 sites in Philippines, 2 sites in Russia, 1 site in Thailand, and 1 site in Vietnam. It is expected that the deposition data collected in the nearest monitoring sites would be used for data interpretation of ecological monitoring at these sites.

However, the current monitoring system seems could not provide evident information on implication between the ecological and deposition data. Monitoring activities for ecosystems and atmospheric impact should be integrated more for discussion on both quantitative and qualitative relations. Catchment-scale monitoring may be one of approaches for this further investigation including some modeling. Hopefully, the regular catchment-scale monitoring will be started in the EANET countries based on the Guideline for Catchment-scale Monitoring (EANET, 2010b) in near future.

Table 2.4.1. Basic survey sites for ecological monitoring and the nearest deposition sites.

Country	Site for deposition monitoring	Plot for soil and vegetation monitoring	Site for monitoring on inland aquatic environment
China	Jinyunshan (Chongqing)	Jinyunshan	Jinyunshan Lake
	Jiwozi (Xi’an)	Dabagou	Jiwozi River
	Xiaoping (Xiamen)	Xiaoping	Xiaoping Dam
	Zhuxiandong (Zhuhai)	Zhuxiandong	Zhuxiandong Stream
Indonesia	Serpong	Bogor Research Forest (Dramaga Experimental Forest)	-
	Bandung	-	Patengang Lake
	-	-	Gunung Lake
Japan	Ijira	Ijira	Ijira Lake
		Yamato	
	Banryu	Banryu-2*2	Banryu Lake
		Iwami “rinku” Factory Park*2	
Lao PDR	Vientiane	-	Nam Hum Lake
Malaysia	-	Pasoh Reserve Forest	-
	Petaling Jaya	Sungai Lalang Reserve Forest	Semenyih Dam
	Danum Valley	-	Tembaling River
Mongolia	Ulaanbaatar	Bogdkhan Mountain	-
	Terelj	-	Terelj River
Philippines	Los Banos	Mt. Makiling	Pandin Lake
		UP Quezon, Land Grant	
	Metro Manila	La Mesa Watershed	-
	Mt. Sto. Tomas	Boneco Long Term Ecological Research Site	Ambulalakao Lake
Republic of Korea	Imsil	Mt. Naejang	-
Russia	Irkutsk	Irkutsk	-
	Listvyanka	Bolshie Koty	Pereemnaya River
		Pereemnaya river catchment	
	Mondy	Ilchir Lake area	-
		Okinskoe Lake area	
		Solar Observatory	
	Primorskaya	Primorskaya area	Komarovka River
Thailand	Vachiralongkorn Dam *3	Vachiralongkorn Dam	Vachiralongkorn Dam
		Vachiralongkorn Puye	
Vietnam	Hoa Binh	Cave of Heaven	Hoa Binh Reservoir
		Thang Ranh	

*Note: S, Soil monitoring; F, Forest monitoring; *1. Classification by FAO/UNESCO has not been reported; *2.The sites around Banryu deposition site were relocated in 2001; *3. Kao Lam Dam was renamed to Vachiralongkorn Dam.*

2.4.5 Overall data quality

Evaluation of the datasets from the four viewpoints mentioned above would have ensured that a large fraction of the measurements in the early stage are acceptable, and recent datasets are improved in the data quality.
However, the room for improvement may be identified in the comparison with established Data Quality Objectives for other international measurement networks. The high quality targets are presented in Table 2.4.2 by the reference to requirement of regional WMO-GAW Precipitation Chemistry network and EMEP.

Table 2.4.2. Data Quality Objectives for Precipitation Chemistry analysis (accuracy in single analysis in laboratory).

Analyzed component in laboratory	Data Quality Objectives established for: GAW-PC network (WMO, 2004)	EMEP network (EMEP, 2001)
pH	0.07 units	0.1 units
conductivity	7%	15 %
acidity	25%	15 %
sulphate	7%	10 %
nitrate	7%	10 %
ammonium	7%	15 %
chloride	10%	15 %
sodium	10%	15 %
potassium	20%	15 %
calcium	15%	15 %
magnesium	10%	15 %

It should be noted that Data Quality Objectives for the most of analyzed constituent in regional WMO-GAW PC laboratories are less in range of value accuracy, than in EANET, especially for the most interested compounds like oxidised sulphur and oxidised nitrogen. The similar limits (as in EANET) were established in EMEP, but recently, the relevant documents on EMEP measurement strategy proposed the changes of those criteria (EMEP, 2009).

It is also useful to consider the comparison of approach being used by different international intercomparison projects for evaluation of analytical results and network laboratories performance as well as an application of some interesting tool like Youden plots (NIVA, 2011) and ring diagrams (WDCPC, 2011).

2.5 Conclusions and recommendations

2.5.1 Observation in the Data Report and the Report on Inter-laboratory Comparison projects

The Network Center has to evaluate the monitoring data and to improve their quality. However, the Network Center could evaluate the data only checking the submitted report and data from the participating country, and it has been difficult to grasp actual QA/QC activities in each participating country.

The deviations of some items indicated decreasing tendency, the deviations of some cations indicated higher CV% and especially, the deviations of electric conductivity and hydronium ion concentration (corresponding to the pH) indicated increasing tendency.

These phenomena can be observed easily ; however, the actual cause investigation for solving the problems is difficult only by detailed checking of submitted reports.

2.5.2 General QA/QC activities

The QA/QC system in the participating countries should be harmonized by establishing the common quality management system. Since only the requirements for QA/QC were described in the Technical Manuals, it may be difficult to recognize and to harmonize how the data quality should be kept.

The preparation of the Technical Manual on the Establishment of Quality System for EANET can be significant.

2.5.3 National Monitoring Plan

The National Monitoring Plan of each participating countries were prepared after starting regular phase. The National Monitoring Plan should be reviewed every year and revised if necessary.

The Network Center is now difficult to grasp current situation of the participating countries because of being kept the National Monitoring Plan unchanged.

When there is a revision in the National Monitoring Plan, the National Monitoring Plan shall be revised immediately and should be submitted to the Network Center.

2.5.4 Implementation of the technical mission

The technical mission has been conducted as one of the additional budget activities of EANET. And it has been effective to build the capacity of acid deposition monitoring in the participating countries.

The problems in the laboratories in charge of the EANET monitoring in the participating countries may not be able to improve their abilities and data quality. Even in the laboratories which have been already regarded as high potential, there might be unrevealed problem. In this case, it may be necessary to reconsider dispatching the technical mission.

2.5.5 Implementation of the Inter-laboratory Comparison project

The Inter-laboratory Comparison project has been conducted to confirm the abilities of the laboratory in charge of EANET monitoring. The Inter-laboratory Comparison samples for wet samples, dry samples, inland aquatic environment samples and soil samples were prepared.

The wet samples contain all of the mandatory items, however, because of the technical aspect, only the limited kinds of the mandatory items are added to the dry samples.

And, the extraction procedure for the dry samples is not standardized, so the variation seemed to be caused by the difference of the extraction efficiency was observed.

Throughout the implementation of Inter-laboratory Comparison project, the submitted data was evaluated by using the prepared values which was specified by the Network Center. Using this prepared value, the bias was observed on the statistic data. The possible likelihood value which should be used for the evaluation can be the median of all of the compiled data.

Items for the improvement of the quality of monitoring data in EANET are listed below:

1) Selection of the monitoring sites;
   The site selection should be done in accordance with the technical manuals. All of the site shall be prepared their regional scale, local scale and on-site scale descriptions. And the sites shall represent the circumstances around the site in each classified region, such as remote,

rural or urban sites. The existing monitoring sites should be reviewed their suitability to the requirements.

2) Establishment of the common point of view to the methodology including sampling and measurement;
3) Implementation of the Inter-laboratory Comparison project;
4) Implementation of the technical mission;
5) Establishment of quality management, and;
6) Reporting of the monitoring data.

2.6 References

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Chapter 3. Wet Deposition in East Asia

3.1 Introduction

The fate of an atmospheric chemical species is determined by a series of processes: 1) emission into the atmosphere, 2) transportation and transformation and 3) deposition to the earth's surface. The deposition of the species from the atmosphere to the surface is the end result of a complex chain of physical and chemical processes in the atmosphere, and is an input to another complex chain of processes that takes place in terrestrial and aquatic ecosystems. Atmospheric deposition has traditionally been separated into two distinct mechanisms, dry deposition and wet deposition. Dry deposition, which is more elaborated in the next chapter, is essentially eddy diffusional transport of gaseous and particulate species to the surface through molecular diffusion to the immediate vicinity of the surface and eventual absorption and/or adsorption onto the surface. Wet deposition, on the other hand, is incorporation of atmospheric species into water droplets with subsequent deposition onto the earth's surface. Droplets in the forms of cloud, rain, snow, or fog will be generally more or less acidified, which is the reason why the wet deposition is commonly known as acid rain or acid precipitation among the general public when this environmental issue was recognized in Europe.

Acid is a group of compounds that will release its hydrogen atom(s) as hydrogen ion(s) when dissolved into water, which suggests acid is intrinsically water soluble and therefore is deeply involved in atmospheric precipitation processes. Most of the acids in the atmosphere are not emitted directly from sources but chemically formed during the course of the transport. Major acids include sulphuric and nitric acids derived from sulphur and nitrogen oxides. Atmospheric acids thus formed acidify atmospheric water droplets including rain, where the acidity is commonly expressed in pH. The acidity of the solution will be decreased more or less, which would allow us to estimate acidification of atmospheric water by pH. Another role of atmospheric acids is to transport basic compounds by incorporating the bases through acid-base reactions. For example, during atmospheric transport of sulphuric and nitric acids, gaseous ammonia is collected to form ammonium salts, which enables the ammonia to be transported over longer distances. These precipitation acids eventually deposit onto earth's surface, which could impact ecological elements like water, soil, vegetation, and materials by acidification, and at least perturb acid-base balance of the elements.

It should be noted, however, precipitation pH is not a conserved quantity, pH of precipitation, as well as that of other aqueous solutions, is determined by the nature and relative proportion of acids and bases in solution. In another words, low pH precipitation is associated with high concentration of acids and/or low concentration of bases and also with smaller amounts of precipitation, the volume of water that dissolves the acids and bases.

Another point to make for scientifically sound interpretation is the ions dissolved in precipitation are not independent species at all but they are released from acids, bases, and salts: cations and anions are always associated with each other. An example is that calcium ion could be associated with carbonate and sulphate ions. In the case of sulphate, the solution is completely neutral, whereas the precipitation is slightly basic. And calcium ion is not basic or acidic at all, because this ion does not contain any potential species of H^+ or OH^- to release in the ion itself, although the ion would be a good measure of calcium carbonate in most cases.

In precipitation chemistry, the following acid-base chemistry will explain the acidity.

$$H_2SO_4 \longrightarrow 2H^+ + SO_4^{2-} \tag{1}$$

$$HNO_3 \longrightarrow H^+ + NO_3^- \tag{2}$$

$$NH_3 + H_2O \longrightarrow NH_3 \cdot H_2O \tag{3}$$

$$NH_3 \cdot H_2O \longrightarrow NH_4^+ + OH^- \tag{4}$$

$$CaCO_3 \longrightarrow Ca^{2+} + CO_3^{2-} \tag{5}$$

$$CO_3^{2-} + H_2O \longrightarrow HCO_3^- + OH^- \tag{6}$$

$$H^+ + OH^- \rightleftarrows H_2O \tag{7}$$

The well known sulphuric and nitric acids are designated as strong acids because the hydrogen atom is released into water as ions, equations. (1) and (2). Atmospheric strong acid includes hydrochloric and hydrofluoric acids. Some acids do not release all the hydrogen than can be released from the compound while the released fraction is controlled by some chemical constants. Such acids are called weak acids, which include sulphurous and nitrous acids and a large group of organic acids. Organic acids also play an important role in controlling precipitation acidity, particularly in areas sufficiently distant from anthropogenic sulphur and nitrogen sources.

Unlike acids, atmospheric bases are all weak bases. Ammonia is dissolved into water to form hydrated ammonia, or ammonium hydroxide, equation (3), which will release some fraction of OH species as OH^- leaving NH_4^+. In the case of calcium carbonate, $CaCO_3$, the chemistry is a little complicated. Because Ca species is very stable in the form of Ca^{2+} in aqueous solution, Ca^{2+} is released from $CaCO_3$ leaving CO_3^{2-}, eq. (5). While no further changes occur for Ca^{2+} as mentioned above, CO_3^{2-} is too instable to form HCO_3^- by reacting with water (hydrolysis), eq. (6), which makes eventually the $CaCO_3$ solution basic. These acids and bases are balanced by neutralization through the reaction, equation (7) with a constraint of $K_w = [H^+][OH^-]$.

3.2 Datasets and Data Quality

The present analysis was applied to volume-weighted annual mean concentrations of the analytical ion suite in order to understand the general regional picture of acid deposition. For this purpose, all available annual means since 2000 were subjected to some data screening only to include the annual mean sets meeting the data completeness criteria for the nine ions including H^+ as pH. Then, all sites that successfully produced at least one full year measurements for all nine analytical sites were selected as shown in Table 3.2.1 covering 57 sites in all of the 13 countries.

Further, another type of screening was carried out for the last five years of the operation, 2005-2009, to provide some average picture of the recent wet deposition. Forty-two out of the 57 sites meet the condition of which annual measurements covered all the five years. For three countries, Cambodia, Lao PDR, and Myanmar, they have intrinsically less than five-year operations because of their time of entry for EANET, and four- or three-year averages for Cambodia and Lao PDR, and Myanmar, respectively, were employed for the five-year averages.

With respect to the unit for chemical species, molar unit is applied in the EANET program because the SI and IUPAC approve this unit system. In this chapter, however, equivalent unit is employed because the equivalent concentration is practically based on a hydrogen ion, H^+, and the quality in this unit is easily converted to molar concentration by multiplying the number of electric charge of an ionic species of interest.

Table 3.2.1. The annual mean sets of data completeness criteria for ions of all EANET participating countries.

Number of years	Country	Site	RF	pH	μeq L^{-1}												C	A	R1
					H^+	NH_4^+	Ca^{2+}	K^+	Mg^{2+}	Na^+	NO_3^-	SO_4^{2-}	Cl^-	nss-SO_4^{2-}	nss-Ca^{2+}	nss-Cl^-			
4	Cambodia	Phnom Penh	1370.7	6.14	0.7	15.1	44.5	5.0	5.1	7.5	9.3	26.5	9.6	24.0	44.2	0.9	78.0	45.4	26.4
5	China	Hongwen	1369.2	4.71	19.6	34.8	62.6	3.6	12.5	32.8	29.2	85.0	46.0	81.2	61.2	7.7	165.9	160.1	1.8
		Jinyunshan	1322.6	4.49	32.3	100.3	135.7	9.4	7.8	9.3	41.3	222.8	13.5	221.7	135.3	2.7	294.8	277.6	3.0
		Jiwozi	352.3	6.20	0.6	41.3	350.2	22.1	81.9	55.0	47.7	242.2	33.1	235.5	347.8	-30.9	551.0	323.0	26.1
		Shizhan	499.1	5.68	2.1	156.9	320.8	15.2	45.2	22.2	54.2	358.1	33.8	355.4	319.8	7.8	562.4	446.1	11.5
		Xiang Zhou	1794.8	4.79	16.3	37.3	13.8	4.1	11.2	34.8	16.5	41.7	42.2	37.6	12.4	1.6	117.5	100.3	7.9
		Xiaoping	1890.2	4.66	22.0	33.6	24.2	7.4	4.8	16.8	21.2	46.2	17.2	44.5	23.5	-2.5	108.9	84.6	12.6
5	Indonesia	Bandung	2009.3	5.11	7.8	45.0	26.5	3.5	6.6	10.9	21.5	58.2	10.9	56.9	26.0	-1.8	100.3	90.7	5.0
		Jakarta	1810.2	4.48	33.2	19.4	27.0	6.4	6.9	14.9	28.8	56.1	21.0	54.3	26.3	3.7	107.8	106.0	0.8
		Kototabang	2270.1	4.82	15.2	9.6	9.4	6.3	3.7	6.5	2.2	10.7	9.0	9.9	9.2	1.4	50.7	21.8	39.8
		Serpong	1631.1	4.62	24.0	44.2	15.2	3.1	5.6	10.7	30.1	56.7	15.5	55.4	14.8	3.0	102.8	102.3	0.3
5	Japan	Banryu	1505.2	4.58	26.4	18.4	14.5	2.8	19.8	81.3	23.1	48.4	93.8	38.6	11.0	-1.0	163.2	165.4	-0.7
		Happo	2415.5	4.87	13.6	10.1	8.6	1.0	1.9	4.9	9.3	22.3	6.3	21.7	8.4	0.7	40.1	37.9	2.8
		Hedo	2056.2	4.98	10.5	8.2	14.1	4.9	43.1	193.8	7.6	37.5	215.5	15.5	5.8	-10.4	274.6	260.6	2.6
		Ijira	2557.4	4.53	29.7	20.0	5.7	1.0	3.8	14.6	22.4	36.8	18.0	35.0	5.1	1.0	74.8	77.2	-1.6
		Ochiishi	999.0	4.88	13.1	9.3	14.3	5.0	45.6	204.3	8.4	41.9	232.6	17.5	5.6	-5.6	291.6	282.9	1.5
		Ogasawara	1710.0	4.98	10.6	4.7	6.9	2.9	25.9	113.7	3.9	23.7	133.2	10.1	2.1	0.6	164.7	160.8	1.2
		Oki	1220.4	4.64	22.7	15.3	27.7	10.6	83.7	361.6	20.6	78.1	414.7	34.5	12.2	-6.9	521.6	513.4	0.8
		Rishiri	1004.0	4.71	19.6	20.3	14.8	4.7	39.7	170.0	14.8	53.0	190.1	32.5	7.6	-8.1	269.0	257.9	2.1
		Sado-seki	1125.6	4.60	24.8	18.9	22.2	6.6	57.7	254.3	20.8	64.4	288.9	33.9	11.3	-7.5	384.6	374.1	1.4
		Tappi	1084.1	4.63	23.5	15.8	20.0	5.5	50.6	225.7	19.1	57.8	254.0	30.9	10.4	-9.1	341.2	330.9	1.5
		Yusuhara	2422.3	4.75	18.0	7.9	4.3	1.0	3.7	14.2	8.1	21.3	16.8	19.6	3.7	0.2	49.1	46.2	3.0
5	Korea	Jeju (Kosan)	990.4	4.56	27.5	31.5	17.9	8.2	18.1	90.6	21.9	61.1	100.7	50.1	14.1	-4.9	193.8	183.7	2.7
		Imsil	1044.7	5.27	5.4	36.9	17.2	24.2	6.0	28.8	23.0	47.9	43.4	44.4	16.0	9.9	118.5	114.3	1.8
		Ganghwa	1119.6	4.53	29.8	45.8	21.6	7.1	9.8	38.5	32.0	69.6	50.5	65.0	20.0	5.5	152.7	152.0	0.2
4	Lao PDR	Vientian	989.2	5.86	1.4	9.6	10.6	2.0	2.9	3.8	6.6	9.2	6.9	8.7	10.5	2.5	30.3	22.7	14.3
5	Malaysia	Danum Valley	2728.0	5.12	7.5	3.9	1.7	1.5	1.0	4.7	1.5	5.8	6.2	5.2	1.5	0.7	20.3	13.5	20.1
		Petaling Jaya	3318.5	4.42	38.0	18.6	8.9	1.5	1.2	4.6	34.7	35.6	7.3	35.1	8.7	2.0	72.8	77.6	-3.2
		Tanah Rata	3128.1	4.98	10.4	3.7	2.3	1.0	0.8	2.4	5.0	7.9	2.0	7.6	2.2	-0.8	20.6	14.9	16.1
5	Mongolia	Terelj	192.6	5.31	5.0	44.5	39.1	6.2	7.4	9.6	16.3	40.0	9.7	38.7	38.7	-1.5	111.8	66.0	25.7
		Ulaanbaatar	169.4	6.19	0.6	56.8	88.7	3.3	9.4	5.9	20.1	52.7	8.9	51.8	88.5	2.1	164.8	81.8	33.7
1	Myanmar	Yangon	3010.4	6.46	0.3	16.2	19.3	6.5	5.7	18.5	7.2	16.0	25.0	13.8	18.5	3.5	66.5	48.3	15.9
5	Philippines	Los Banos	2351.2	5.33	4.7	15.6	10.0	4.7	7.1	18.6	7.8	21.3	22.0	19.1	9.2	0.3	60.7	51.0	8.6
		Metro Manila	2948.0	5.22	6.0	34.4	20.3	7.5	7.3	18.4	14.2	38.2	22.9	36.0	19.5	1.4	93.8	75.2	11.0
5	Russia	Irkutsk	446.9	5.00	9.9	33.3	54.6	10.8	12.5	11.1	17.1	63.9	15.8	62.5	54.1	2.9	132.2	96.8	15.5
		Listvyanka	374.6	4.72	19.3	16.2	18.4	1.9	3.8	2.9	15.7	37.4	4.4	37.1	18.3	1.0	62.4	57.5	4.1
		Mondy	274.3	5.38	4.2	14.1	11.8	1.8	2.7	2.1	6.6	13.3	4.5	13.1	11.7	2.1	36.6	24.4	20.1
		Primorskaya	753.1	4.89	12.8	38.2	34.9	6.2	9.6	18.5	23.2	68.6	19.2	66.4	34.1	-2.4	120.3	111.0	4.0
5	Thailand	Bangkok	1804.4	5.07	8.5	55.1	30.5	2.1	4.7	9.8	28.5	35.5	10.1	34.3	30.1	-1.4	110.7	74.0	19.9
		Chiang Mai	1225.1	5.61	2.4	18.9	11.2	1.9	3.7	2.5	8.4	10.3	4.5	10.0	11.1	1.6	40.6	23.2	27.4
		Kanchanaburi	2045.0	5.67	2.1	14.6	7.0	1.4	2.4	7.8	4.4	6.8	7.9	5.9	6.7	-1.2	35.5	19.1	30.1
		Pathumthani	1448.1	4.94	11.5	32.3	20.6	1.5	3.0	6.3	20.6	23.3	6.7	22.5	20.3	-0.7	75.2	50.6	19.6
		Samutprakan	1727.8	5.38	4.1	46.2	24.9	4.1	6.9	14.2	17.4	33.6	12.6	31.9	24.3	-4.0	100.4	63.6	22.5
5	Vietnam	Hanoi	1705.4	5.77	1.7	54.1	38.7	3.2	11.4	11.5	18.7	68.9	14.3	67.6	38.2	0.9	120.6	102.0	8.4
		Hoa Binh	1821.7	5.37	4.3	25.1	26.3	2.9	6.3	4.5	12.4	35.3	7.6	34.8	26.1	2.3	69.4	55.2	11.4

3.3 pH

The state of acid deposition will be assessed in terms of pH in this chapter. In the discussion of “acid rain”, pH of rainwater is a focus among the general public. A description of pH is provided as the starting point of understanding of the wet deposition. It should be noted that interpretation of pH might not be so straight forward, of which chemistry will be addressed in more detail in section 3.6.

The regional pHs were assessed in terms of a histogram for the annual mean pHs for the entire period from 2000 through 2009 (Figure 3.3.1). The pH ranged from 4.18 to 6.94 with an arithmetic mean of 5.07. The histogram had a tail to higher pHs with a median of pH 4.94. The pHs below 4.30 were: 4,18 (Jinyunshan, 2001), 4.20 (Haifu, 2008), 4.22 (Nansan, 2000, Haifu, 2009), 4.23 (Petaling Jaya, 2002), 4.25 (Ganghwa, 2005), 4.25 and 4.28 (Petaling Jaya, 2001 and 2003), respectively.
In contrast, the pH over 6.5 were 6.94 (Jiwozi, 2008), 6.85 (Jiwozi, 2007), 6.72 and 6.69 (Phnom Penh, 2006 and 2007, respectively), and 6.53 (Shuzhan, 2009).

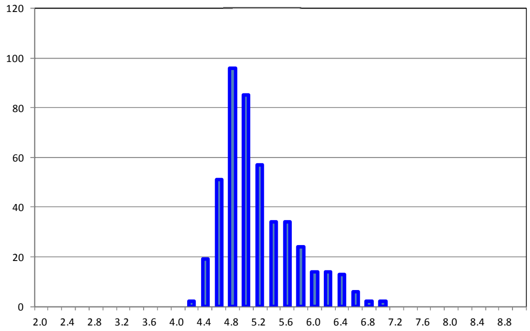


Figure 3.3.1. Histogram of annual mean pH for 2000-2009.

The pH was also assessed on basis of the last five-year operation. The top two pHs (6.20 and 6.19 for Jiwozi and Ulaanbaatar, respectively) were exceptionally high for the distribution, and the remaining pHs were smoothly decreasing to the minimum pH, 4.42 for Petaling Jaya (Figure 3.3.2).

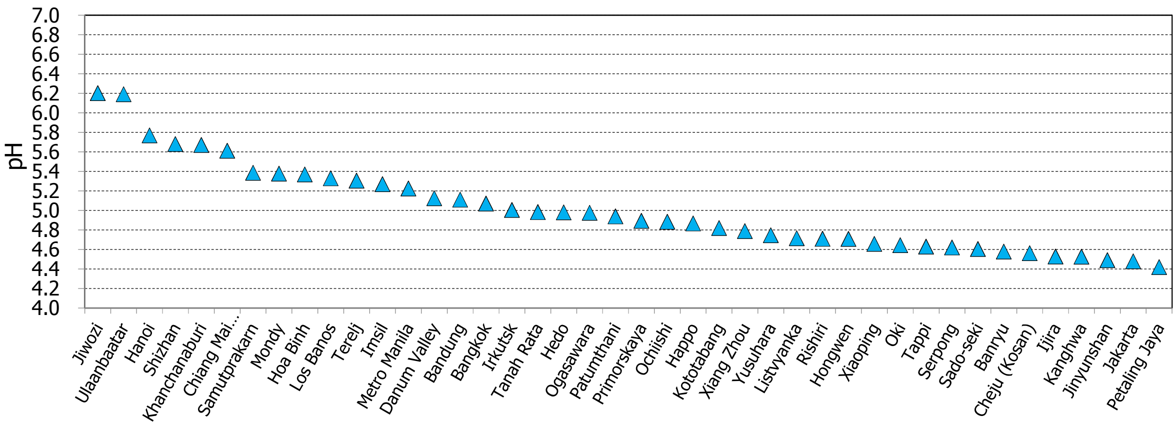


Figure 3.3.2. Five-year average for the annual mean pHs (2005 – 2009)

It is difficult to set a scientifically reasonable criterion of “acid rain” in terms of pH although “pH 5.6”, seems to be socially accepted as the standard based on the gas-water equilibrium of an acidic gas, carbon dioxide, in the atmosphere. However, natural air free from anthropogenic pollution contains other acid gases like sulphur dioxide due to volcanic emissions and oxidation of maritime dimethylsulfide, $(CH_3)_2S$. Moreover, some bases are derived from natural sources including soil and animals.

In spite of these facts, it could be useful to set a pH point for preliminary evaluation of the observations. In this chapter, a pH of 5.0 is proposed as a guide for that purpose because the value is an integer. Under this assumption, 26 sites out of the 42 sites (62%) recorded pHs lower than this guide value. No sites covered pH equal to or less than 4.0. The lowest three pHs were less than pH 4.5.

Although some more chemistry should be explored for more detailed discussion, it suffices here to mention that precipitation in East Asia is significantly acidic.

3.4 Concentrations of Major Ions

Similar illustrations were applied to the concentrations of major ions involved in the acid-base chemistry.

Before entering into the discussion of sulphate and calcium ions, it is worthwhile to address the non-seasalt fraction concept. Precipitation incorporates seasalt particles containing seawater components including sulphate and calcium ions. Therefore, the seasalt fractions should be discriminated from the non-seasalt ions. Non-seasalt fractions of sulphate and calcium ions were usually estimated with equations (8) and (9), respectively, on the basis of sodium ion as a tracer of seawater under the assumption that the relative composition of these seasalt ions are conserved throughout the atmospheric processes (EANET Manual for Wet Deposition).

$$[\text{nss-}SO_4^{2-}] = [SO_4^{2-}] - ([SO_4^{2-}] / [Na^+])_{\text{seawater}} \times [Na^+] \qquad (8),$$

$$[\text{nss-}Ca^{2+}] = [Ca^{2+}] - ([Ca^{2+}] / [Na^+])_{\text{seawater}} \times [Na^+] \qquad (9).$$

where $([SO_4^{2-}] / [Na^+])_{\text{seawater}}$ and $([Ca^{2+}] / [Na^+])_{\text{seawater}}$ are the equivalent concentration ratios of seawater components and numerically 0.1206 and 0.0432, respectively. In this manner, sulphate derived from sulphuric acid converted from SO_2 (nss-SO_4^{2-}) in precipitation could be differentiated from that of natural seasalt (ss-SO_4^{2-}) while nss-Ca^{2+} would be due to soil-derived $CaCO_3$ being distinguished from Ca^{2+} ion in the seawater.

Concentration maps were produced for the mean annual concentrations of major ions for 2005-2009 (Figures. 3.4.1 to 3.4.5). With this map in reference, the discussion will apply to each of the major ions.

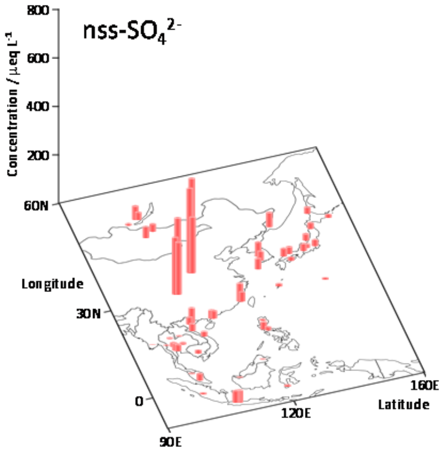


Figure 3.4.1. Distribution of five-year average concentrations of nss-SO$_4^{2-}$ ion.

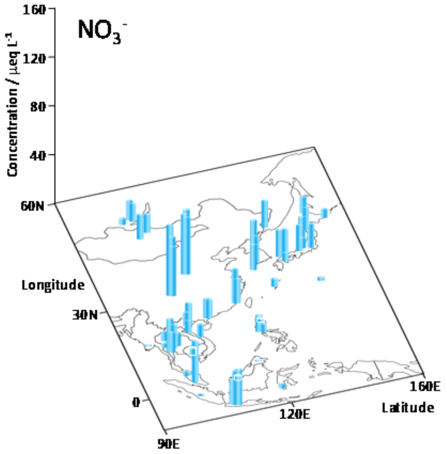


Figure 3.4.2. Distribution of five-year average concentrations of NO$_3^-$ ion.

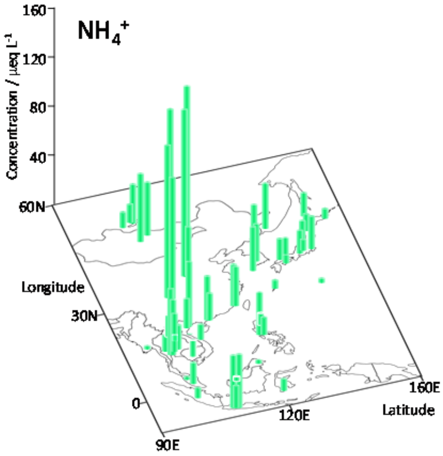


Figure 3.4.3. Distribution of five-year average concentrations of NH$_4^+$ ion.

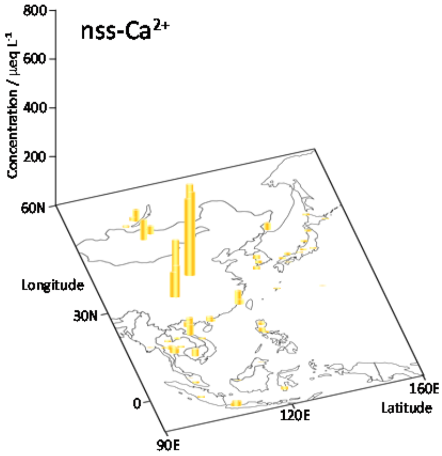


Figure 3.4.4. Distribution of five-year average concentrations of nss-Ca^{2+} ion.

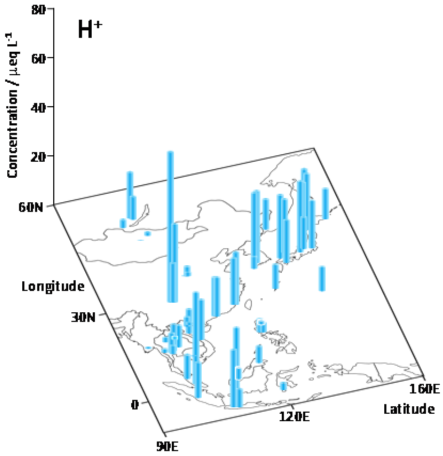


Figure 3.4.5. Distribution of five-year average concentrations of H$^+$ ion.

Figure 3.4.6 depicts a graph for the annual mean concentrations of nss-SO_4^{2-} for 2000 to 2009 at 57 sites in the 13 countries. The graph is produced for logarithmic concentration classes because the ionic species showed a broad range of concentration. One should note that the graph employs the logarithmic equivalent concentration, 1 μeq L^{-1} (or 1x10^{-6} eq L^{-1}) is transformed to “-6” in the abscissa.

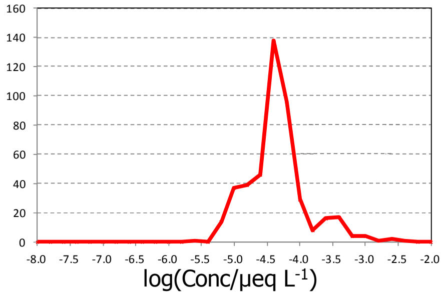


Figure 3.4.6. An annual mean concentration of nss-SO_4^{2-}.

The nss-SO_4^{2-} concentration is not smoothly distributed in the region. The graph was bimodal where one broad small peak at -3.5 of the abscissa and one large peak at -4.4 with a shoulder at 5.0. This pattern denotes that some sites are associated with unusually high concentration whereas the remaining sites have smoothly distributed concentrations although the lowest concentrations occur exclusively at some selected sites.

The concentration ranged from 2.4 μeq L^{-1} at Mt. Sto. Tomas (2006) to 3,000 μeq L^{-1} at Weishuyuan (2001). The high concentration over 1,000 μeq L^{-1} occurred in addition to this Weishuyuan concentration, 1,989, 1,670 and 1,068 μeq L^{-1} at Weishuyuan (2002) and Shizhan (2002 and 2000), respectively. Concentrations of 4.1, 4.1, 4.6, 4.8, and 4.9 μeq L^{-1} for Danum Valley (2007), Kanchanaburi (2007), Danum Valley (2008), Kanchanaburi (2005) and Danum Valley (2009) followed the minimum at Mt. Sto. Tomas.

The five year means were also produced for nss-SO_4^{2-} (Figure 3.4.7). Non-smooth distribution for the concentration is also discernible for this figure. Top three sits recorded unusually high concentration in the region.

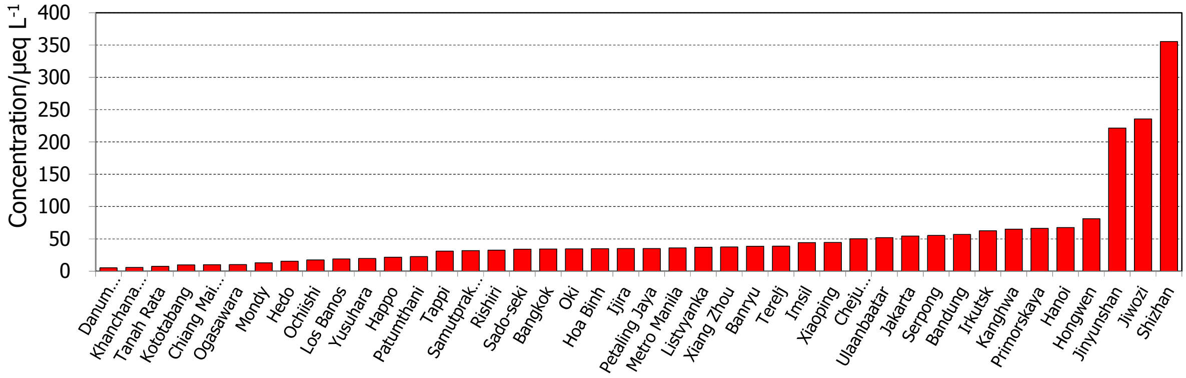


Figure 3.4.7. Five-year annual mean concentrations of nss-SO_4^{2-} (2005 – 2009)

For NO_3^-, a mono-modal pattern was noted with a peak at -4.6 of the abscissa with a very small shoulder at all at -5.1 of the abscissa (Figure 3.4.8). The highest six concentrations over 140 µeq L^{-1} occurred: 210, 168, 153, 150, and 140 µeq L^{-1} at Weishuyuan (2001, and 2002), Shizhan (2001, and 2002), and Jiwozi (2001). The lowest five were 1.0, 1.0, 1.4, 1.4, and 1.4 for Danum Valley (2007), Kototabang (2009), Danum Valley (2008), Kototabang (2006), and Danum Valley (2006), respectively.

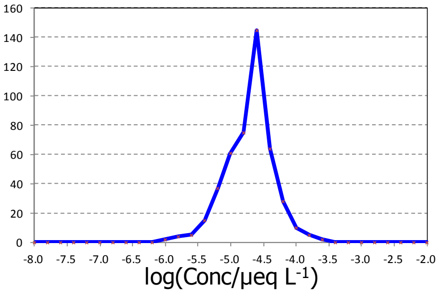


Figure 3.4.8. An annual mean concentration of NO_3^-.

The five year mean for NO_3^- is shown in Figure 3.4.9. Top three concentrations were significantly higher than the others and some lowest two or twelve could be very low in the NO_3^- concentration spectrum.

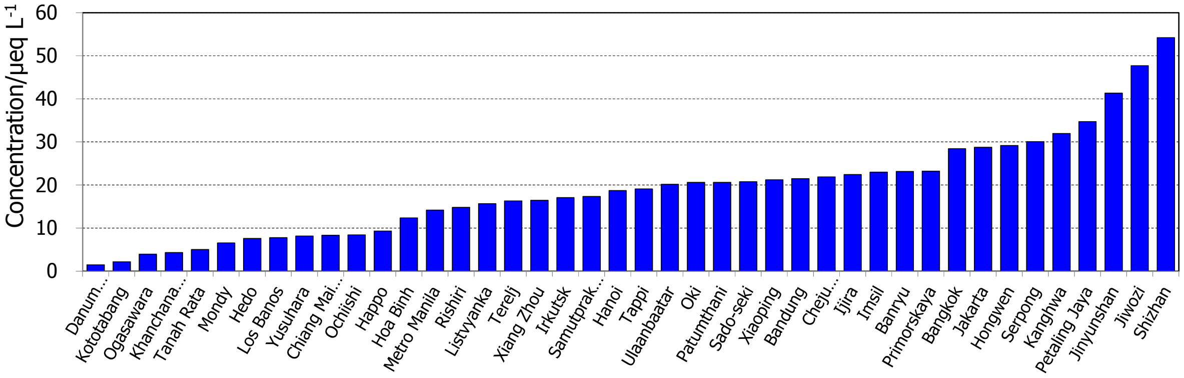


Figure 3.4.9. Five-year annual mean concentrations of NO_3^- (2005 – 2009)

Ammonium ion, NH_4^+, was explored in the same manner as nss-SO_4^{2-} and NO_3^-. The corresponding graph is displayed in Figure 3.4.10 where a somewhat skewed mono modal pattern is noted with some shoulders at both tails. The maximum concentration, 426 µeq L^{-1} took place at Weishuyuan (2000) followed by 421, 354, 319, and 255 µeq L^{-1} at Shizhan (2000), Weishuyuan (2001, 2002) and Shizhan (2001), respectively. The minimum, 1.0 µeq L^{-1} was recorded at Mt. Sto. Tomas (2006) and next minimums were 1.3, 1.8, 2.0 and 2.0 µeq L^{-1} at Danum Valley (2007), Kototabang (2003), and Danum Valley (2005 and 2006), respectively.

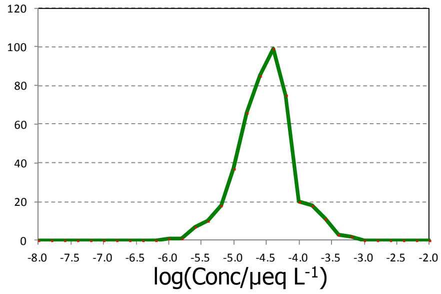


Figure 3.4.10. An annual mean concentration of NH_4^+.

Five year means were summarized in Figure 3.4.11 for ammonium ion. Top two, Shizhan and Jinyunshan concentration had exceptionally high in the EANET region.

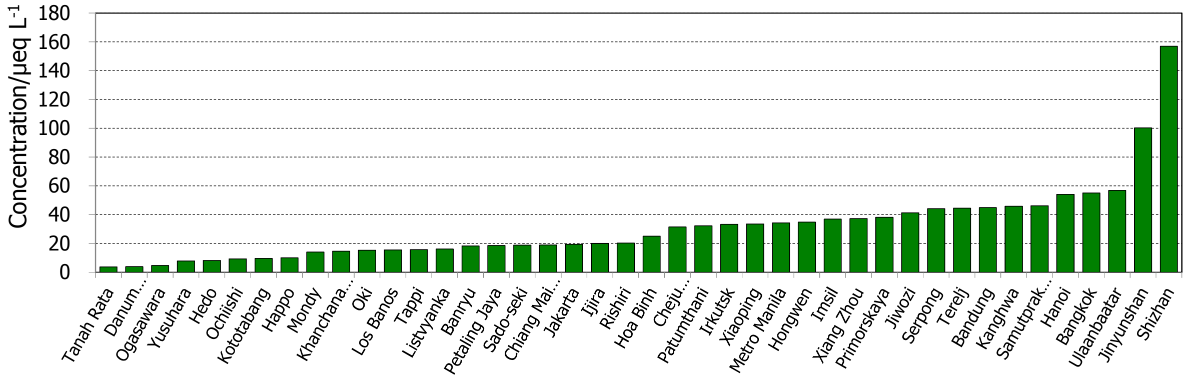


Figure 3.4.11. Five-year annual mean concentrations of NH_4^+ (2005 – 2009)

Non-seasalt calcium, nss-Ca^{2+} was subjected to the graph analysis (Figure 3.4.12). Concentration of over 500 μeq L^{-1} were observed in three sites; 1334, 948, 514, 600 at Shizhan (201, 2002, 2003, 2006), 606, 2300, 1224, 656 at Weishuiyuan (2000, 2001, 2002, 2006), 850, 546, 690, 528 at Jiwozi (2001, 2002, 2007, 2008). The lowest five happened at three sites: 1.0, 1.3, 1.8, 2.0, and 2.0μeq L^{-1} for Mt. Sto. Tomas. (2006), Danum Valley (2007), Kototabang (2003) and Danum Valley (2005 and 2006). For nss-Ca^{2+} of the five year covering 2005 to 2009, the top three sites, Jiwozi, Shizhan, and Jinyunshan, the concentrations were really prominent in the five-year averages (Figure 3.4.13).

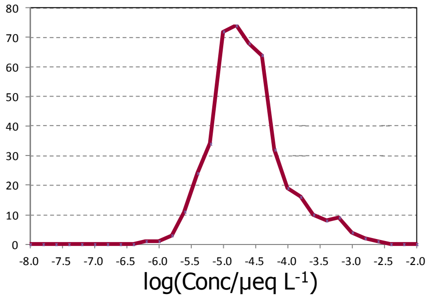


Figure 3.4.12. An annual mean nss-Ca^{2+} concentration.

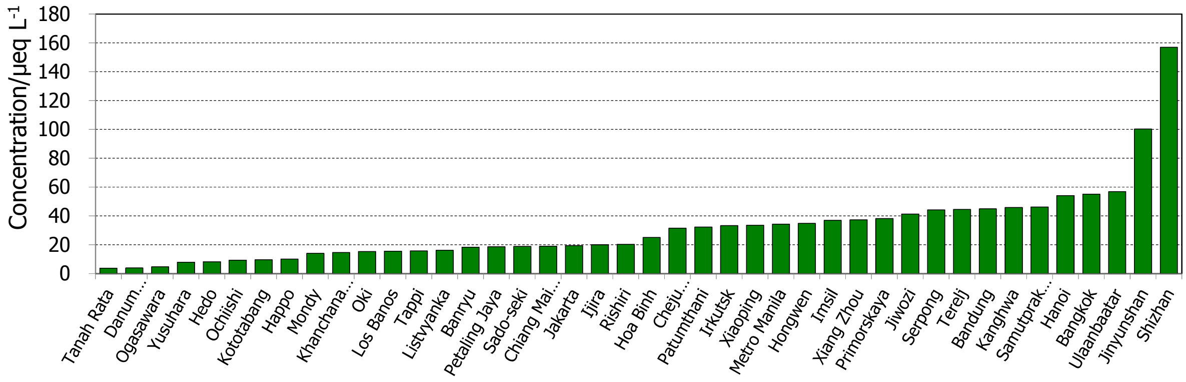


Figure 3.4.13. Five-year average annual mean nss-Ca^{2+}(2005 – 2009)

The concentration graph was extended to seasalt ion, Na$^+$ (Figure 3.4.14) and Cl$^-$(Figure 3.4.15). Both graphs are bimodal with a large peak centred at approximately -5.0 and -5.1, respectively, on the abscissa and a small peak at some -3.6 and -3.4, respectively. The diagrams for these species are generally similar to each other and small peaks would correspond to measurements in remote islands and sites close to the ocean.

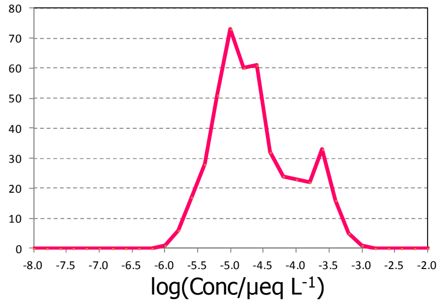


Figure 3.4.14. An annual mean Na$^+$ concentration.

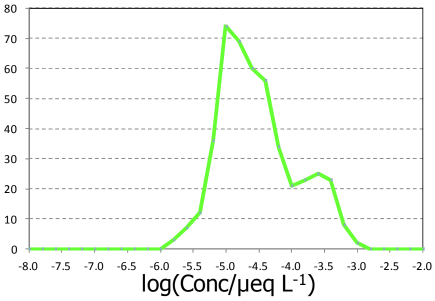


Figure 3.4.15. An annual mean Cl$^-$ concentration.

3.5 Composition

Ionic composition provides the chemical nature of the wet deposition. Ions in aqueous solution are derived directly from acids, bases, and salts. These compounds, however, are originally formed with acids and/or bases. The following is the significant original compounds associated with respective ions. Each of the analytical suits would divulge the history of the precipitation.

Hydrogen ion, H$^+$: This ion is derived from acids, and practically from sulphuric and nitric acids. Some organic acids like HCOOH and CH_3COOH are kept in mind as well as other mineral acids including HCL and HF. This ion easily react with OH$^-$ to form H_2O where the concentration product is constrained: $[H^+][OH^-] = K_w$.

Ammonium ion, NH_4^+: The ion is due to ammonia gas regardless some possible reactions with acids in the atmosphere. Ammonia has a broad range of sources: biomass burning, animal feedlots, solid waste, and automobiles.

Calcium ion, Ca^{2+}: This ion is derived from seasalt, soil and road dust. In most cases, the calcium ion is regarded to be from $CaCO_3$, a weakly basic compound.

Potassium ion, K^+: This ion is regarded as from K_2CO_3 which is formed from combustion of vegetation.

Magnesium ion, Mg^{2+}: This species has origins similar to calcium ion because the chemical behaviors of the two elements are very similar. Magnesium carbonate, $MgCO_3$ would be the original form of this ion.

Sodium ion, Na^+: This ion is interpreted to be exclusively from seasalt, and non-seasalt fractions of sulphate and calcium ions are evaluated on the basis of this species. Actually, however, solid waste facilities and wind-blown dust could be sources in some cases.

Nitrate, NO_3^-: Nitric acid, HNO_3, is the source of this ion.

Sulphate, SO_4^{2-}: This ionic species has two sources: 1) nss-sulphate ion from sulphuric acid, H_2SO_4 which is derived from SO_2, and 2) sea salt-sulphate in seawater. Note that this ion itself is neither acidic nor basic. If the sulphate is derived entirely from H_2SO_4, this ion is a good measure of sulphuric acid. However, if it is derived from $CaSO_4$, the solution is neutral and the sulphate ion never works as the measure of sulphuric acid. This kind of discussion generally applies to wet deposition chemistry.

Chloride, Cl^-: This ion is originated from seasalt. In some cases, however, hydrochloric acid is the source of this ion, which should be considered for sites under the influence of volcanic emission.

With the explanation, the ionic composition of wet deposition is displayed in the form of two bar charts representing cations and anions. Figures 3.5.1 to 3.5.43 are volume-weighted concentrations of the ion suite for individual sites for the period 2005-2009.

Taking the case of Banryu (Figure 3.5.18), some interpretations of the ionic composition will be addressed.

(1) Data quality: Agreement between the cations and anions should be assessed at first. Because the concentration is expressed in equivalent, the length of the respective bars is corresponding to the amount of electric charge. The ion balance will be checked with such a comparison of lengths.

(2) Degree of neutralization: Concentrations of nss-sulphate and nitrate are a good measure to estimate original concentration of sulphuric and nitric acids. If the sum of the two ions is compared with hydrogen ion, degree of neutralization will be evaluated. When the hydrogen ion concentration is very low compared with the sulphate and nitrate, the neutralization took place to a considerable extent. In other words, the original acidity, or input acidity, might be considerably high in this case.

(3) Relative contribution of nitrate and nss-sulphate: This shows the contributions of sulphuric and nitric acids to the precipitation acidity although a certain fraction has been usually neutralized.

(4) Relative contribution of ammonium and nss-calcium ions: Under the assumption in this section, which bases, ammonia or calcium carbonate, would be classified to the major base for the neutralization of the input acidity.

(5) Sodium and chloride ion concentrations: The relative concentration of these ions is associated with seasalt contribution. For sites on an islands or coastal areas, these ions compose a significant fraction of the ionic concentration. Banryu is the case, and island sites featured the seasalt contribution: e.g. Sado-Seki (Figure 3.5.15), Oki (Figure 3.5.18), Hedo (Figure 3.5.21) and Ogasawara (Figure 3.5.22) .

(6) Others: It should be noted that the above discussion is not always successful because some other sources might have contributed to the ions of interest.

In some cases, particularly the precipitation acidity is very low, hydrogen carbonate, HCO_3^-, has a visible contribution where as this ion is not included in the analytical suite. Other than HCO_3^-, hydrogen chloride, and organic acids could be some significant components of the precipitation ions.

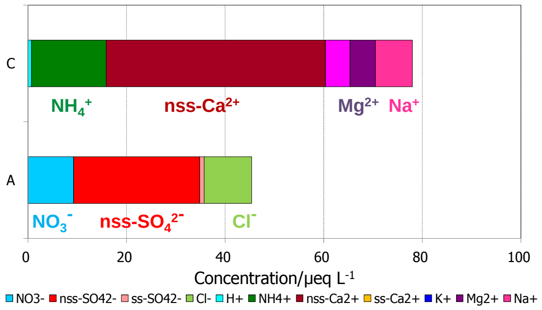


Figure 3.5.1. Ionic composition of wet deposition in Phnom Penh.

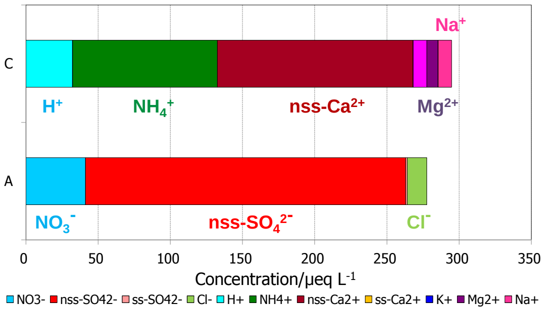


Figure 3.5.2. Ionic composition of wet deposition in Chongqing - Jinyunshan.

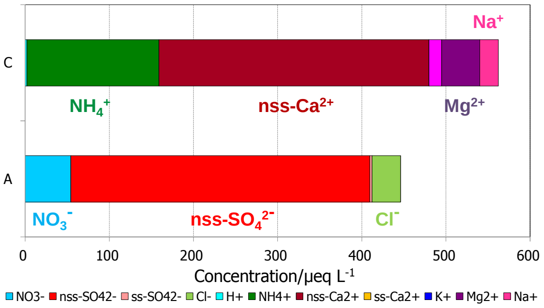


Figure 3.5.3. Ionic composition of wet deposition in Xi'an - Shizhan.

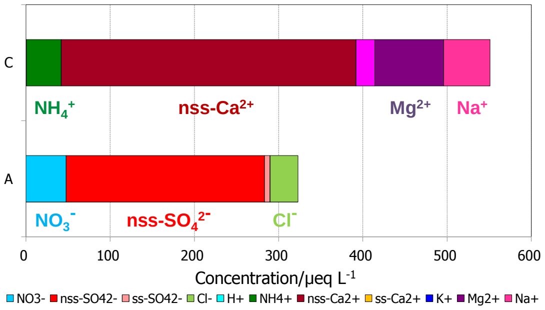


Figure 3.5.4. Ionic composition of wet deposition in Xi'an - Jiwozi.

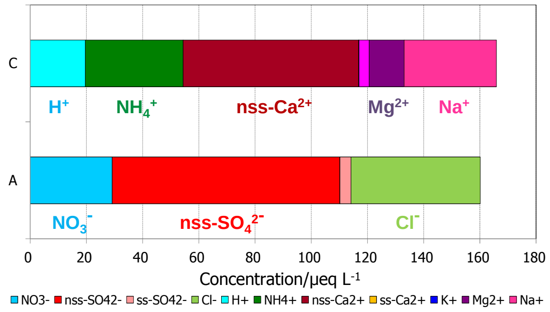


Figure 3.5.5. Ionic composition of wet deposition in Xiamen - Hongwen.

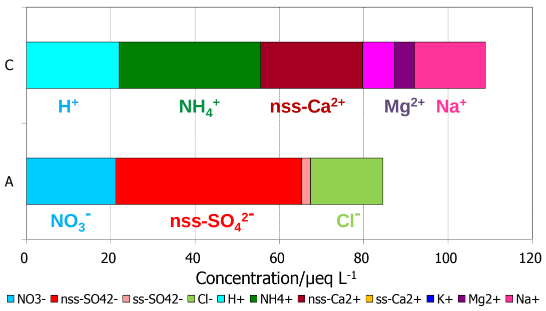


Figure 3.5.6. Ionic composition of wet deposition in Xiamen - Xiaoping.

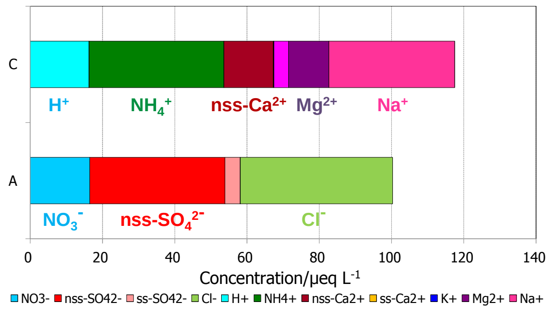


Figure 3.5.7. Ionic composition of wet deposition in Zhuhai - Xiang Zhou.

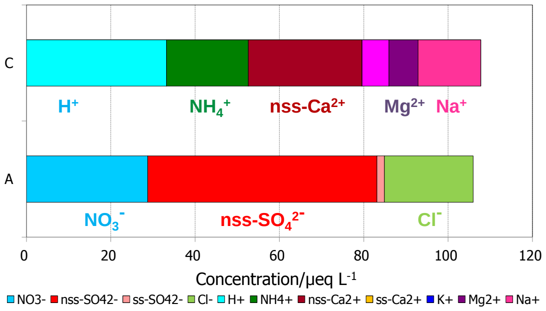


Figure 3.5.8. Ionic composition of wet deposition in Jakarta.

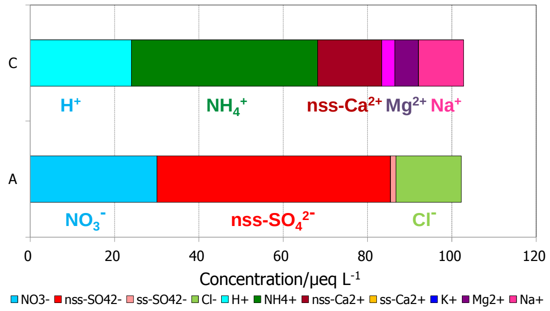


Figure 3.5.9. Ionic composition of wet deposition in Serpong.

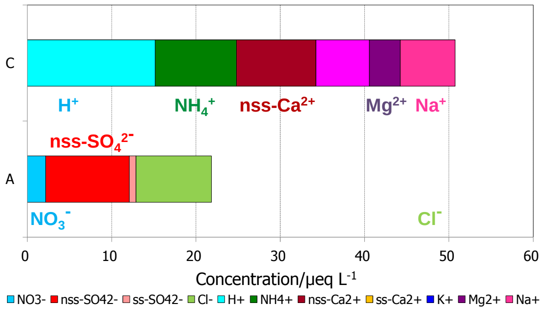


Figure 3.5.10. Ionic composition of wet deposition in Kototabang.

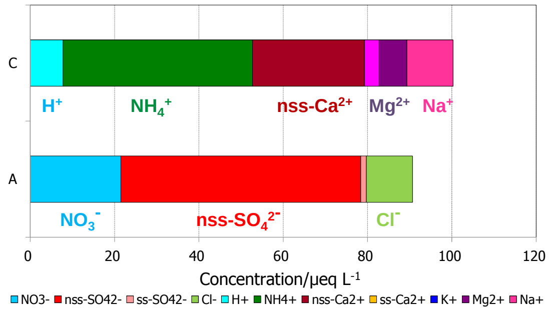


Figure 3.5.11. Ionic composition of wet deposition in Bandung.

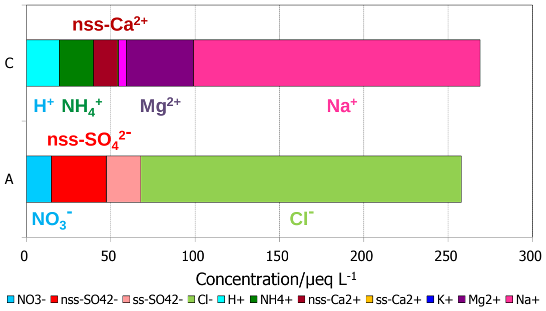


Figure 3.5.12. Ionic composition of wet deposition in Rishiri.

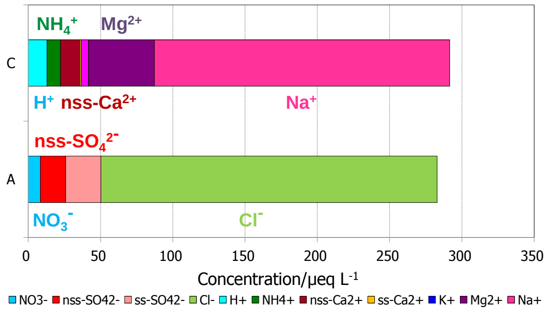


Figure 3.5.13. Ionic composition of wet deposition in Ochiishi.

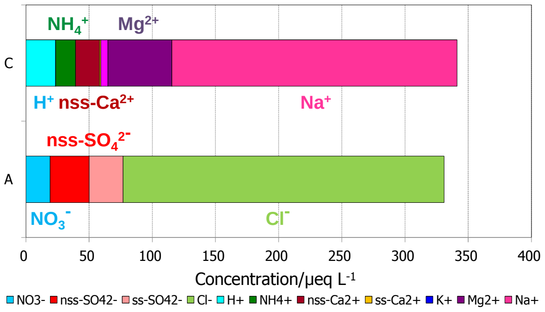


Figure 3.5.14. Ionic composition of wet deposition in Tappi.

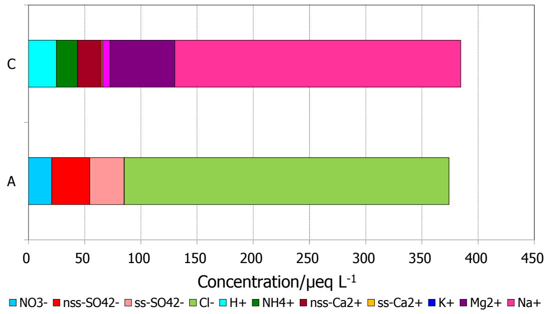


Figure 3.5.15. Ionic composition of wet deposition in Sado-Seki.

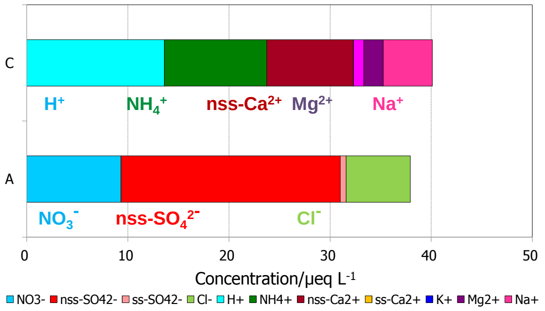


Figure 3.5.16. Ionic composition of wet deposition in Happo.

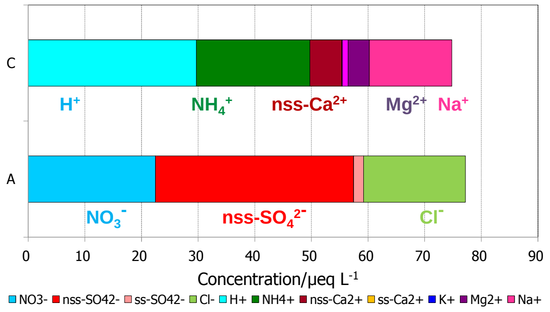


Figure 3.5.17. Ionic composition of wet deposition in Ijira.

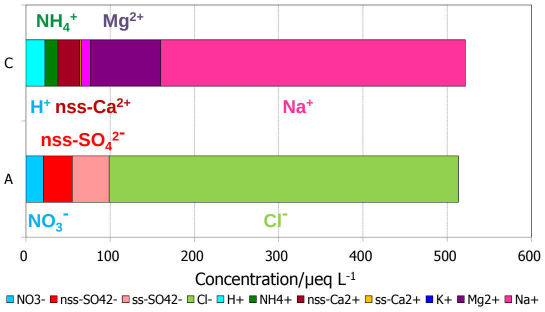


Figure 3.5.18. Ionic composition of wet deposition in Oki.

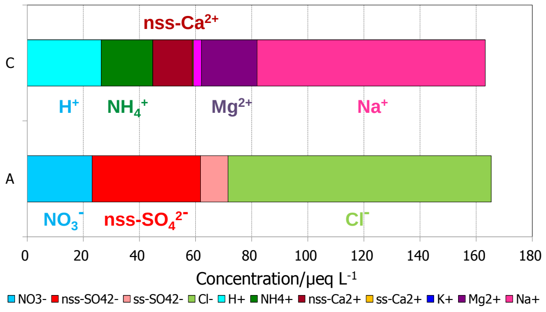


Figure 3.5.19. Ionic composition of wet deposition in Banryu.

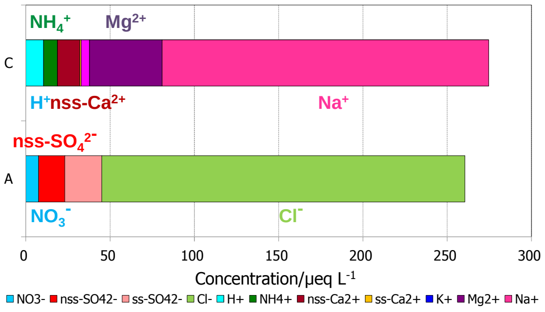


Figure 3.5.20. Ionic composition of wet deposition in Yusuhara.

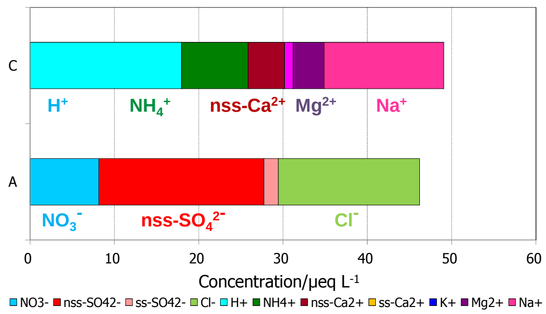


Figure 3.5.21. Ionic composition of wet deposition in Hedo.

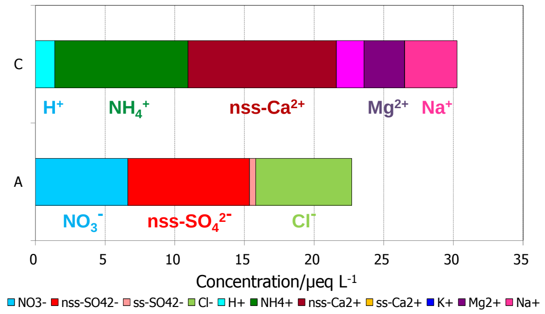


Figure 3.5.22. Ionic composition of wet deposition in Ogasawara.

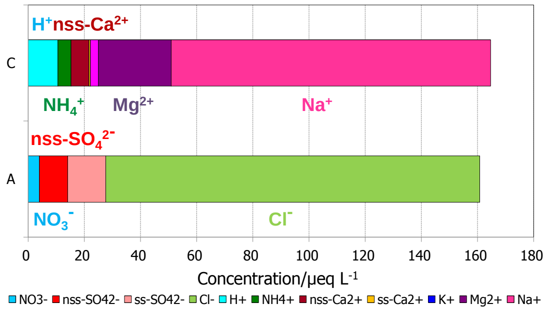


Figure 3.5.23. Ionic composition of wet deposition in Vientiane.

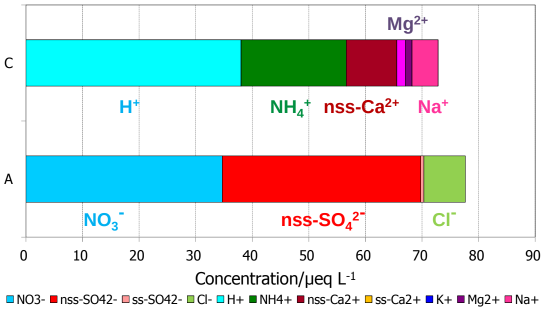


Figure 3.5.24. Ionic composition of wet deposition in Tanah Rata.

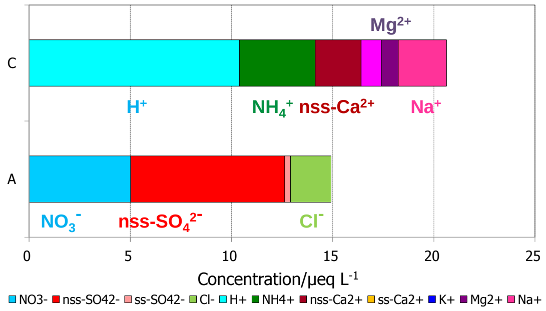


Figure 3.5.25. Ionic composition of wet deposition in Petaling Jaya.

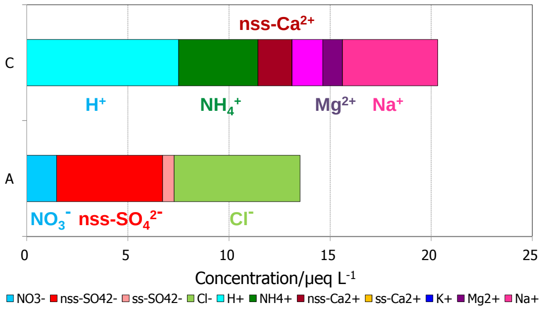


Figure 3.5.26. Ionic composition of wet deposition in Danum Valley.

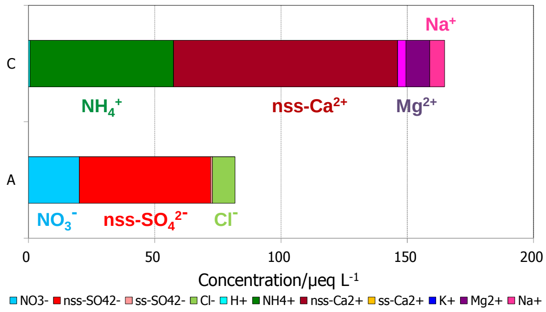


Figure 3.5.27. Ionic composition of wet deposition in Ulaanbaatar.

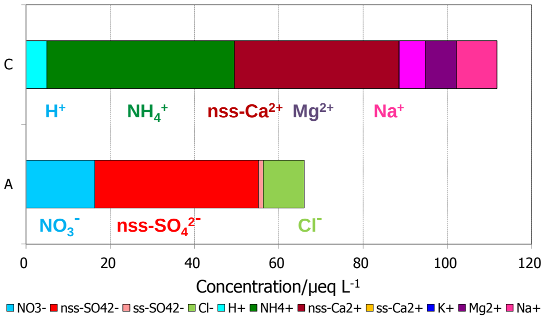


Figure 3.5.28. Ionic composition of wet deposition in Terelj.

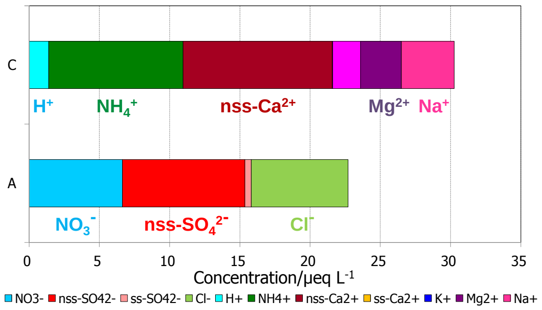


Figure 3.5.29. Ionic composition of wet deposition in Yangon.

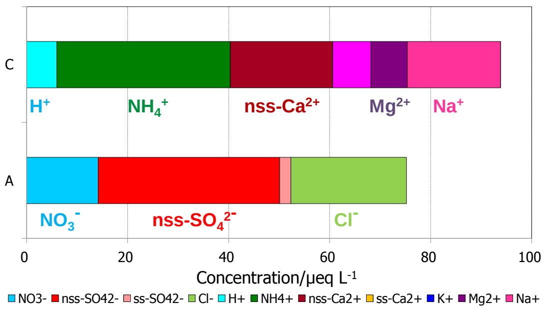


Figure 3.5.30. Ionic composition of wet deposition in Metro Manilla.

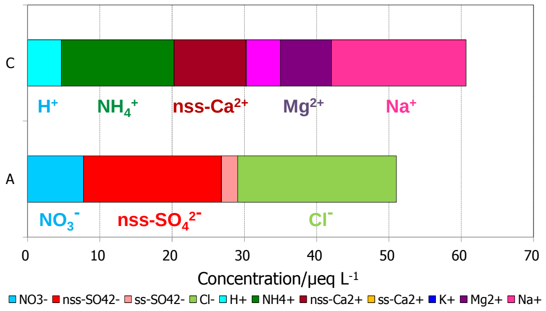


Figure 3.5.31. Ionic composition of wet deposition in Los Banos

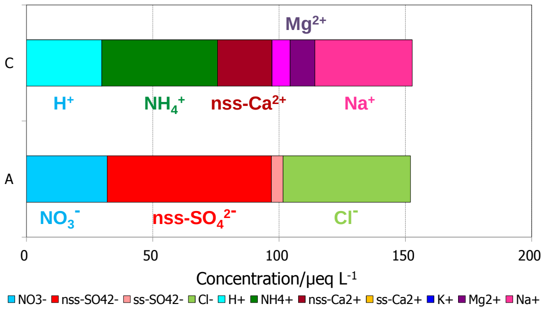


Figure 3.5.32. Ionic composition of wet deposition in Ganghwa.

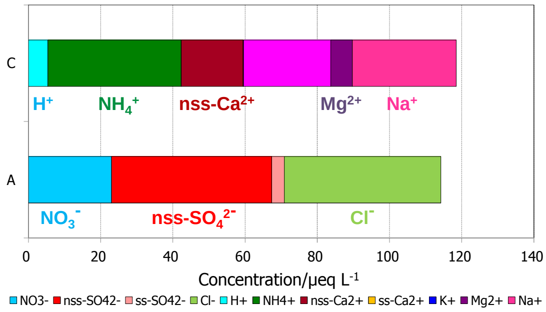


Figure 3.5.33. Ionic composition of wet deposition in Imsil.

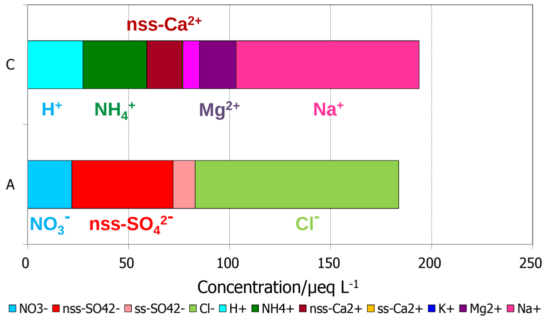


Figure 3.5.34. Ionic composition of wet deposition in Jeju.

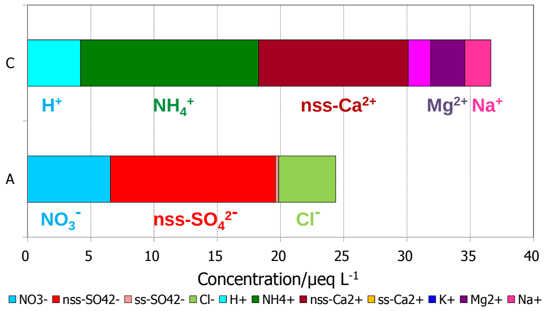


Figure 3.5.35. Ionic composition of wet deposition in Mondy.

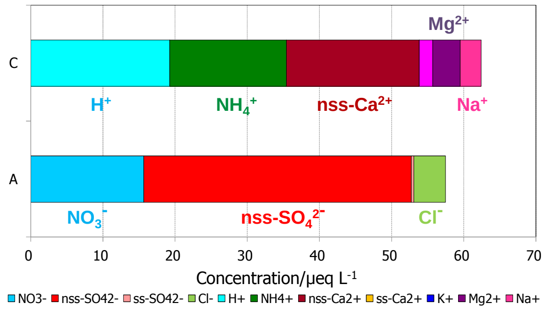
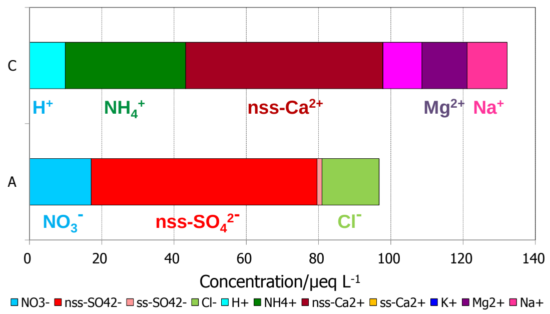


Figure 3.5.36. Ionic composition of wet deposition in Listvyanka.



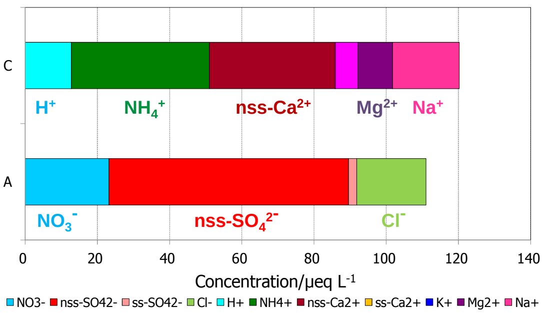
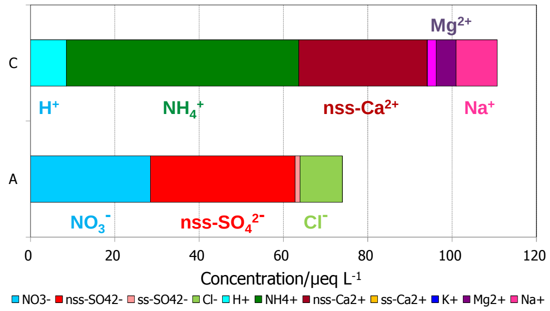


Figure 3.5.37. Ionic composition of wet deposition in Irkutsk.

Figure 3.5.38. Ionic composition of wet deposition in Primorskaya.



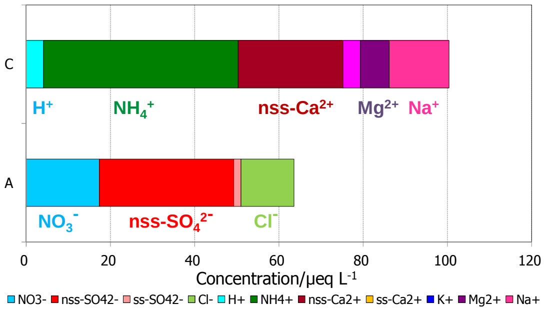
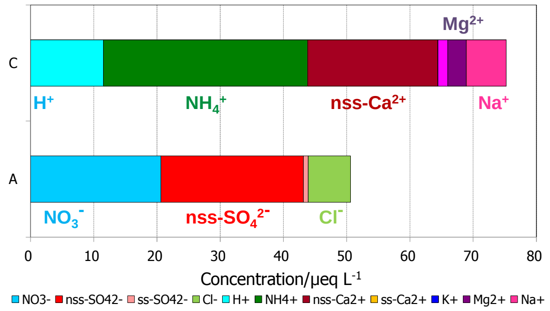


Figure 3.5.39. Ionic composition of wet deposition in Bangkok.

Figure 3.5.40. Ionic composition of wet deposition in Samutprakan.



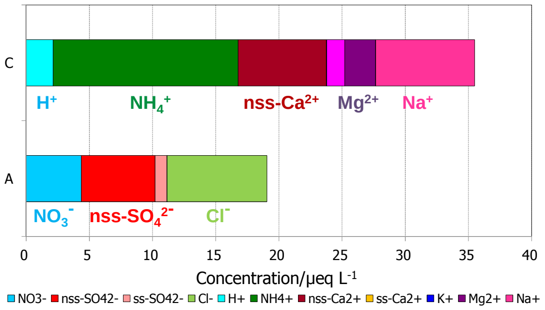
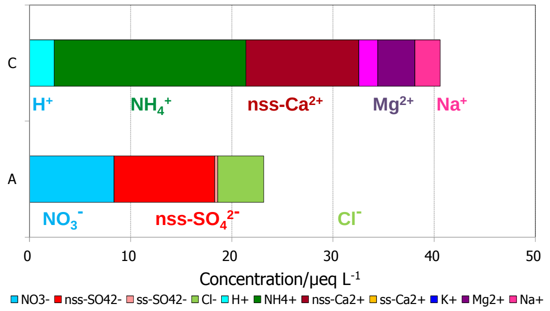


Figure 3.5.41. Ionic composition of wet deposition in Pathumthani.

Figure 3.5.42. Ionic composition of wet deposition in Kanchanaburi.



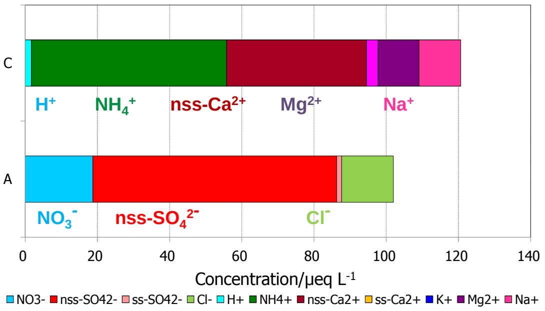


Figure 3.5.43. Ionic composition of wet deposition in Chaing Mai.

Figure 3.5.44. Ionic composition of wet deposition in Hanoi.

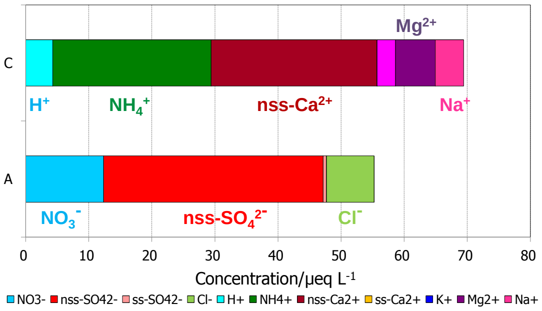


Figure 3. 5.45. Ionic composition of wet deposition in Hoa Binh.

3.6 pH and pA$_i$

pH should be properly interpreted on the basis that pH is determined by the nature and balance of acids and bases in the aqueous solution. Specifically, neutralization between acids and bases should be taken into consideration in a quantitative manner. For this purpose, pA$_i$ has been proposed as below in a similar manner to pH where two major acids, both sulphuric and nitric acids, are assumed to determine the input acidity (A$_i$) in order to draw a gross picture of acid-base chemistry.

$$pA_i = -\log\{[A_i]\} = -\log\{[\text{nss-}SO_4^{2-}]+[NO_3^-]\}$$
$$pH = -\log\{[H^+]\}$$

These two indices are complementary to each other: pH is an observable to show the acidity after neutralization whereas pA$_i$ is a conceptual parameter corresponding to the acidity before neutralization. If no neutralization takes place, the pH is identical with the pA$_i$. During the course of neutralization, the pH increases with additional bases whereas the pA$_i$ remains constant throughout the process.

The pH-pA$_i$ concept was applied to the five-year annual mean datasets for 2005 to 2009. Figure 3.6.1 illustrates that pH and pAi respectively ranged from 4.4 to 6.5 and 3.6 to 5.4. The highest pHs were 6.46, 6.20, 6.19, 6.14 for Yangon, Jiwozi, Ulaanbaatar, and Phnom Penh, respectively. In terms of pA$_i$, however, they are significantly different: the corresponding pA$_i$ were 4.85, 3.78, 4.34, and 4.67. Take Jiwozi, for instance, the input acidity would have been unusually high (it could be as low as pH 3.78 before neutralization!), but due to a high-degree neutralization, the input acidity was relaxed to show a pH of 6.20 eventually. Shizhan and Jinyunshan have pA$_i$s of 3.63 and 3.82, well below 4.0. However, different basic contribution seemed to have happened to the two systems, the resultant pHs were pH 5.68 and pH 4.49 for Shizhan and Jinyunshan, respectively.

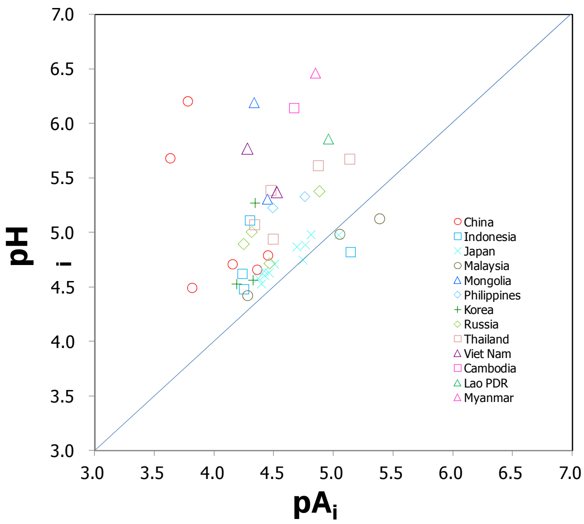


Figure 3.6.1. pA$_i$-pH diagram for the 13 counties of EANET.

It should be pointed out that all plots should be located above the diagonal line of pA$_i$ pH diagram according to the definition. Remarkably well below the line, however, are noted for points for Kototabang (5.15, 4.82) Danum Valley (5.39, 5.12), and Ogasawara (4.98, 5.04). This discrepancy could be explained by possible contribution of organic acids because of the locations and considerably high pHs.

The diagrams for individual countries are provided in Figures. 3.6.2 to 3.6.12. For annual mean pHs, and pA$_i$s derived from annual mean nss-sulphate and nitrate concentrations. This pA$_i$- pH diagram will shed lights on the interpretation of pH by enabling us to understand the acid-base interactions in the determination of precipitation pH.

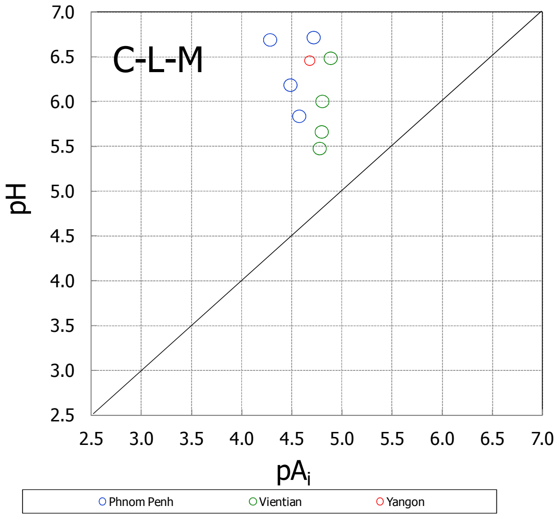


Figure 3.6.2. pA$_i$-pH diagram for Cambodia, Lao PDR, and Myanmar.

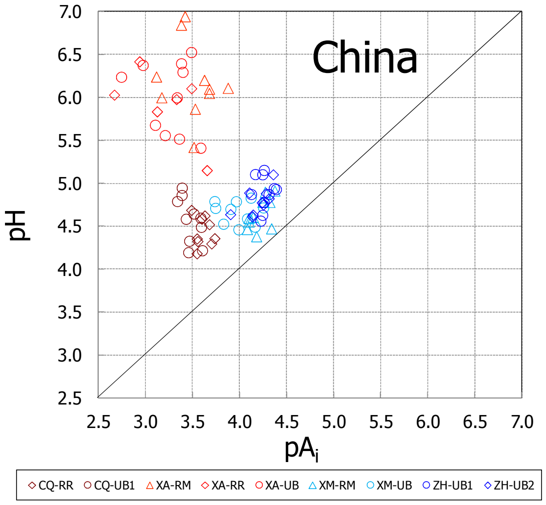
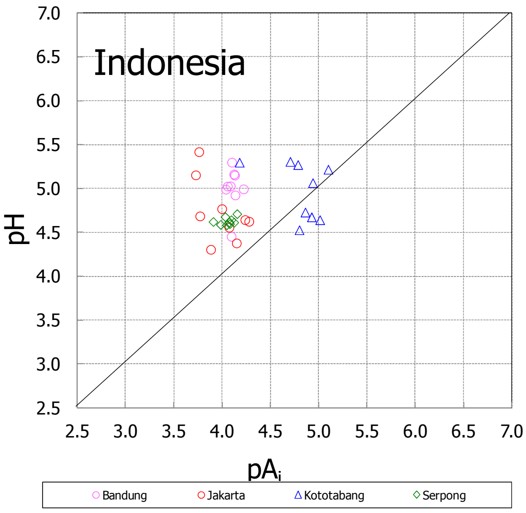


Figure 3.6.3. pA$_i$-pH diagram for China.



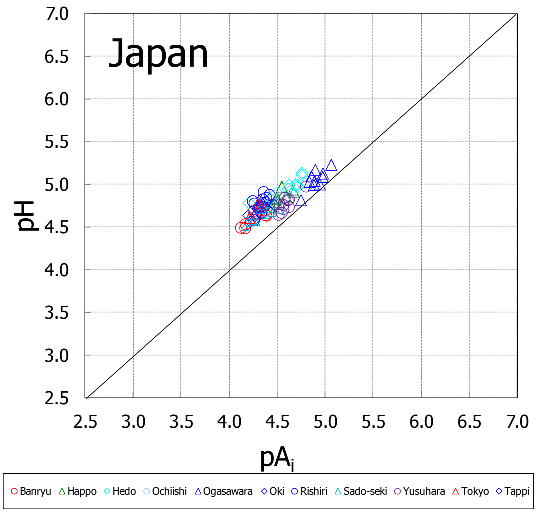
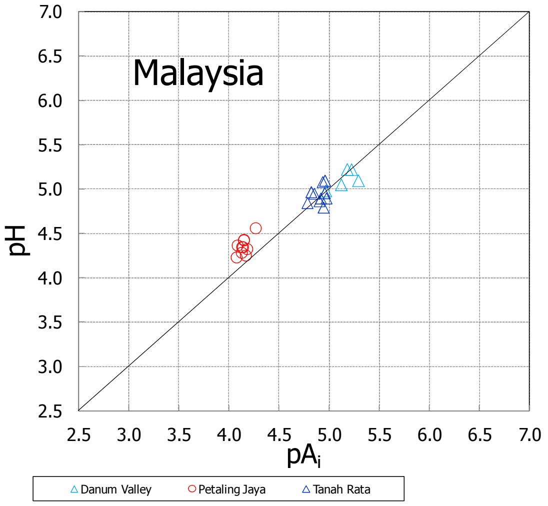


Figure 3.6.4. pA$_i$-pH diagram for Indonesia.

Figure 3.6.5. pA$_i$-pH diagram for Japan.



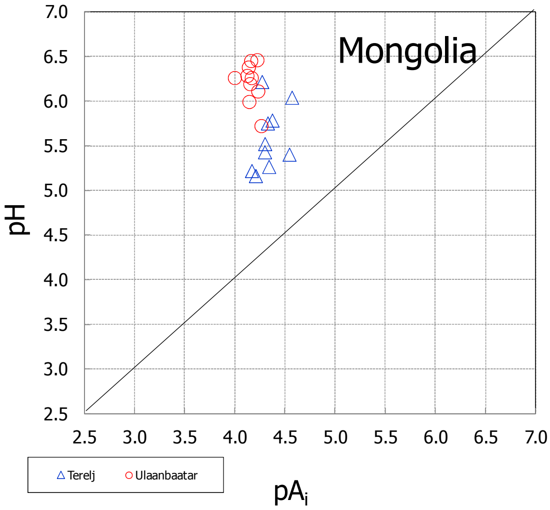
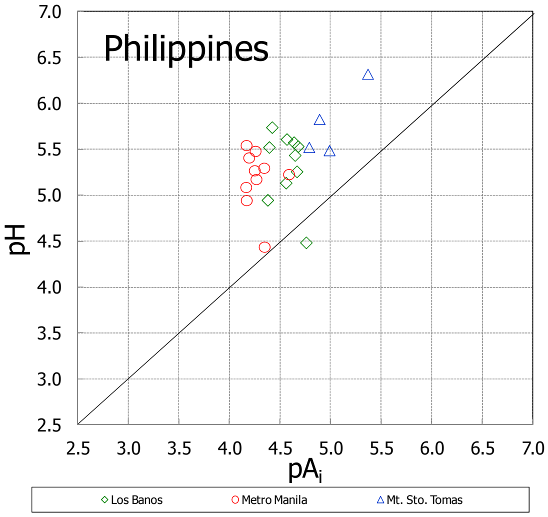


Figure 3.6.6. pA$_i$-pH diagram for Malaysia.

Figure 3.6.7. pA$_i$-pH diagram for Mongolia.



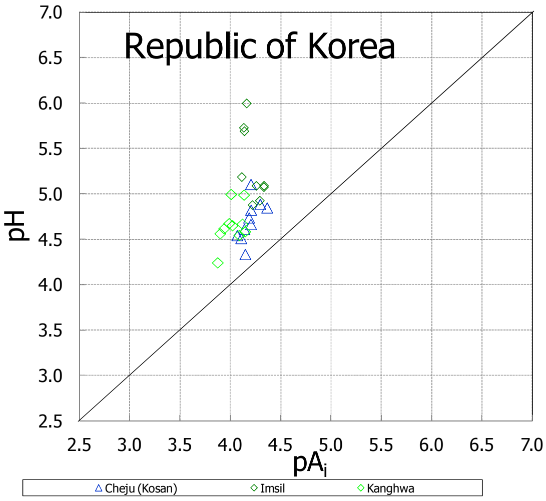


Figure 3.6.8. pA$_i$-pH diagram for the Philippines.

Figure 3.6.9. pA$_i$-pH diagram for Republic of Korea.

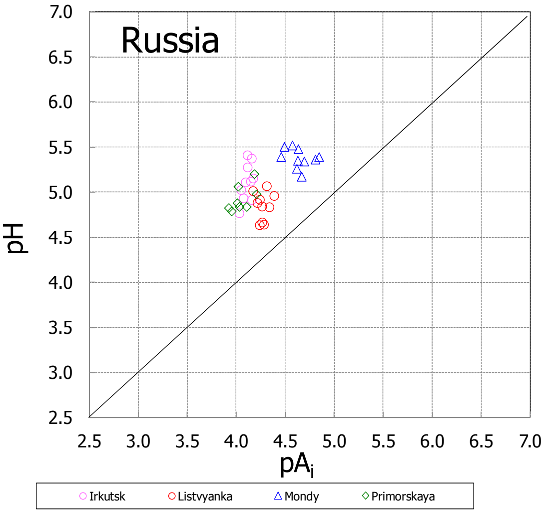


Figure 3.6.10. pA$_i$-pH diagram for Russia.

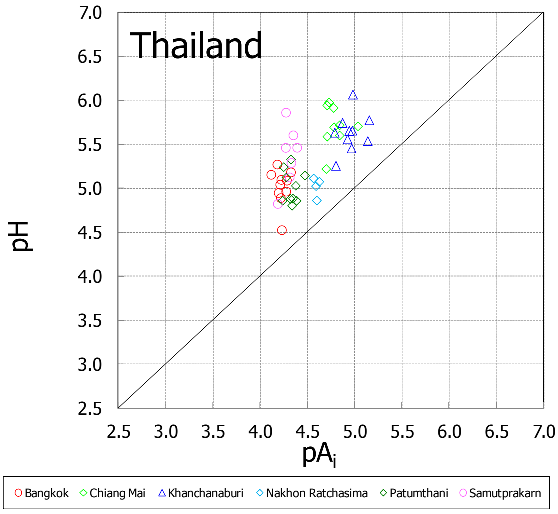


Figure 3.6.11. pA$_i$-pH diagram for Thailand.

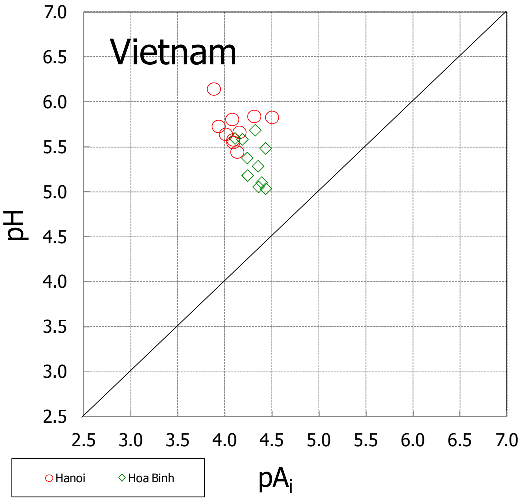


Figure 3.6.12. pA$_i$-pH diagram for Vietnam.

3.7 Relative contribution of some ion pairs

***NO_3^- and nss-SO_4^{2-}*: ** In acid deposition chemistry, two acids, sulphuric and nitric acids play an exclusively important role in acidification of the atmosphere and ecology although some significant roles are played by some other acids including hydrochloric and hydrofluoric ands and a wide range of organic acids. The relative contribution of the two acids are reasonably evaluated in terms of corresponding anions, NO_3^- and nss-SO_4^{2-} because these anions are so stable that they are free from further acid-base interactions. Next point is how to quantify the relative contribution for the entire concentration range for the two anions. Two ratios, nss-SO_4^{2-}/NO_3^- and NO_3^-/nss-SO_4^{2-} have different dependence for concentration variation of one specific ion. In this work, therefore, the relative contribution was quantified in the form of NO_3^- fraction for the sum of the ion pair, NO_3^- and nss-SO_4^{2-}: $[NO_3^-]/([NO_3^-] + [\text{nss-}SO_4^{2-}])$, which is denoted as fraction #1, or F1 for simplicity.

$$F1 = [NO_3^-]/([\text{nss-}SO_4^{2-}]+[NO_3^-])$$

The F1 was calculated for the five-year annual means for the 42 sites (Figure 3.7.1). The maximum, 0.50, occurred at Petaling Jaya, which means two acids, H_2SO_4 and HNO_3, were equally significant in acidification of rain and snow. This value was followed by 0.48, 0.46, 0.45, 0.43, and 0.42 for Pathumthani, Chiang Mai, Bangkok, Vientiane, and Kanchanaburi, respectively. The ratio was lowest, 0.13, for Shizhan, and Jinyunshan (0.16), Jiwozi (0.17), Kototabang (0.18), where H_2SO_4 was four times or more dominant acid than HNO_3 on an equivalent basis.

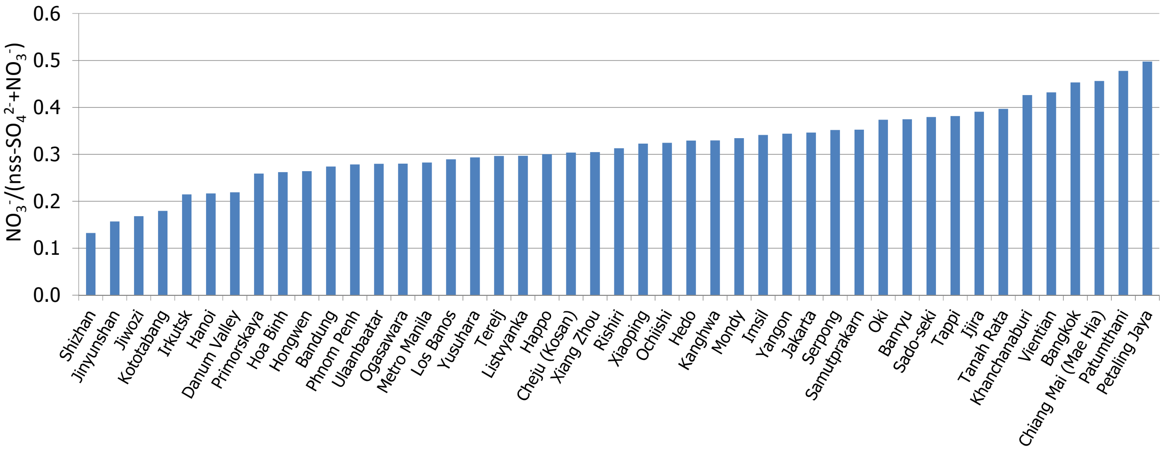


Figure 3.7.1. Equivalent fraction of NO_3^-, $NO_3^-/(nss\text{-}SO_4^{2-} + NO_3^-)$.

***NH_4^+ and nss-Ca^{2+}*:** Where as F1 could focus on the two acids, a similar discussion would be applicable to a pair of two basic compounds, NH_3 and $CaCO_3$. In precipitation samples, these compounds would be represented by NH_4^+ and nss-Ca^{2+}, which suggested to define a fraction, F2, to assess the importance of atmospheric NH_3 relative to $CaCO_3$ as:

$$F2 = [NH_4^+]/([nss\text{-}Ca^{2+}] + [NH_4^+]).$$

Figure 3.7.2 displays this ratio for the five-year annual means in order of the increasing ratio. Values for F2 are mostly over 0.50 where the two compounds equally work as bases in the interaction. Only three sites displayed less than 0.33 for F2: 0.11, 0.25, and 0.33 for Jiwozi, Phnom Penh, and Shizhan, respectively. The maximum was 0.80 for Ijira, and more seven sites had F2 over 0.70: 0.75, 0.75, 0.72 for Serpong, Xiang Zhou, and Danum Valley, respectively. In this way, ammonia is the dominant base in comparison with $CaCO_3$.

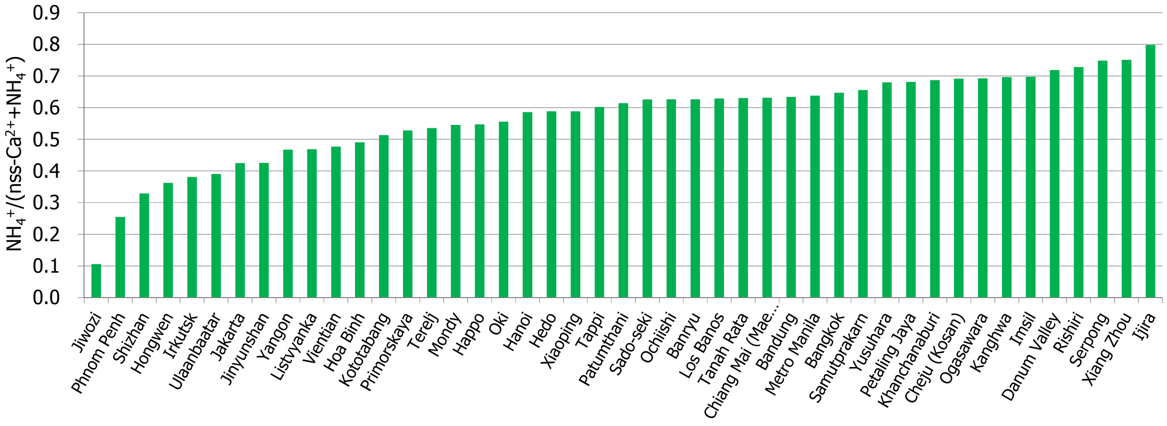


Figure 3.7.2. Equivalent fraction of NH_4^+, $NH_4^+/(nss\text{-}Ca^{2+} + NH_4^+)$.

***NH_4^+ and NO_3^-*: ** Nitrogen saturation is another significant environmental issue, and a third fraction, F3, was discussed for the relative importance of nitrogen species

where F3 is $[NH_4^+]/([NH_4^+] + [NO_3^-])$.

As shown in Figure 3.7.3, ten of the 45 sites displayed less than 0.50 of F3 where the minimum was 0.35 for Petaling Jaya and Jakarta, Oki, Tanah Rata, Banryu, and Tappi recorded 0.40, 0.43, 0.43, 0.44, and 0.55, respectively. The maximum was 0.82 for Kototabang, and nine more stations showed F3s of more than 0.70.

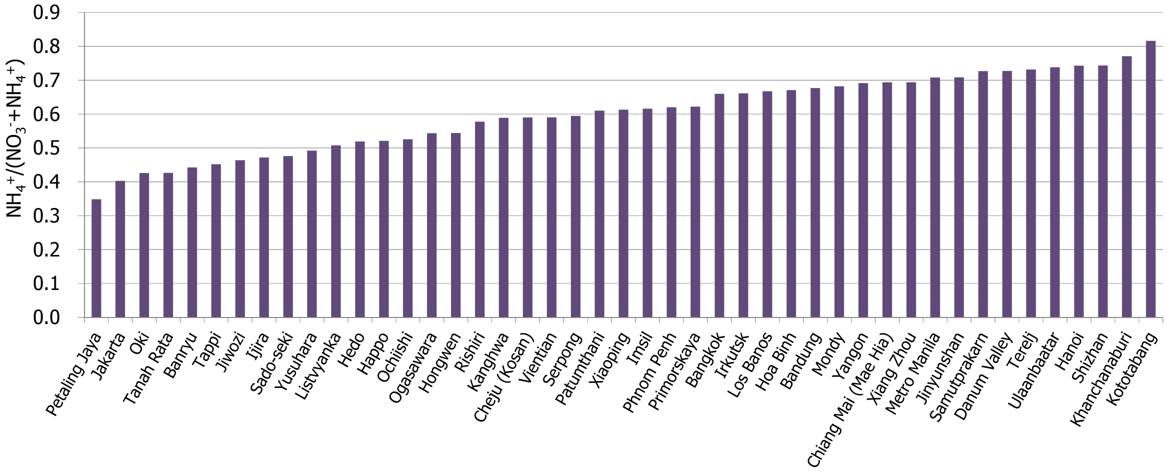


Figure 3.7.3. Equivalent fraction of NH_4^+, $NH_4^+/(NH_4^+ + NO_3^-)$.

Together with the discussion in Figure 3.7.3, ammonia/ammonium is noted to be a predominant species in atmospheric chemistry.

3.8 Deposition of Major Ions and Chemical Indices

Deposition of major ions was discussed in a similar way as the corresponding ionic concentrations. While deposition is calculated by the product between concentration and rainfall amount, the discussion begins with graph for the annual rainfall amounts. Figure 3.8.1 is the graph for annual rainfall amounts skewing to the upper end and covering two orders of magnitude. Six rainfalls were over 4,000 mm yr^{-1} occurred since 2000: 5,078, 4,727, 4,671, 4,286, 4,036, and 4,009 mm yr^{-1} for Mt. Sto. Tomas (2008, 2009), Kuching (2009, 2008), Metro Manila (2000), and Petaling Jaya (2006), respectively. Six lowest records were 69, 83, 89, 100, 111, and 119 mm yr^{-1} for Weishuyuan (2001), Ulaanbaatar (2004), Terelj (2001), Jiwozi (2007), Terelj (2009), and Mondy (2009), respectively. Also five-averages of rainfall amounts were summarized in Figure 3.8.3.

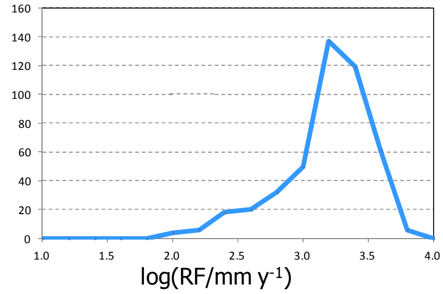


Figure 3.8.1. Graph of annual mean rainfall amounts.

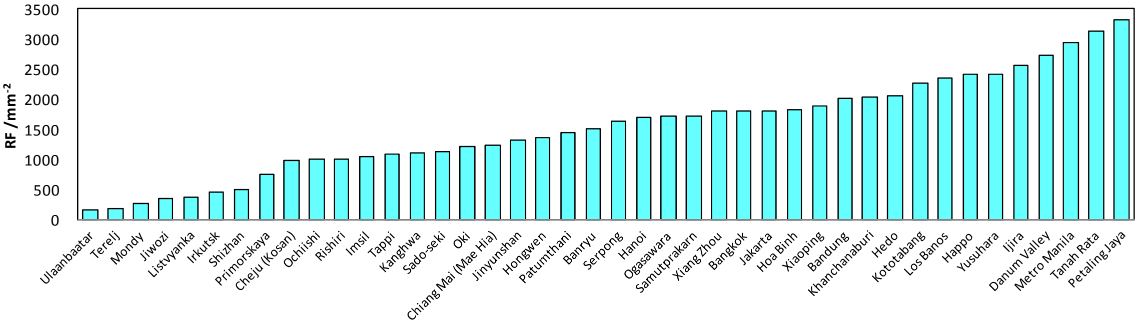


Figure 3.8.2. Five-year average annual precipitation amounts (2005 – 2009)

For deposition, however, some chemical indices, ΣN and H^{+}_{eff} defined below were included in the deposition analysis.

Because "nitrogen saturation" is another environmental focus in relationship with acid deposition or atmospheric deposition, the summation of ionic deposition for ammonium and nitrate were calculated as wet deposition of inorganic nitrogen: $\Sigma N = NH_4^+ + NO_3^-$ where the chemical symbols denote the amount of deposition of the respective species.

The other index, effective hydrogen ion deposition, H^{+}_{eff}, is defined to be (H^{+} +2 NH_4^{+}) where the symbols express the deposition of the species of the symbols on the basis of the biological nitrification of ammonium in the soil environment : $NH_4^{+} + 2O_2 \rightarrow 2H^{+} + NO_3^{-} + H_2O$.

The graph for nss-SO_4^{2-} deposition gave a mono modal pattern different from that for its concentration (Figure 3.8.3). The deposition over 400 meq $m^{-2}yr^{-1}$ was observed for five cases: 583, 506, 478, 461, 457 meq $m^{-2}yr^{-1}$ for Shizhan (2002, 2003), Weishuyuan (2000, 2002) and Guanyinqiao (2004). Less than 3 meq $m^{-2}yr^{-1}$ deposition was reported to be 1.0, 1.3, 1.6, 1.8, and 2.9 meq $m^{-2}yr^{-1}$ for Mt. Sto. Tomas (2006), Vientiane (2009), Terelj (2001), and Mondy (2009 and 2008). The five-year average annual deposition is available for nss-SO_4^{2-} in Figure 3.8.4.

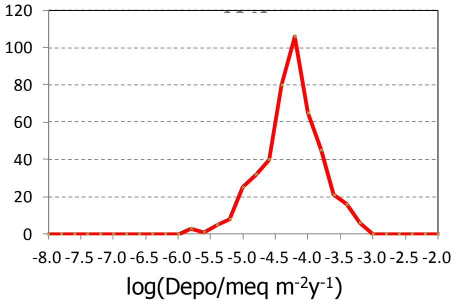


Figure 3.8.3. Graph of annual deposition nss-SO_4^{2-}.

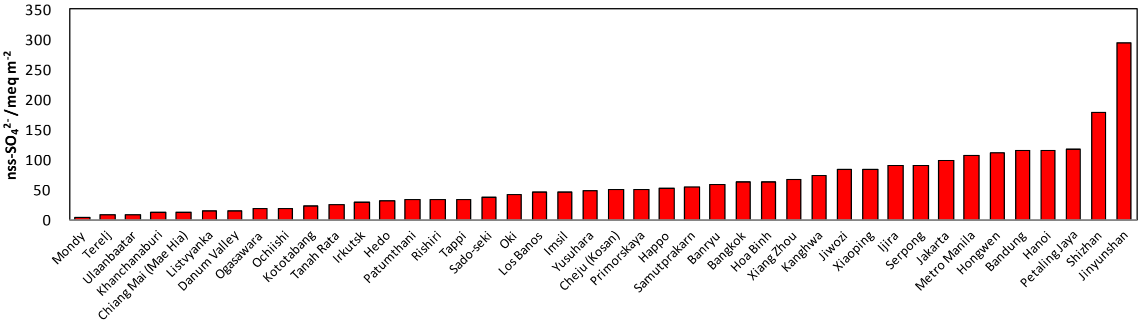


Figure 3.8.4. Five-year average annual deposition of nss-SO_4^{2-} (2005 – 2009)

Nitrate deposition has a mono modal peak with some small shoulders on the lower end (Figure 3.8.5). Kototabang, Petaling Jaya, and Jakarta observed over 100 meq $m^{-2}yr^{-1}$ deposition: 154, 137, 135, 120, 115, 114, and 102 meq $m^{-2}yr^{-1}$ for Kototabang (2003), Petaling Jaya (2009, 2008, and 2006), Jakarta (2005 and 2004), and Petaling Jaya (2005). The smallest amounts of deposition were less than 1.5 meq $m^{-2}yr^{-1}$: 0.8, 0.8, 0.9, 1.3, and 1.4 for Mt. Sto. Tomas (2006), Terelj (2001), Mondy (2009), Ulaanbaatar (2004), and Mondy (2005). For this ion, the five-year average figure is provided in Figure 3.8.6.

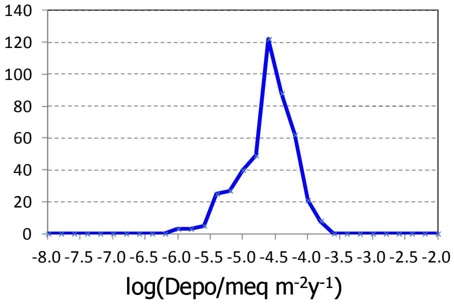


Figure 3.8.5. Graph of annual deposition of NO_3^-.

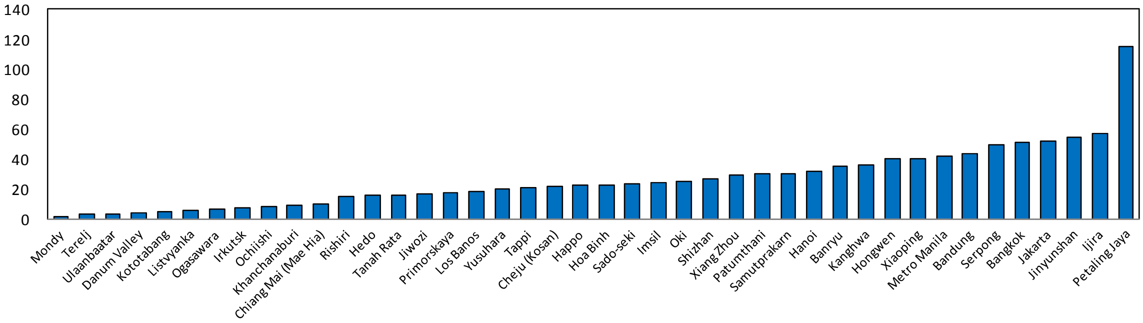


Figure 3.8.6. Five-year average annual deposition of NO_3^- (2005 – 2009)

The ammonium graph is essentially a mono modal pattern with a peak slightly split into two. (Figure 3.8.7). The deposition over 180 meq $m^{-2}yr^{-1}$ occurred at three sites: 235, 190, 189, 188, 183, and 181 meq $m^{-2}yr^{1}$ for Shizhan (2000), Weishuyuan (2000), Guanyinqiao (2004), Weishuyuan (2003), Shizhan (2003), and Guanyinqiao (2007). The five lowest deposition: 0.4, 1.1, 1.5, 2.0, and 2.4 meq $m^{-2}yr^{-1}$for Mondy (2009, 2008, and 2006), Jiwozi (2007), and Mt. Sto. Tomas (2006). The five-year averaged deposition is also provided for this ion (Figure 3.8.8).

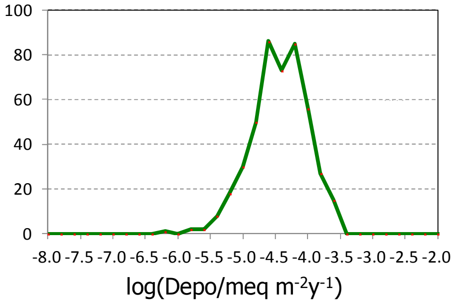


Figure 3.8.7. Graph of annual deposition of NH_4^+.

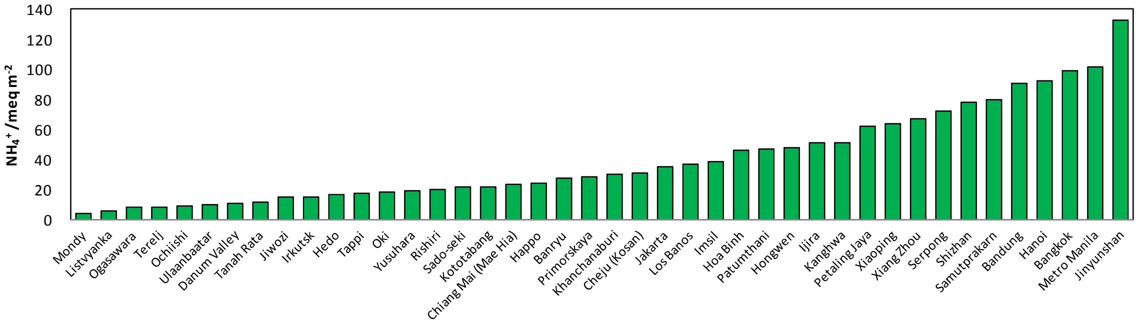


Figure 3.8.8. Five-year averaged deposition of NH_4^+ (2005 – 2009)

The graph for nss-Ca^{2+} deposition was an essentially mono modal peak with two small shoulders on the upper end (Figure 3.8.9). The extreme upper values were: 654, 621, 489, 336, 330, and 327 meq m^{-2}yr^{-1} for Shizhan (2003), Weishuyuan (2003), Shizhan (2001), Guanyinqiao (2005), Shizhan (2002), and Jakarta (2003), respectively. The complementary lower values for nss-Ca^{2+} were: 1.2, 1.9, 1.9, 1.9, 2.4, 2.4, 2.4, and 2.5 for Mondy (2009), Xiaoping (2000), Danum Valley (2008), Vientiane (2009), Ogasawara (2005), Mondy (2005), Ochiishi (2008), Ogasawara (2004), and Ochiishi (2003), respectively. The five-year averaged deposition is also given in Figure 3.8.10.

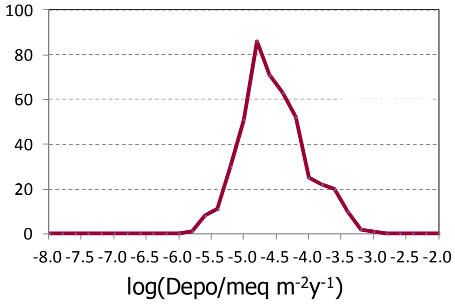


Figure 3.8.9. An annual deposition of nss-Ca^{2+}.

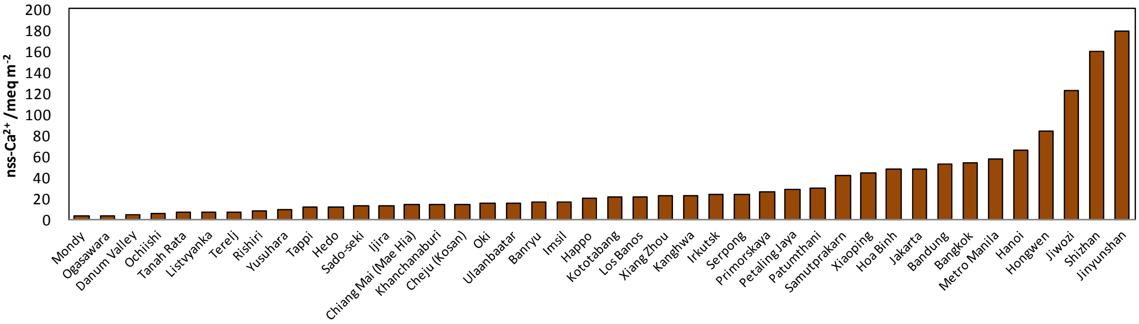


Figure 3.8.10. Five-year averaged deposition of nss-Ca^{2+} (2005 – 2009)

The deposition of H^+ was compiled in the form of graph (Figure 3.8.11). This pattern is different form the others so far. It is a mono modal peak but skewed to the upper end and has a long tailing to the lower end. Some limited sites had considerably larger amount of the deposition whereas some more sites received significantly smaller amount of deposition, which is easily visible for the five-year average diagram in Figure 3.8.12. In terms of a single-year annual deposition, however, highest deposition took place exclusively in Petaling Jaya: 173 (2001), 158 (2003), 157 (2002), 151 (2006), 142 (2004), 140 (2008), and 134 meq $m^{-2}yr^{-1}$ (2009). The lower ends were 0.07, 0.07, 0.07, 0.07, 0.05, 0.03, 0.03 and 0.01 meq $m^{-2}yr^{-1}$ for Ulaanbaatar (2002, 2007), Terelj (2009), Ulaanbaatar (2006), Jiwozi (2008), Weishuyuan (2001), Ulaanbaatar (2004), and Jiwozi (2007), respectively.

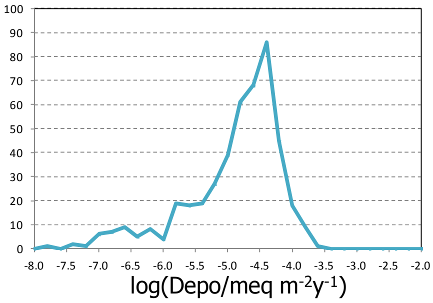


Figure 3.8.11. An annual deposition of H^+.

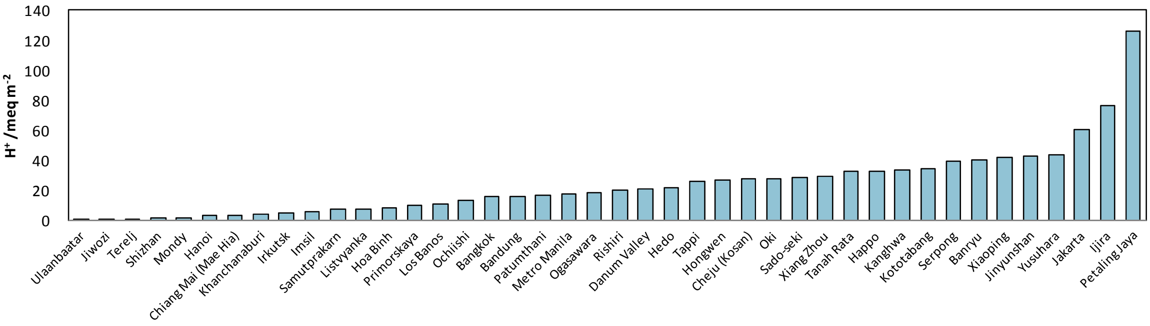


Figure 3.8.12. Five-year average annual deposition of H^+ (2005 – 2009)

The discussion was extended to two chemical indices, ΣN and $H^+_{eff,}$ for possible ecological impacts. The graph for ΣN was an essentially mono modal pattern with a small peak on the lower end (Figure 3.8.13). Four cases were associated with deposition equal to or more than 250 meq $m^{-2}yr^{-1}$: 284, 267, 258, 252, and 250 meq $m^{-2}yr^{-1}$ for Shizhan (2000 and 2003), Weishuiyuan (2003), and Petaling Jaya (2001), and Guanyingqiao (2004). Lowest cases were less than 5 meq $m^{-2}yr^{-1}$: 1.2, 3.3, 3.5, 3.6 and 3.6, 4.2, and 4.9 meq $m^{-2}yr^{-1}$ for Mt. Sto. Tomas (2006), Mondy (2009), Terelj (2009), Mondy (2008, 2006, and 2005) and Vientiane (2009). The five-year mean deposition would be useful (Figure 3.8.14) for inspection of specific sites to understand its significance in the EANET context.

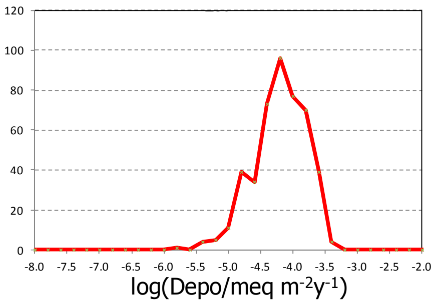


Figure 3.8.13. An annual mean deposition of ΣN.

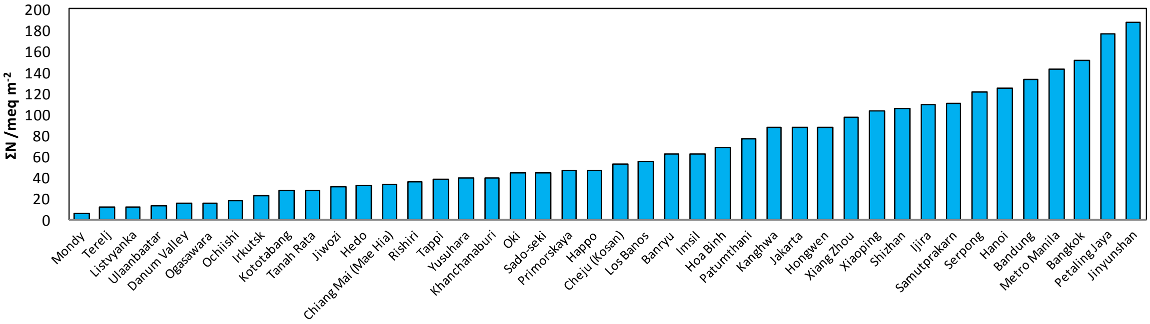


Figure 3.8.14. Five-year average annual deposition of ΣN (2005 – 2009)

Another index for potential impacts on ecological elements, $H^+_{eff,}$ was analysed in terms of graph (Figure 3.8.15). The maximum deposition was 531 meq $m^{-2}yr^{-1}$ for Petaling Jaya (2001), followed by 472, 415, 382, and 392 meq $m^{-2}yr^{-1}$ for Shizhan (2000), Guanyinqiao (2004, 2000, 2007), respectively. The minimum, 1.1 meq $m^{-2}yr^{-1}$ occurred in Mt. Sto. Tomas (2006), and 2.3, 4.7, 5.2, 5.4, and 5.5 meq $m^{-2}yr^{-1}$ followed for Jiwozi (2007), Mondy (2006, 2009, and 2008), and Terelj (2001), respectively. The five-year mean was also provided for this index (Figure 3.8.16).

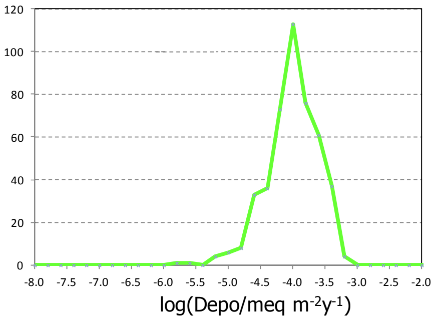


Figure 3.8.15. An annual deposition of H^+_{eff}.

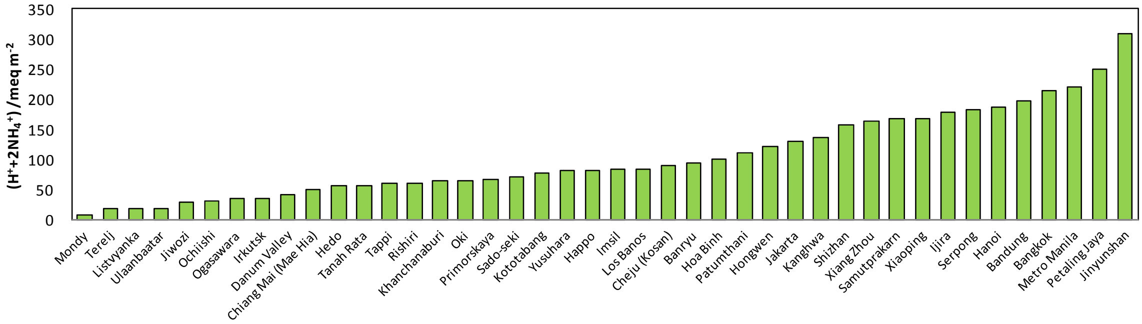


Figure 3.8.16. Five-year average annual depositions of H^{+}_{eff}. (2005 – 2009)

3.9 Temporal Variation of Concentration and Deposition

Temporal variations in the ionic deposition and concentration of major ions and some indices were explored by applying linear regression analysis to the 2000-2009 annual mean concentration and deposition. The slope of the regression line was statistically tested with 95% confidence.

Significant cases are summarized for concentration or deposition of ionic species where some sites implied increasing variation and others decreasing. (Table 3.9.1 and Figures 3.9.1 to 3.9.16). In addition to a variety of species, some site showed increasing nature whereas others decreasing implications. It is interesting to note that Bangkok site showed significantly increasing temporal variations for a number of items (Figures 3.9.1 and 3.9.2). Concentration and deposition of ΣN and H^{+}_{eff} are likely to be increasing, which seemed derived from increasing trends for ammonium ion. Petaling Jaya and Tanah Rata displayed some decreasing variations for plural items (Figures. 3.9.3 to 3.9.6).

Table 3.9.1. Temporal variations in the ionic deposition and concentration of major ions and some indices.

Country	Site	Species	C/D	n	Slope
Thailand	Bangkok	NO_3^-	D	10	3.117
	Bangkok	NH_4^+	C	10	2.739
	Bangkok	NH_4^+	D	10	8.848
	Bangkok	ΣN	C	10	3.986
	Bangkok	ΣN	D	10	13.035
	Bangkok	$(H^+)_{eff}$	C	10	4.738
	Bangkok	$(H^+)_{eff}$	D	10	17.449
Malaysia	Petaling Jaya	nss-SO_4^{2-}	C	10	–1.035
	Petaling Jaya	nss-Ca^{2+}	C	10	–0.374
	Petaling Jaya	nss-Ca^{2+}	D	10	–1.049
Malaysia	Tanah Rata	nss-Ca^{2+}	C	10	–0.268
	Tanah Rata	nss-Ca^{2+}	D	10	–0.716
	Tanah Rata	H^+	C	10	–0.639
Indonesia	Jakarta	nss-SO_4^{2-}	C	9	–5.107
	Jakarta	nss-SO_4^{2-}	D	9	–6.058
Japan	Ijira	nss-Ca^{2+}	C	10	–0.350
	Ijira	nss-Ca^{2+}	D	10	–0.908
Japan	Banryu	NO_3^-	C	10	1.070
	Banryu	NO_3^-	D	10	1.305
China	Xiaoping	NO_3^-	C	10	1.269
	Xiaoping	nss-SO_4^{2-}	C	10	1.352
Thailand	Chiang Mai	NO_3^-	C	9	0.488
	Chiang Mai	ΣN	C	9	0.941
Thailand	Pathumthani	nss-SO_4^{2-}	C	10	–0.930
China	Jinyunshan	NO_3^-	D	9	2.834
Indonesia	Bandung	NH_4^+	C	10	1.928
Japan	Yusuhara	NH_4^+	C	10	0.322

C = Concentration, D = Deposition

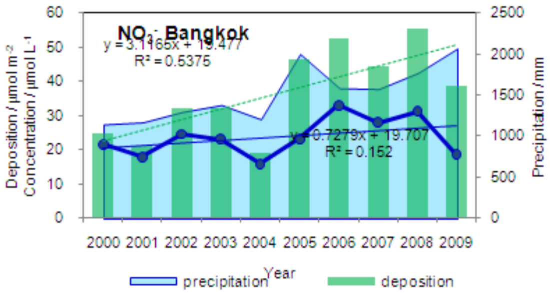


Figure 3.9.1. Temporal variations of concentration and deposition of NO_3^- for Bangkok.

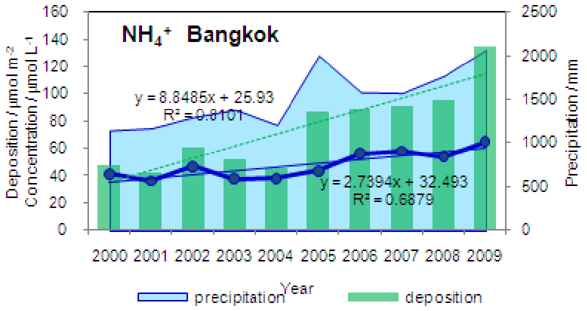


Figure 3.9.2. Temporal variations of concentration and deposition of NH_4^+ for Bangkok.

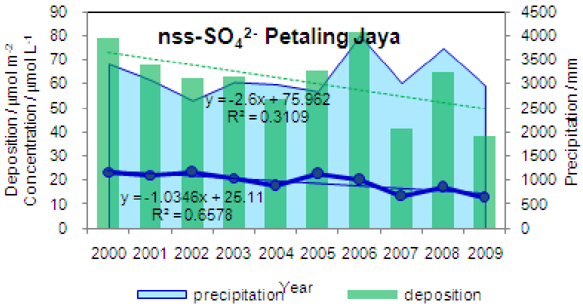


Figure 3.9.3. Temporal variations of concentration and deposition of nss-SO$_4^{2-}$ for Petaling Jaya.

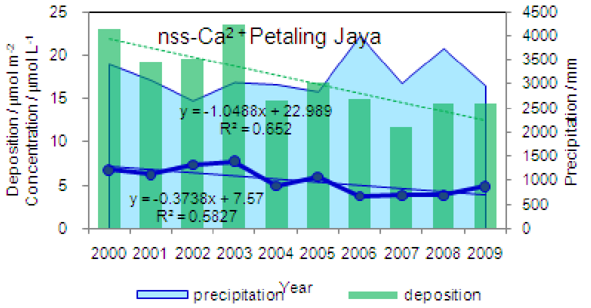


Figure 3.9.4. Temporal variations of concentration and deposition of nss-Ca^{2+} for Petaling Jaya.

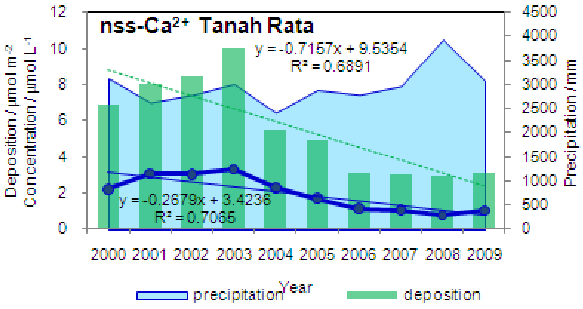


Figure 3.9.5. Temporal variations of concentration and deposition of nss-Ca^{2+} for Tanah Rata.

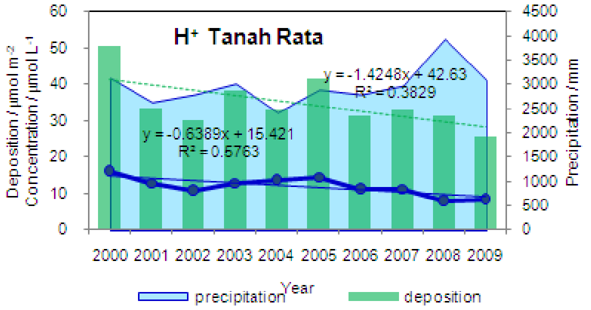


Figure 3.9.6. Temporal variations of concentration and deposition of H$^+$ for Tanah Rata.

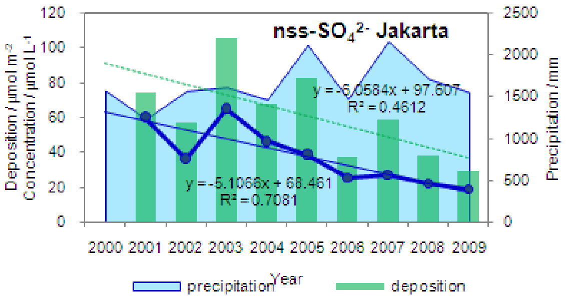


Figure 3.9.7. Temporal variations of concentration and deposition of nss-SO$_4^{2-}$ for Jakarta.

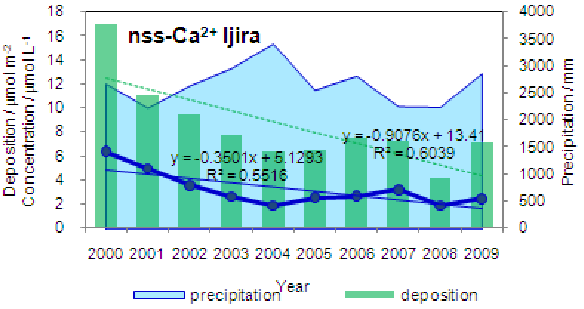


Figure 3.9.8. Temporal variations of concentration and deposition of nss-Ca^{2+} for Ijira.

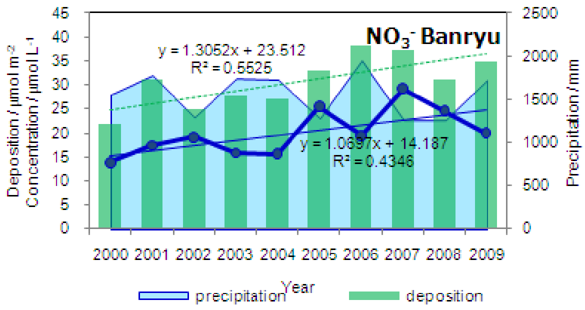


Figure 3.9.9. Temporal variations of concentration and deposition of NO_3^- for Banryu.

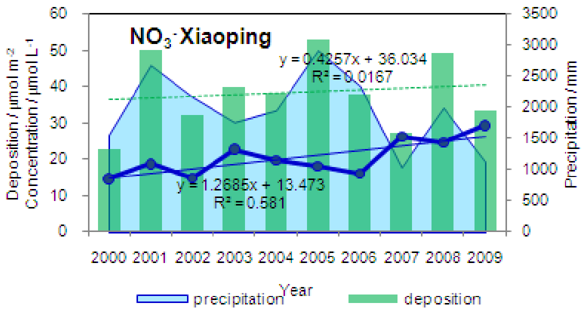


Figure 3.9.10. Temporal variations of concentration and deposition of NO_3^- for Xiaoping.

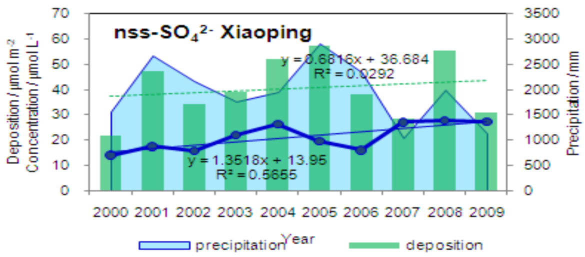


Figure 3.9.11. Temporal variations of concentration and deposition of nss-SO_4^{2-} for Xiaoping.

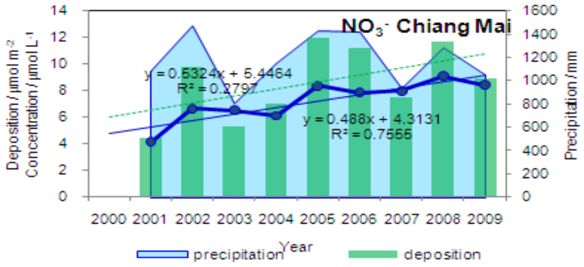


Figure 3.9.12. Temporal variations of concentration and deposition of NO_3^- for Chiang Mai.

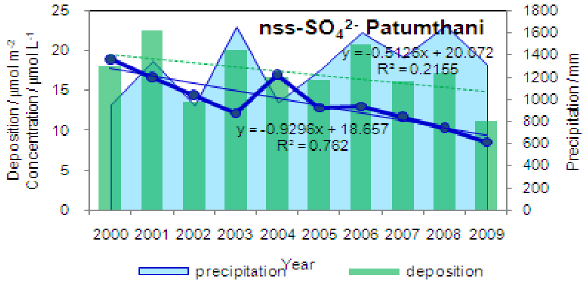


Figure 3.9.13. Temporal variations of concentration and deposition of nss-SO_4^{2-} for Pathumthani

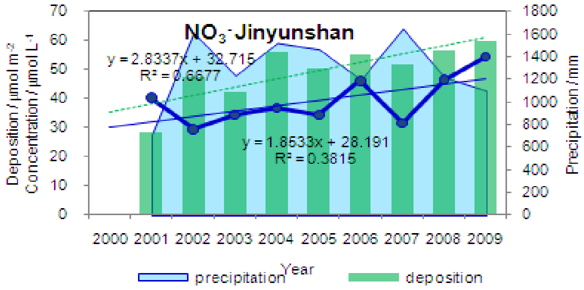


Figure 3.9.14. Temporal variations of concentration and deposition of NO_3^- for Jinyunshan.

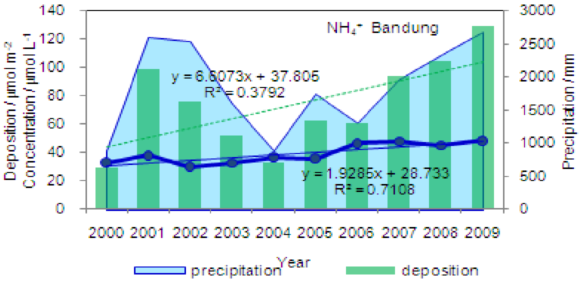


Figure 3.9.15. Temporal variations of concentration and deposition of NH_4^+ for Bandung.

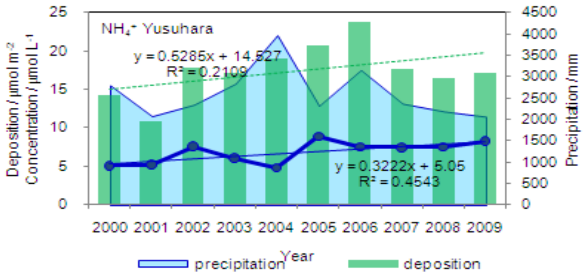


Figure 3.9.16. Temporal variations of concentration and deposition of NH_4^+ for Yusuhara.

Although the EANET has accumulated ten-year record, the length of the period does not suffice for temporal trend analysis because the wet deposition has a number of factors most of which are quite variable with time, the existing time trend could be concealed unlike gas and aerosol species. This would stress the significance of a long-term high-quality monitoring is indispensable to detect some symptoms at all for elimination and mitigation of potential impacts.

3.10 Summary and Recommendations

Another five-year monitoring has produced high quality datasets for wet deposition for East Asia. The precipitation is obviously acidic for most part of the region. Major findings are summarized as below:

(1) The feature of the wet deposition in East Asia is a wide range of concentration and deposition which may well reflect some complex nature of geography and climatology.

(2) In terms of pH, most of the precipitations were acidic in the EANET region whereas the measurements covered a wide range of pHs.

(3) pH-pA$_i$ analysis clarified that even when the observed pH was high, the precipitation had been originally very low, and the high pH was explained by additional bases neutralizing the input acidity.

(4) Sulphuric acid contributed more to precipitation acidity than nitric acid. Some other acids including hydrochloric and hydrofluoric acids and organic acids could be invoked to explain the observed wet deposition chemistry.

(5) In the deposition of basic species, ammonia/ammonium species were more prevailing than basic calcium compounds.

(6) In connection with the point (5), some impacts other than acidification should be considered because indices of ΣN and H^+_{eff} were calculated to be high.

(7) In comparison with European and North American deposition, the EANET region.

Quality of the conclusion of any investigations should be consistent with the quality of the data from which the conclusion is derived whereas a saying tells us "garbage in, garbage out." In order to continue to draw meaningful conclusions for scientific and political purposes, the quality of the monitoring operation should be critically reviewed on a regular basis.

The above mentioned findings therefore make the following recommendations:
(1) Monitoring operations at sites which record tops of concentration and deposition should be closely reviewed with reference to the operational manual and SOP as well as the results of Inter-Laboratory Comparison project. Particularly, mega data should be far more properly elaborated in the routine operation by observing the qualities of siting, sampling frequency, chemical analysis, and QA/QC.
(2) At least in the next Periodic Report of the State of Acid Deposition, a) the wet deposition analysis should include discussion of the monthly and daily datasets, and b) comprehensive discussion should be carried out. c) Trend analysis studies are encouraged for the coming fifteen-year datasets. And d) the wet deposition should be interpreted with accomplishments of emission studies and transport and deposition modelling. For this purpose, no site should be closed and new sites are greatly recommended for improving the conclusions.
(3) In accordance with the regional assessment, national scale studies of the measurements obtained so far are strongly recommended to be investigated in each participating country favourably guided by its SAC members.

3.11. References

EANET. 2006. Data Report on the Acid Deposition in the East Asian Region 2005. Network Center for EANET.
EANET. 2007. Data Report 2006. Network Center for EANET.
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Chapter 4 Dry and Overall Deposition in East Asia

4.1 Introduction

Dry deposition is the transport of gaseous and particulate species from the atmosphere onto surfaces in the absence of precipitations. The relative amount of dry deposition, as compared with wet deposition principally depends on the amount of precipitation in the region. EANET monitoring scheme has facilitated to collect both the wet and dry concentrations and to determine the amount of wet and dry depositions. Within the dry deposition study in EANET region, the priority gases to be monitored include SO_2, O_3, NO, NO_2 (urban), HNO_3, HCl and NH_3 and the particulate components include SO_4^{2-}, NO_3^-, Cl^-, NH_4^+ and PM_{10}. The second priority chemicals which are not a mandatory include NO_2 (rural and remote) and $PM_{2.5}$ (EANET, 2005).

The primary gases: SO_2, NO_x (NO and NO_2) and NH_3 are important in relation to atmospheric deposition. Once emitted to the atmosphere, some of SO_2 and NO_x become oxidized to sulphate and nitrate through both gas and aqueous–phase processes. Also, atmospheric oxidation of volatile organic compounds (VOCs) produces organic acids. The result of these reactions is the formation of acids in the gas phase (HNO_3, HCl, HCOOH, CH_3COOH, etc.) and in the aerosol phase (sulphate, nitrate, chloride, organic acids, etc.). Those primary gases and the aerosols formed by secondary reaction eventually will deposit onto surfaces. The rate of deposition depends on chemical and physical properties of the species, types of surfaces and climatology. Field studies on dry deposition fluxes of various chemical species have been done explicitly.

The atmospheric chemistry of sulphur compounds in gas phase can be expressed as follows,

$$SO_2 + OH^\cdot + M \rightarrow HOSO_2^\cdot + M$$

$$HOSO_2^\cdot + O_2 \rightarrow HO_2^\cdot + SO_3$$

where M is any atmospheric molecules (nitrogen and oxygen in effect) called a third body of the reaction.

Sulphur trioxide, in the presence of water vapor, is converted rapidly to sulphuric acid

$$SO_3 + H_2O \rightarrow H_2SO_4$$

In the case of nitrogen compounds,

$$NO_2 + OH + M \rightarrow HNO_3 + M$$

is the major oxidation pathway to give nitric acid. Another reaction pathway,

$$NO_2 + O_3 \rightarrow NO_3 + O_2$$

$$NO_3 + NO_2 + M \leftrightarrows N_2O_5 + M$$

$$N_2O_5 + H_2O \rightarrow 2HNO_3$$

occurs mainly in the nighttime, as during the daytime NO_3 photolyzes rapidly (Seinfeld and Pandis, 1998).

In the present of halogen species, i.e. sea-salt, the strong acids such as H_2SO_4 and HNO_3 can react to form secondary aerosols,

$$H_2SO_4 + 2NaCl \rightarrow 2HCl + Na_2SO_4$$

and $N_2O_5 + NaCl \rightarrow ClNO_2 + NaNO_3$

Sulphuric and nitric acids are neutralized by NH_3 in the atmosphere as follows.

$$H_2SO_4 + 2NH_3 \rightarrow (NH_4)_2\,SO_4$$

$$HNO_3 + NH_3 \leftrightarrows NH_4NO_3$$

Sulphuric acid formed by oxidation of SO_2 may in turn neutralized by NH_3 to form ammonium sulphate. The conversion of H_2SO_4 to $(NH_4)_2SO_4$ changes the physical and chemical properties of the species resulting in changes in deposition characteristics. Gaseous nitric acid formed by oxidation of NO_2 may be neutralized by NH_3 to form ammonium nitrate solid aerosol. This equilibrium reaction between NH_3 and HNO_3 is particularly important from the point of dry deposition, since the very large deposition velocity of HNO_3 is lowered substantially by converting to NH_4NO_3 (Erisman and Draaijers, 1995).

Tropospheric ozone is produced from the photochemical reaction of nitrogen oxides (NOx) and volatile organic compounds (VOCs) under sunlight. NOx and VOCs are emitted from automobiles and industries among other sources. Since the emission of nitrogen oxides in East Asia is rapidly increasing due to rapid economic development, the concentrations of tropospheric ozone are increasing and are predicted to increase further in the future (Erisman and Draaijers, 1995). Under photochemically active condition to produce high concentration of O_3, the concentration of OH, which is the most important oxidant in the atmosphere, is also high to facilitate the reaction of NO_2 to form HNO_3. O_3 is also an important oxidant in cloud water droplet to oxidize SO_2 to H_2SO_4. Therefore, ozone is a very important species indirectly attributable to acid deposition. Furthermore, adverse impacts of ozone have recently been paid attention. When ozone is inhaled into a human body, it may cause health effects such as respiratory and cardiology diseases. High concentration of ozone may also cause agricultural production losses and forest damages.

Particulate matter is a complex mixture of small particles and liquid droplets. It is composed of a number of components, including acids (such as nitrates and sulphates), organic substances, metals, and soil or dust particles. The size of particle matters is related to health problems. PM_{10}, particles that are 10 micrometers in diameter or smaller, are passing through the throat and nose and enter the lungs. Once inhaled, these particles can affect the heart and lungs and cause serious health effects. $PM_{2.5}$ that are 2.5 micrometers in diameter or smaller can be emitted from combustion sources such as diesel engines and forest fires, or formed in the atmospheric reactions of SO_2, NOx and VOCs. Epidemiological studies in the United States, Europe, and Asia have shown a remarkable correlation between the mortality rate of a population and the concentration of fine particles with $PM_{2.5}$ mass concentration (Dockery *et al.*, 1993).

Furthermore, ozone and some kinds of particulate matter such as sulphate and black carbon affect the global radiative forcing. These ozone and PM are important air pollutant in view of not only acid deposition but also regional air quality and climate change.

The dry deposition fluxes have been identified as being necessary for evaluation at the regional level in addition to wet deposition, in order to provide more comprehensive data on atmospheric impacts on the environment. There were 44 air concentration monitoring sites operating in the EANET network in 2009. The set of compounds whose atmospheric concentrations are monitored differs depending on the methodology at each monitoring site. Filter pack method has been introduced to EANET as the most suitable method for monitoring gas and aerosol concentration. For nitrogen oxides (NO_x) and ozone (O_3), passive samplers are used at several sites. Automatic air monitors have been introduced in some countries. The Task Force on Monitoring for Dry Deposition (TFMDD) decided to employ the inferential method for dry deposition measurement in EANET, i.e. the dry deposition flux is calculated by the product of atmospheric concentration, and dry deposition flux of each species under consideration. The deposition velocity is in turn estimated based on land surface properties and meteorological parameters. Direct flux observation has been conducted at selected sites on a research base for future improvement of the inferential method.

4.2 Dry deposition assessment methodology

Method of estimation

Based on the Strategy Paper for Future Direction of Dry Deposition Monitoring of EANET (Network Center for EANET, 2005), the Inferential Method is adopted to estimate the dry depositions of EANET priority chemical species. The method can estimate the dry depositions around the monitoring sites using the data measured at the sites. Since deposition velocity depends on height, a reference height $z_{ref} = z - d$, where z is height above ground d is the so-called zero-plane displacement height, should be determined. In this report, the target area and reference height for the estimation were set at 1 km around the sites and 20 m, respectively. Dry deposition amounts of gaseous and particle species are estimated as the product of an atmospheric concentration and a deposition velocity expressed as, $F_{dry} = V_d \times C$, where F_{dry}, V_d, and C denote the dry deposition flux, deposition velocity, and atmospheric concentration, respectively for each species. The V_d was calculated using a parameterization arranged by Matsuda (2008) based on recent field studies in East Asia (Matsuda *et al.*, 2007).

The atmospheric concentrations of sulphur and nitrogen compounds were monitored using the four-stage filter-pack method and following the procedures described in the Technical Document for

Filter Pack Method in East Asia (Network Center for EANET, 2003). The filter-pack setup was connected with the sampling system, and the air flow rate of the sampling pump was set either at 2.0 $l.min^{-1}$ or 1.0 $l.min^{-1}$ for different sites. Filter-pack sampling was performed every 2 weeks which followed the schedule of wet deposition monitoring since most sites locate in remote area. The sampling height exceeded 3 m above the ground at most sites.

Parameterization terms to be used by EANET countries

The V_d was calculated using a parameterization arranged by Matsuda (2008) based on recent field studies in East Asia (Matsuda *et al*., 2007). Although land-use type near the observation sites includes forest, grass and other types of surfaces, dry deposition was calculated only for forest and grass surfaces in this report, because the atmospheric deposition on the land area was highlighted for the sake of evaluation of impact on terrestrial environment. In this estimation, the hourly F_{dry} ($mmol.m^{-2}hr^{-1}$) was calculated as the product of the modeled hourly V_d and 2-week-averaged atmospheric concentrations obtained by the four-stage filter-pack method. To calculate hourly F_{dry}, hourly atmospheric concentration was used as same value during the sampling period (2 weeks). The annual F_{dry} ($mmol.m^{-2}yr^{-1}$) was the means of at least 70% available 2-week deposition fluxes in a year following the estimation by Baumgardner *et al*. (2002). The V_d of gases was obtained by the resistance model (Matsuda, 2008) which was consisted of three resistances, the aerodynamic resistance (R_a), the quasi-laminar layer resistance (R_b) and the surface resistance (R_c), respectively. In this report, these resistances were estimated for forest and grass surfaces. Ra and Rb were determined based on Erisman and Draaijers (1995). To calculate R_a and R_b, the reference height was set at 20 m, and canopy heights (h) were defined as 10 m for forest and 1 m for grass. The zero-plane displacement height and roughness length were set as 0.7 h and 0.1 h, respectively. The atmospheric stability determined by Paquill's classification was taken into account for the calculation.

R_c was based on Wesely's parameterization (1989) except the outer surface resistance (R_{lu}) of SO_2 and NH_3. Regarding land-use types defined by Wesely (1989), "mixed forest including wet land" was used for the forest surface, and "mixed agricultural and range land," for the grass surface. Seasonal categories defined by Wesely (1989) were set in accordance with local climate. The wet leaf surface can act as an effective sink for soluble gases, such as SO_2 and NH_3 (Erisman and Draaijers, 1995). Previous studies in Japan also suggested the enhancement of the uptake of SO_2 by the wet surface (Matsuda *et al*., 2002; Takahashi *et al*., 2002). In this estimation, the R_{lu} for SO_2 and NH_3 were obtained by the meteorological parameters related to surface wetness (Erisman, 1994; Smith *et al*., 2000). The V_d of particulate matter for forest and grass surfaces was calculated referring to Brook *et al*. (1999), in which V_d of particulate matter estimated for low and high canopies separately (Wesely *et al*., 1985; Erisman *et al*., 1997). Meteorological parameters, such as the temperature, relative humidity, solar radiation, and wind speed, necessary for the V_d calculation, were continuously measured at each EANET site. Currently hourly meteorological parameters were reported for Japanese sites, and thus dry deposition estimation was implemented only for Japanese sites. To determine Paquill's stability classification, cloud coverage data at the nearest meteorological observation stations from the respective sites were used. Since V_d of gases and particulate matter are different in different types of land surfaces, the V_d at each site was first calculated for forest and grass surfaces, and the weighted-average of V_d was then estimated according to the land-use ratio of forest and grass within 1 km square from the site.

In this report, estimation of dry deposition flux was made only for Japanese site due to availability of hourly meteorological data and land use data which are necessary parameters described in the Technical Manual on Dry Deposition Flux Estimation in East Asia (Network Center for EANET, 2010).

4.3 Spatial variation of air concentration

EANET regions are varied greatly in topographical and climatological characteristics, according to these significant spatial characteristic differences, analyses on dry deposition in the EANET regions are to be reported in two broadly classified as the Northeast and Southeast Asian sub-regions.

Northeast Asia of EANET region comprises the eastern part of Russia, Mongolia, China, the Republic of Korea and Japan. The Russian Far East under the influence of the Pacific Ocean has a monsoon climate that reverses the direction of wind in summer and winter, sharply differentiating temperatures. Mongolia's terrain is one of mountains and rolling plateaus, with a high degree of relief. The country has an average elevation of 1,580 m and extremely continental climate with long cold winters and short summers with most precipitation fall. The country averages 257 cloudless days a year, and average precipitations in the North are 2,000-3,500 $mm.yr^{-1}$, and lowest in the South, which receives 1,000-2,000 $mm.yr^{-1}$. Average temperatures over most of the country are below freezing from November through March and are about freezing in April and October. January and February averages of -20°C are common. For China, the climate of China is extremely diverse; subtropical in the South to subarctic in the North. Tremendous differences in latitude, longitude, and altitude give rise to sharp variations in precipitation and temperature within China, its climatic patterns are complex. The Korean Peninsula is under the influence of monsoon region has a temperate climate with four distinct seasons. The movement of air masses from the Asian continent exerts greater influence on the Southern Korea's weather. It is about 74% of Japan is mountainous, with a mountain range running through each of the main islands and scattered plains and intermontain basins. Japan belongs to the temperate zone with four distinct seasons. Japan is generally a rainy country with high humidity. These are all climatic information of the Northeast Asian region.

Spatial air pollution variations in the Northeast Asian regions are presented based on average annual observed values. SO_2, HNO_3, NH_3 and aerosol components are as SO_4^{2-}, NO_3^- and NH_4^+ within the areas classified as urban, rural and remote.

For the most urban Eastern Asia, the observed values of NH_3 are higher than SO_2 and much larger than HNO_3 (Figure 4.3.1). This phenomenon was unexpected for the city to occur because NH_3 is generally thought to be emitted in agricultural areas. The NH_3 concentrations were observed in ranges of 7.9–11.2 ppb at Hongwen site; 7.4–10.7 ppb at Ulaanbaatar site; 1.8–3.8 ppb at Irkutsk site. The SO_2 concentrations were found in ranges of 5.2–10.9 ppb at Hongwen site, 1.8–4.8 ppb at Ulaanbaatar site, 2.2–5.1 ppb at Irkutsk site. The urban concentrations at Banryu site contain much lower SO_2 (0.6–1.1 ppb) and NH_3 (1.0–1.2 ppb) comparing with all other urban areas within the East Asia of EANET region.

The aerosol concentrations of sulphate (SO_4^{2-}) were found to be higher than NO_3^- and NH_4^+ at all urban sites (Figure 4.3.2). The concentrations of SO_4^{2-} were observed in ranges of 10.58–13.38 $\mu g.m^{-3}$ at Hongwen site, 5.11–6.55 $\mu g.m^{-3}$ at Banryu site, 4.62–5.36 $\mu g.m^{-3}$ at Tokyo site, 1.33–5.68 $\mu g.m^{-3}$ at Ulaanbaatar site and 2.13–2.91 $\mu g.m^{-3}$ at Irkutsk site.

The nitrate concentrations were found in ranges of 5.81–8.10 $\mu g.m^{-3}$ at Hongwen site, 1.35–1.57 $\mu g.m^{-3}$ at Banryu site, 3.85–4.03 $\mu g.m^{-3}$ at Tokyo site, 0.9–1.74 $\mu g.m^{-3}$ at Ulaanbaatar site and 0.61–0.89 $\mu g.m^{-3}$ at Irkutsk site. The observed values of NH_4^+ were substantially lower than NO_3^- in most East Asia urban sites. Although ammonium ions react readily with sulphate to form ammonium sulphate, $(NH_4)_2SO_4$ but the reaction $NH_3(g) + HNO_3(g) \leftrightharpoons NH_4NO_3(s)$ can form ammonium nitrate particles. This reaction is in favor only in low sulphur atmosphere (Stelson, 1982).

The gaseous components of HNO_3 and NH_3 were not monitored at Imsil and Ganghwa sites before the year 2006. Imsil and Ganghwa sites observed low ranges of SO_2 (1.4–2.5 ppb and 2.8–3.4 ppb, respectively) but higher for NH_3 (3.2–9.7 ppb and 5.0–6.0 ppb, respectively). All measured chemical component at Ijira site are comparatively low. The SO_2 concentrations at Listvyanka and Primosrskaya rural sites were observed in ranges of 1.1–6.0 ppb and 0.7–1.6 ppb, respectively and the NH_3 were found in ranges of 0.8–4.4 ppb and 1.2–3.6 ppb, respectively. The HNO_3 concentration was significantly low for all the East Asia rural sites.

The three gas components, SO_2, HNO_3 and NH_3 at Jeju remote site were noticeably higher than those at the other remote sites within the East Asia region. The observed concentrations of SO_2 were in ranges of 2.2–3.4 ppb; HNO_3, 0.5–0.7 ppb; NH_3, 3.7–4.9 ppb. These values are in close proximity to the gas concentrations at Ganghwa site but higher than that at Imsil site, the two rural sites of the Republic of Korea. Gas concentrations observed at the six remote sites in Japan varied with increasing values from the monitoring sites from Rishiri site in the north down to Oki site in the south, except at the two island southernmost remote sites, Hedo and Ogasawara, which showed very low concentrations. Rishiri site contained the lowest SO_2 concentration, 0.1–0.3 ppb, followed by Ochiishi site, 0.2 ppb; Tappi site, 0.2–0.5 ppb; Sado-Seki site, 0.4–0.6 ppb; Happo site, 0.4–0.8 ppb; Oki site, 0.6–0.9 ppb; Yusuhara site, 0.9–1.2 ppb. The two southern islands, Hedo contained very low SO_2 concentration of 0.2–0.4 ppb, and SO_2 was non-detectable at Ogasawara site. The ammonia concentrations at all Japan remote sites were varied within a range of 0.2–1.2 ppb. The concentration of HNO_3 at all East Asia remote sites was low within a range of 0.1– 0.7, especially at Mondy, the value was non-detectable.

The observed values of aerosol component, SO_4^{2-} were significantly high comparing with NO_3^- and NH_4^+ at all remote East Asia sites. These results indicate that the source of sulphur may derive from both human activities and the natural sulphate. By looking at the four chemical correlated species, i.e. SO_4^{2-}, Na^+, Cl^- and Ca^{2+}, the Japan sites that showed high value of SO_4^{2-} component tended to show also high Na^+, Cl^- and Ca^{2+}. For example, Hedo, the island site, the SO_4^{2-} concentration found in a range of 5.32–6.51 μg.m^{-3} and Na^+, Cl^- and Ca^{2+} were found in ranges of 3.77–5.22 μg.m^{-3}, 4.40–7.29 μg.m^{-3} and 0.34–0.48 μg.m^{-3}, respectively. At the same period, Happo, the inland mountainous site, SO_4^{2-} concentration found in a range of 5.32–6.51 μg.m^{-3} and Na^+, Cl^- and Ca^{2+} were found in ranges of 3.77–5.22 μg.m^{-3}, 4.40–7.29 μg.m^{-3} and 0.34–0.48 μg.m^{-3}, respectively. Relatively, the Happo site located in the mountainous site of central Japan shows all low concentrations of SO_4^{2-} (0.59 – 2.79 μg.m^{-3}), Na^+ (0.25–0.29 μg.m^{-3}), Cl^- (0.01–0.03μg.m^{-3}) and Ca^+ (0.07–0.17 μg.m^{-3}). At Jeju Site, in the Republic of Korea, the SO_4^{2-} concentrations show no relationship with Na^+, Cl^- and Ca^{2+}. The SO_4^{2-} concentration was considerably high for the remote area (4.68–8.05 mg.m^{-3}) while Na^+, Cl^- and Ca^{2+} are in low ranges (0.35–0.51 μg.m^{-3}, 0.13–0.32 μg.m^{-3} and 0.05–0.11 μg.m^{-3}, respectively). The measured concentrations of all the aforementioned chemical components are substantially low in Terelj, Mongolia, and Mondy, Russia. The SO_4^{2-}, Na^+, Cl^- and Ca^{2+} are found in ranges of 0.85–1.42 μg.m^{-3}, 0.08–0.14 μg.m^{-3}, 0.19–0.36 μg.m^{-3} and 0.27–0.42 μg.m^{-3} at Terelj site, respectively and 0.22–0.49 μg.m^{-3}, non-detectable – 0.04 μg.m^{-3}, non-detectable – 0.03 μg.m^{-3} and 0.03 – 0.07 μg.m^{-3} at Mondy site, respectively. It should be noted that other particulate components, i.e. Mg^{2+} and K^+, the concentration values are within the Ca^{2+} ranges at all East Asia remote sites.

Other important aerosol components, i.e. nitrate (NO_3^-) and ammonium ion (HN_4^+), their concentration distributions are considerable low at all remote sites.

NO_2, NOx and PM_{10} concentrations are high in sites in China, whereas it showed lower concentrations at Japanese sites, most of which are classified as remote site. Annual O_3 concentrations were higher at mountainous Japanese and Russian sites (Figure 4.3.3).

Southeast Asia lies on two geographic regions. Continental Southeast Asia comprises Cambodia, Laos, Myanmar, Thailand, Vietnam and Peninsular Malaysia, and Maritime Southeast Asia comprises Indonesia and the Philippines. The climate in Southeast Asia is mainly tropical and sub-tropical – hot and humid all year round with plentiful rainfall. Southeast Asia has a wet and dry season caused by seasonal shift in winds or monsoon. The tropical rain belt causes additional rainfall during the monsoon season. The climatology of countries near equator (Malaysia and Indonesia) is rain forest. The mountain areas in the Northern region (Laos and Myanmar) have milder temperatures and drier landscape.

The monitoring results in 2005-2009 show substantially high concentration of NH_3 in most of urban sites when comparing with SO_2 (Figure 4.3.1). Their concentrations are generally 3 times higher than the measured SO_2 values. It is interesting to see the observed NH_3 is high while HNO_3 is apparently very low in the urban areas. Since the oxidative reactions nitrogen oxides to produce HNO_3 should have been taken place. High concentration of NH_3 is presumably derived from fertilization consumed in the rural areas under hot and humid conditions. Agricultures are major activities in Most Southeast Asia countries.

Although Jakarta (Indonesia) and Samutprakan (Thailand) sites monitored only SO_2 and the observed values are noticeably high. The concentrations spanned in ranges of 7.4–11.0 ppb and 4.8–6.6 ppb, respectively. The SO_2 concentration in Bangkok during the past 5 years is improving. The observed value in the year 2005 is 7.3 ppb and down to 1.9 ppb in the year 2009. The government policy to improve vehicle fuel quality has been effective. Other urban sites at Petaling Jaya (Malaysia), Metro Manila (Philippines) and Hanoi (Vietnam), the SO_2 concentration found in ranges of 1.8–6.3 ppb, 1.2–3.1 ppb, 0.8–7.3, 1.3–3.9 ppb, respectively. The measured values of NH_3 are in ranges of 6.1–16.9 ppb, 3.8–12.5 ppb, 7.8–10.2 ppb and 2.8–5.0 ppb at Petaling Jaya, Metro Bangkok and Hanoi, respectively. The amounts of HNO_3 are reportedly very low at all the urban sites. This component has short-live in the atmosphere since it can be formed readily in humid atmosphere and then washed out by heavy monsoon rain in the tropical region.

Variation in aerosol compositions at EANET unban sites in South Asia are observed (Figure 4.3.2). The sulphate components are proportionately much greater than the other two acid components, whether the site is located in high land (Hanoi), hot and humid in central (Bangkok), the Malay Peninsula (Petaling Jaya) or pacific island (Metro Manila). The SO_4^{2-} concentrations at Petaling Jaya, Metro Manila, Bangkok and Hanoi were found in ranges of 2.26–3.32 $\mu g.m^{-3}$, 2.37–3.31 $\mu g.m^{-3}$, 2.13–4.71 $\mu g.m^{-3}$ and 8.77–20.35 $\mu g.m^{-3}$. It is noted that Hanoi had improved markedly in reducing quantitatively the ambient aerosols during 2005-2009. For other components, i.e. NO_3^- and NH_4^+, their measured values are small.

There is spatial variation of chemical compositions among the Southeast Asia rural sites. High ammonia concentration observed at all the EANET rural sites indicates large effects of Nitrogen constituent emissions from agriculture in the vicinity of the monitoring sites and less influence of mass transport of sulphur component from urban areas. The concentrations ratio of NH_3/SO_2 at Serpong site (Indonesia) is 7; Los Banos (Philippines), 11; Chiang Mai (Thailand), 6 and about equal proportion at Hoa Binh (Vietnam), 1.5. High amount of NH_3 and low HNO_3 and NH_4+ indication slowed reverse oxidation in the atmosphere of NH_3 to HNO_3 and NH_4^+. Similarly, the acid chemical distribution at all the Southeast Asia remote sites followed the same pattern as the urban and the rural sites.

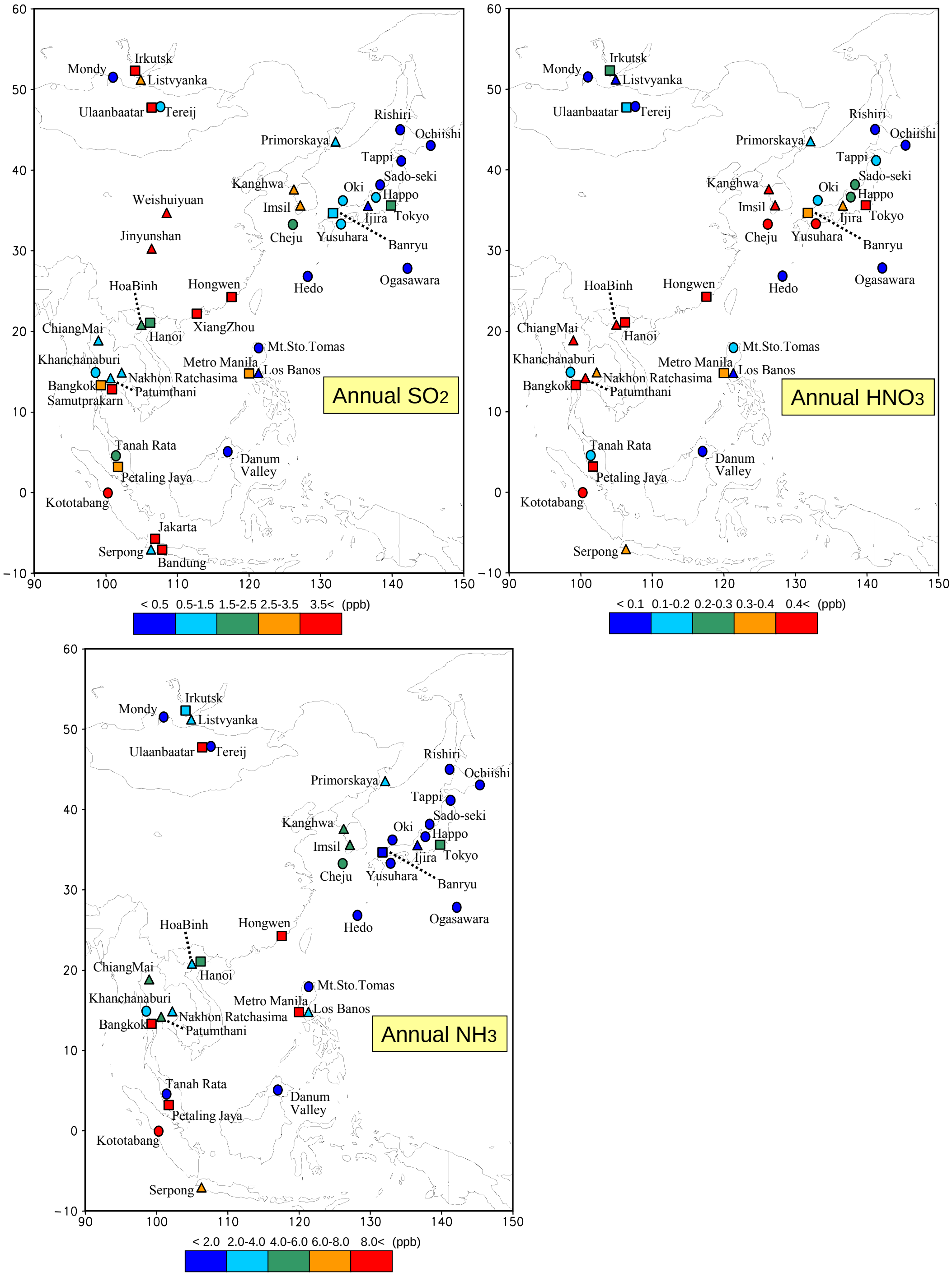


Figure 4.3.1. Spatial variation in gas concentrations (SO_2, HNO_3 and NH_3) at EANET sites. The concentrations show the annual average of 2005-2009. *Square, triangle and circle symbols denote urban, rural and remote sites, respectively.*

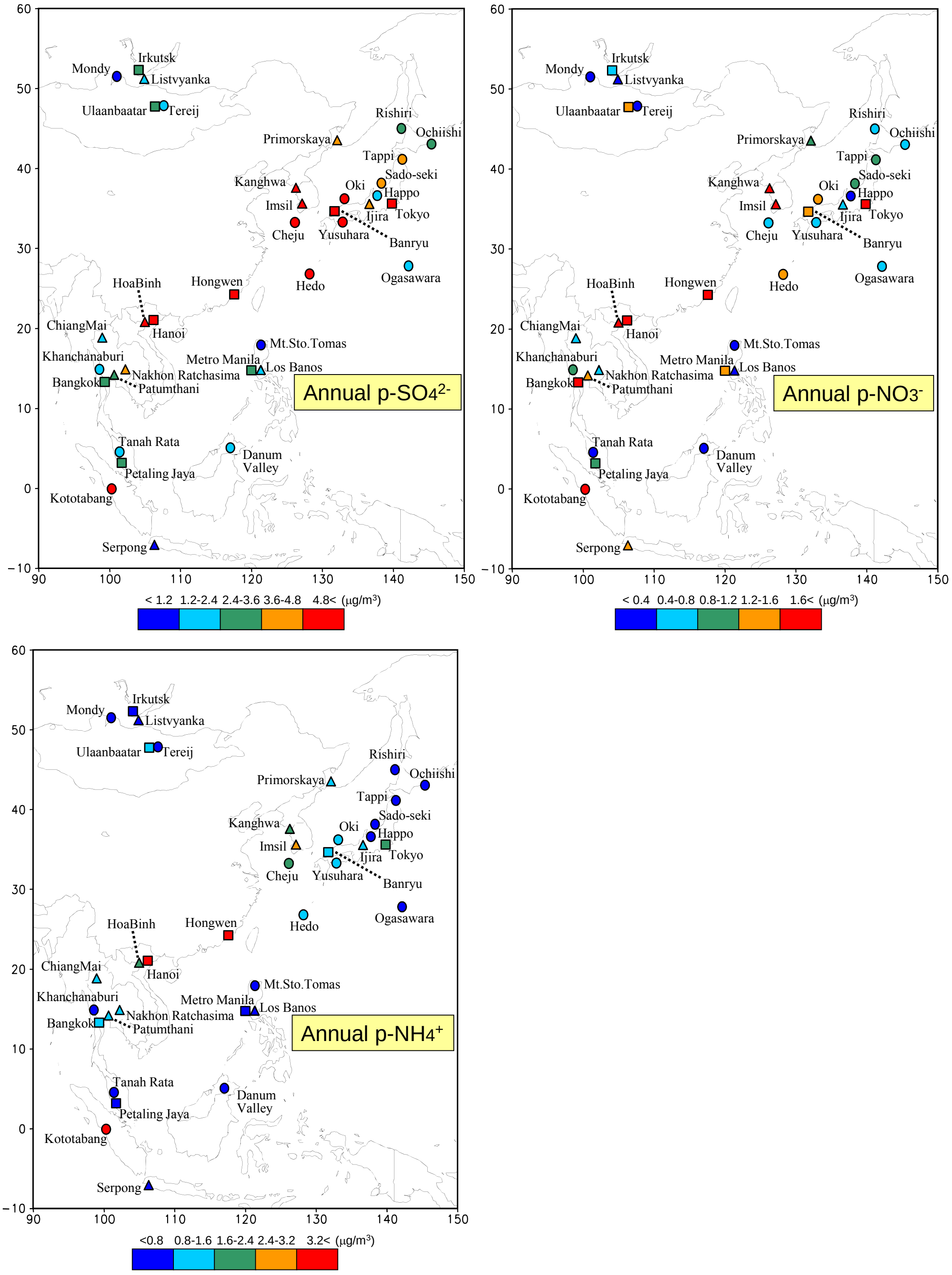


Figure 4.3.2. Spatial variation in aerosol concentrations (SO_4^{2-}, NO_3^- and NH_4^+) at EANET sites. The concentrations show the annual average of 2005-2009. *Square, triangle and circle symbols denote urban, rural and remote sites, respectively.*

NO_2 and NOx concentrations are high in urban sites in Indonesia, and Thailand. Annual O_3 concentrations were low in Thailand. For annual PM_{10} concentrations, moderate PM_{10} concentrations were observed at urban sites in Thailand (Figure 4.3.3).

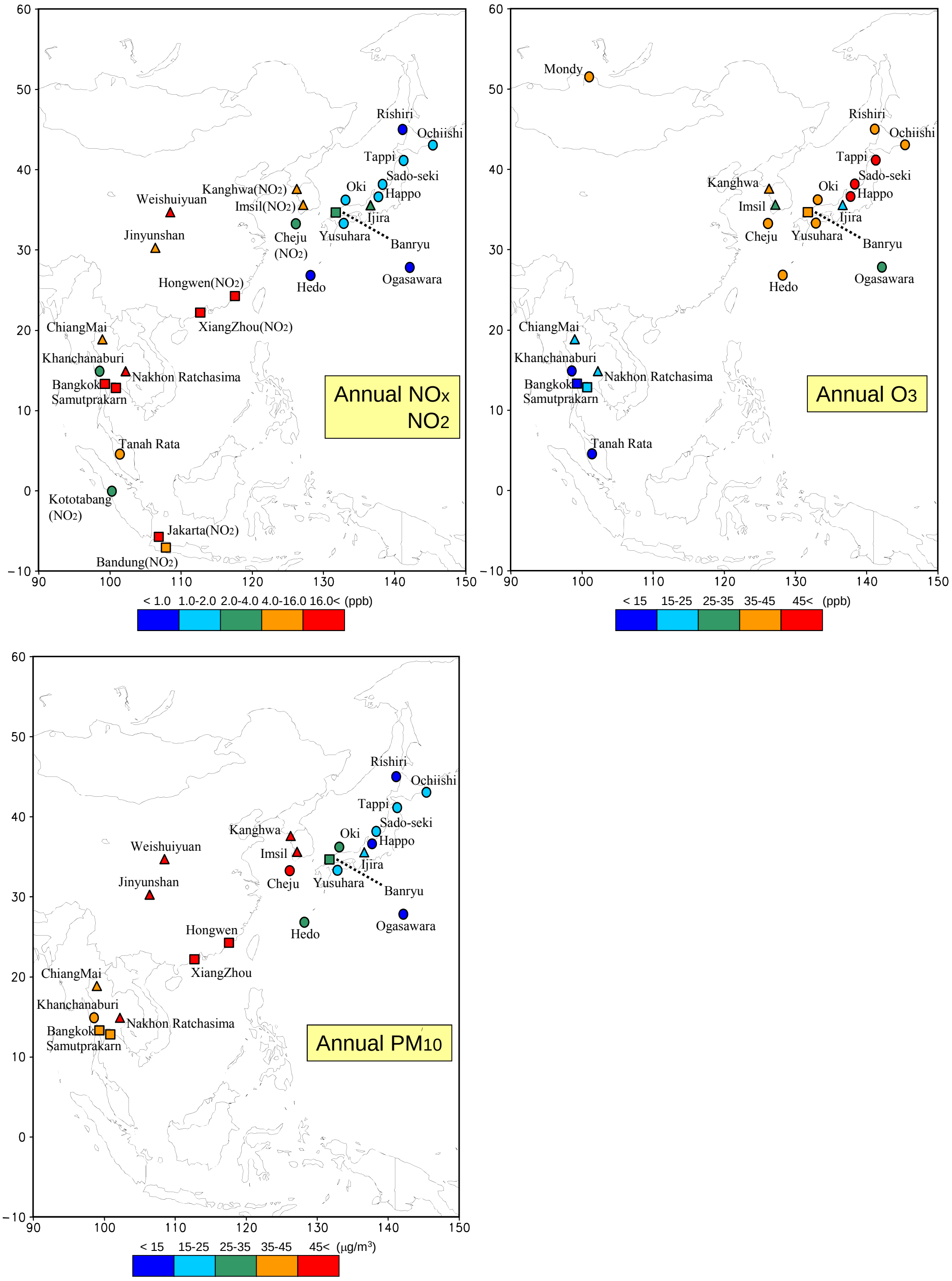


Figure 4.3.3. Spatial variation in gas concentrations (NOx, NO_2, O_3, PM_{10}) at EANET sites. The concentrations show the annual average of 2005-2009. *Square, triangle and circle symbols denote urban, rural and remote sites, respectively.*

4.4 Temporal variation of air concentration

4.4.1 Seasonal characteristic in the Northeast Asia regions

SO_2 and SO_4^{2-} concentrations

The sulphur components at all monitoring sites in China (Xiamen-Hongwen and Xiang Zhou), Mongolia (Ulaanbaartar and Terelj) and Russia (Irkutsk, Listvyanka, Primorskaya and Mondy) show distinctively seasonal cyclic pattern during 2005–2009 (Figures 4.4.1 and 4.4.3). The observed concentrations of SO_2 and SO_4^{2-} are low in the mid-summer and high during the winter period. This indicates high energy utilization during the low temperature climate. The SO_2 concentration in China is seen some temporally decrease but in Mongolia and Russia are fluctuating in the last 5 years. The maximum concentration of SO_2 at the urban site (Irkutsk) in Russia is 4-5 times higher than the remote site (Mondy). In fact the SO_2 concentration in Mondy is quite small in 2009 (0.1 ppb) where there is a significant improvement from the year 2005 (1.6 ppb).

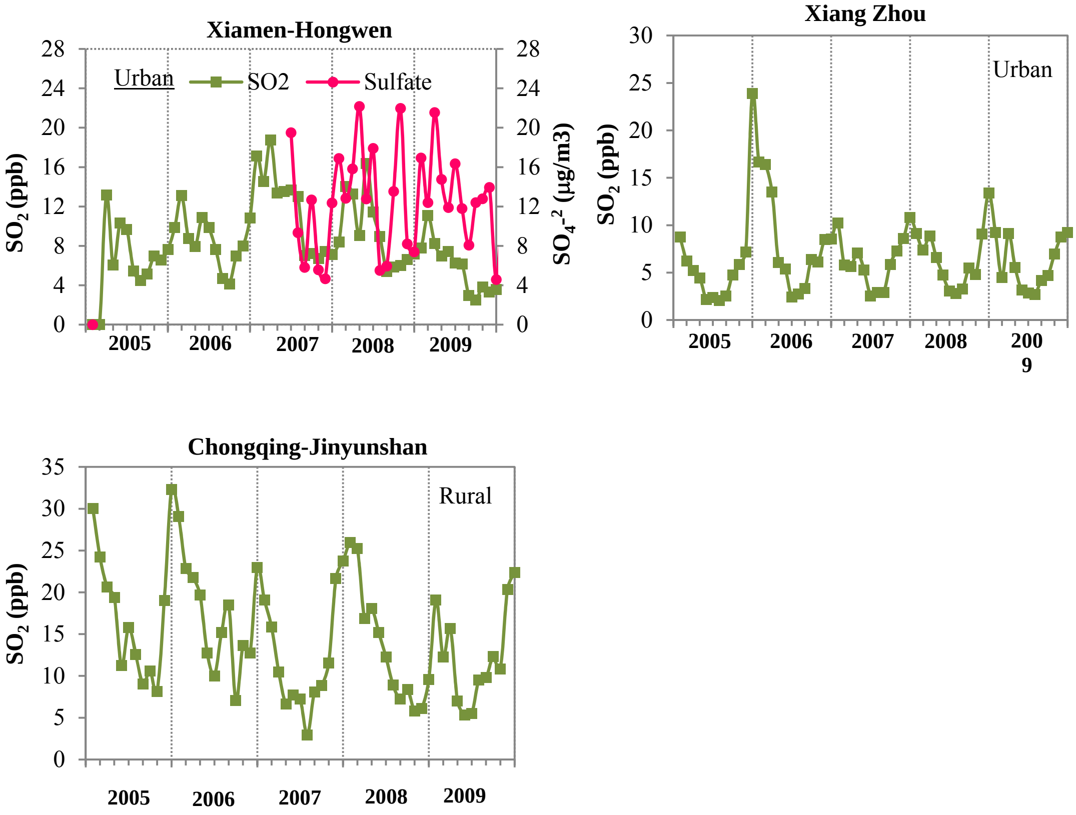


Figure 4.4.1. Seasonal characteristics of SO_2 and sulphate concentrations at urban and rural monitoring sites in China.

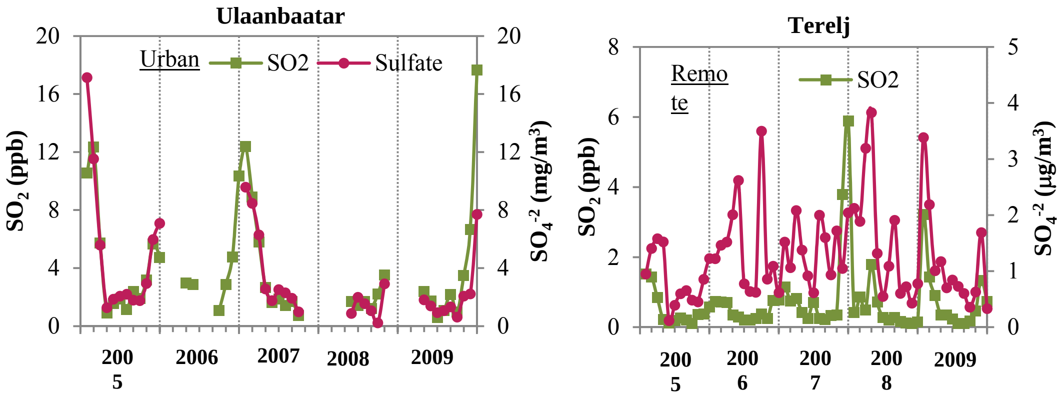


Figure 4.4.2. Seasonal characteristics of SO_2 and sulphate concentrations at urban and remote monitoring sites in Mongolia.

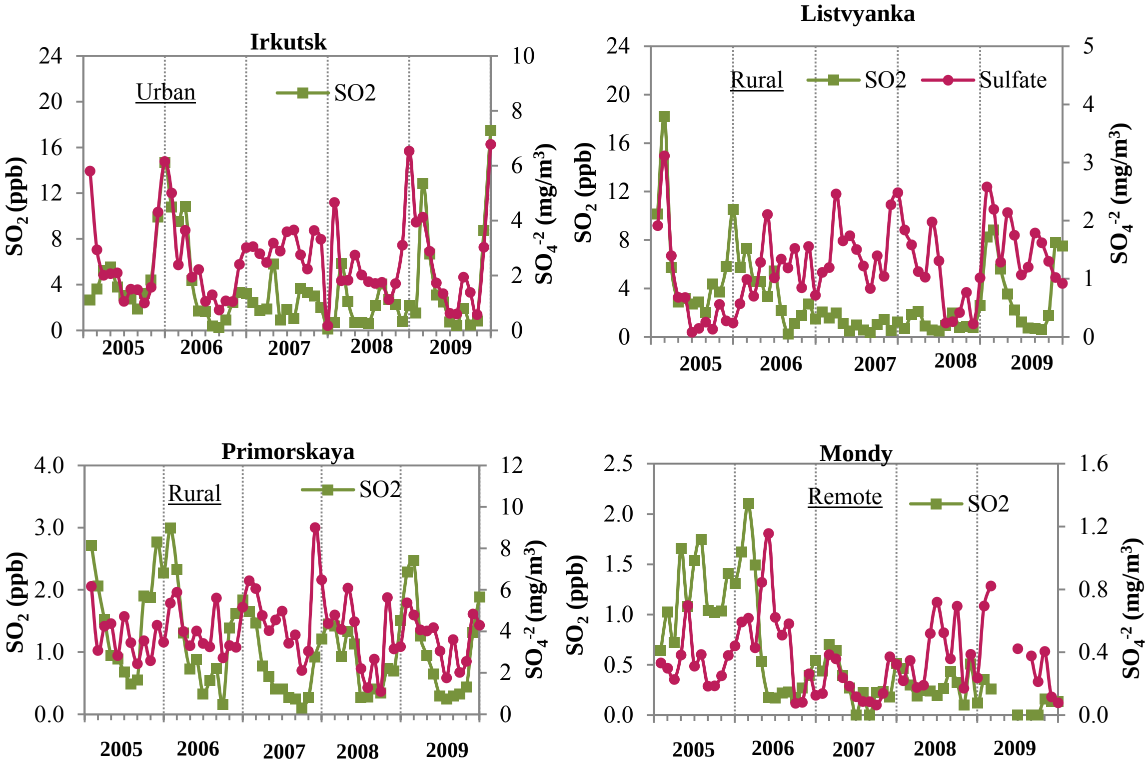


Figure 4.4.3. Seasonal characteristics of SO_2 and sulphate concentrations at urban and rural monitoring sites in Russia.

The Imsil, Ganghwa and Jeju monitoring sites in the Republic of Korea also shows fluctuating cyclic pattern of sulphuric gas and aerosol. The peak of sulphur components reaches a maximum value in the winter (around January) and low in the summer seasons, except in the year 2005 and 2007 (Figure 4.4.4). The SO_2 concentration at Ganghwa in 2005 and 2007 noticeably low in the mid 2007 but the sulphate concentration is oppositely reaching a peak value. Also at Jeju, the remote site, the observed value of SO_2 in 2008 is very low (monthly average of 0.1 ppb) compared with other years while the concentration of sulphate component is exceptionally very high within the same year.

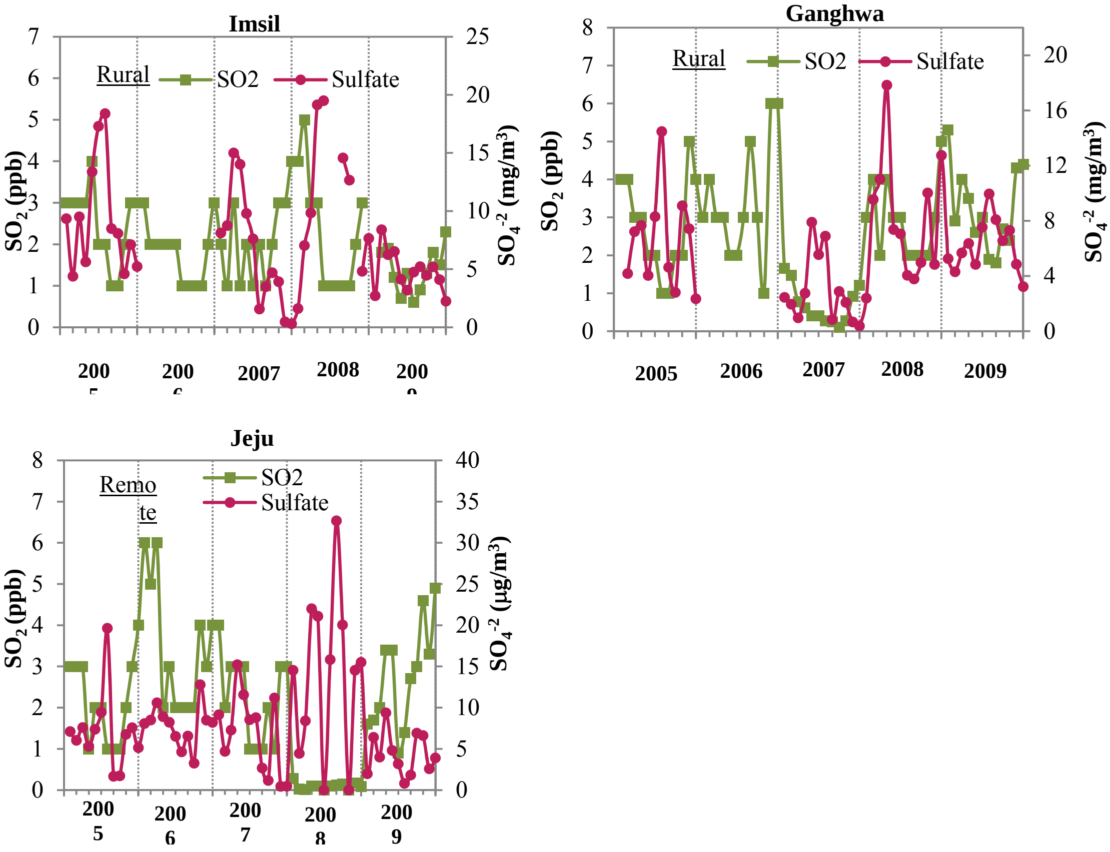


Figure 4.4.4. Seasonal characteristics of SO_2 and sulphate concentrations at rural and remote monitoring sites in the Republic of Korea.

Although the sulphuric gas and aerosol in Japan show similarity with other Northeast Asia sites in which the values respond to seasonal verily, but there was a delay of sulphate peak for 2-3 months after SO_2 reaching the peak. For the remote island site, Ogasawara, the observed SO_2 concentrations are almost all non-detectable during 2005–2009, except the year 2008 (Figure 4.4.5).

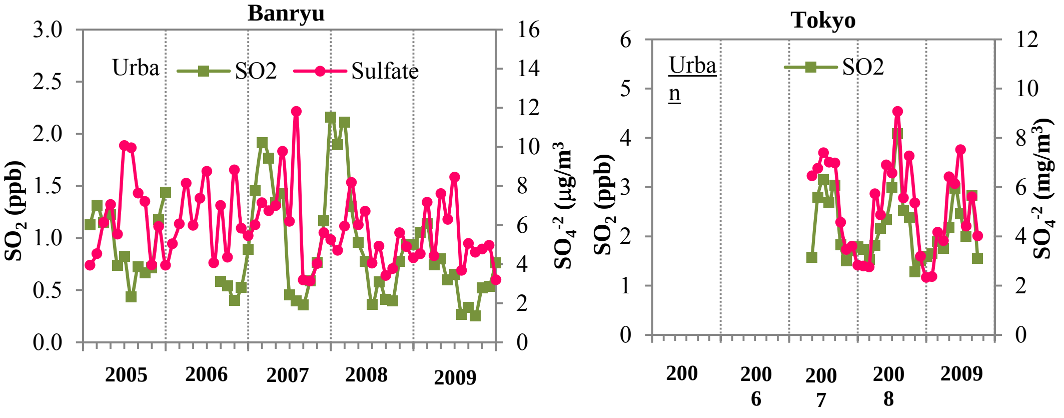


Figure 4.4.5. Seasonal characteristics of SO_2 and sulphate concentrations at urban, rural and remote monitoring sites in Japan.

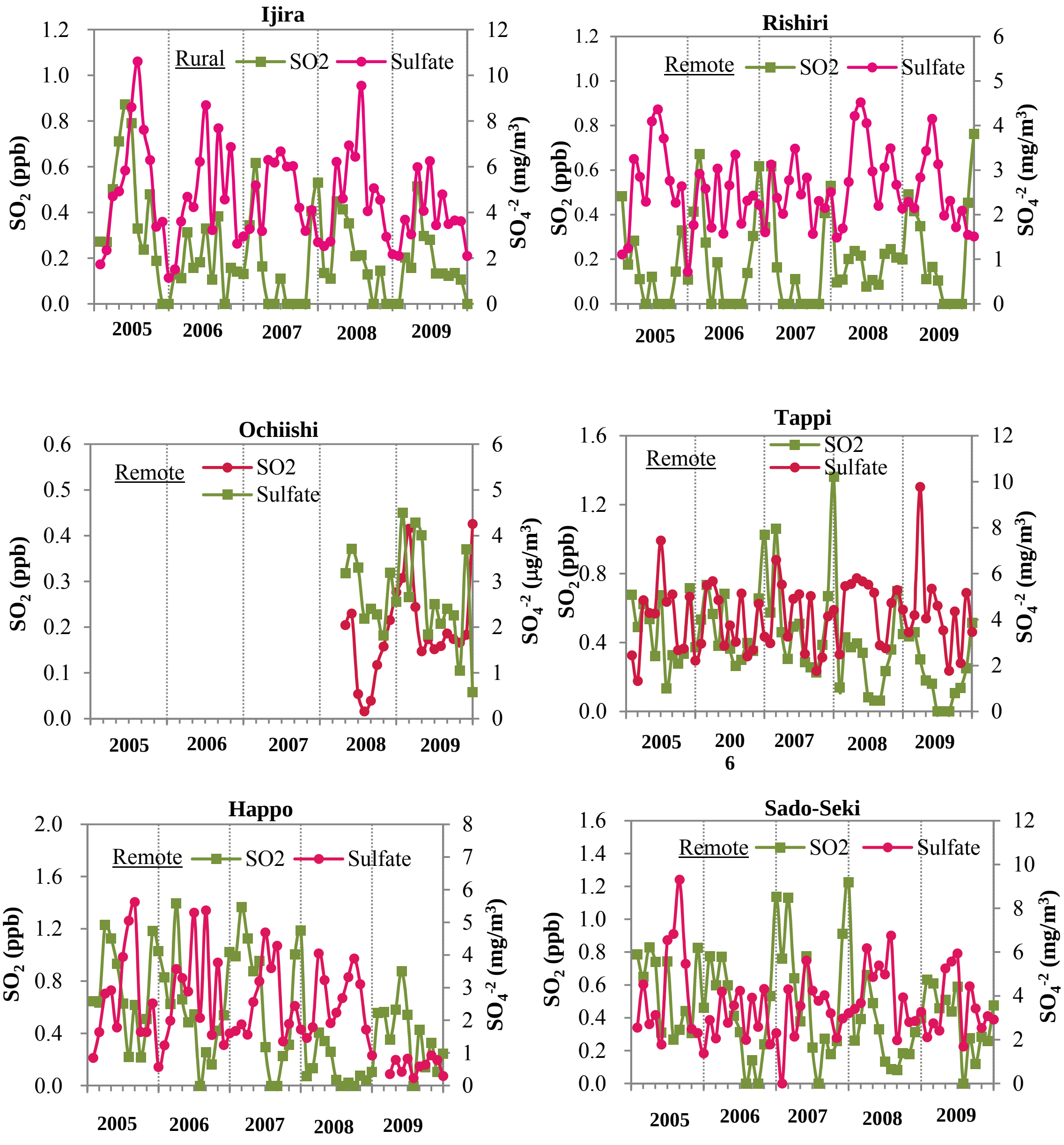


Figure 4.4.5. *(continued).*

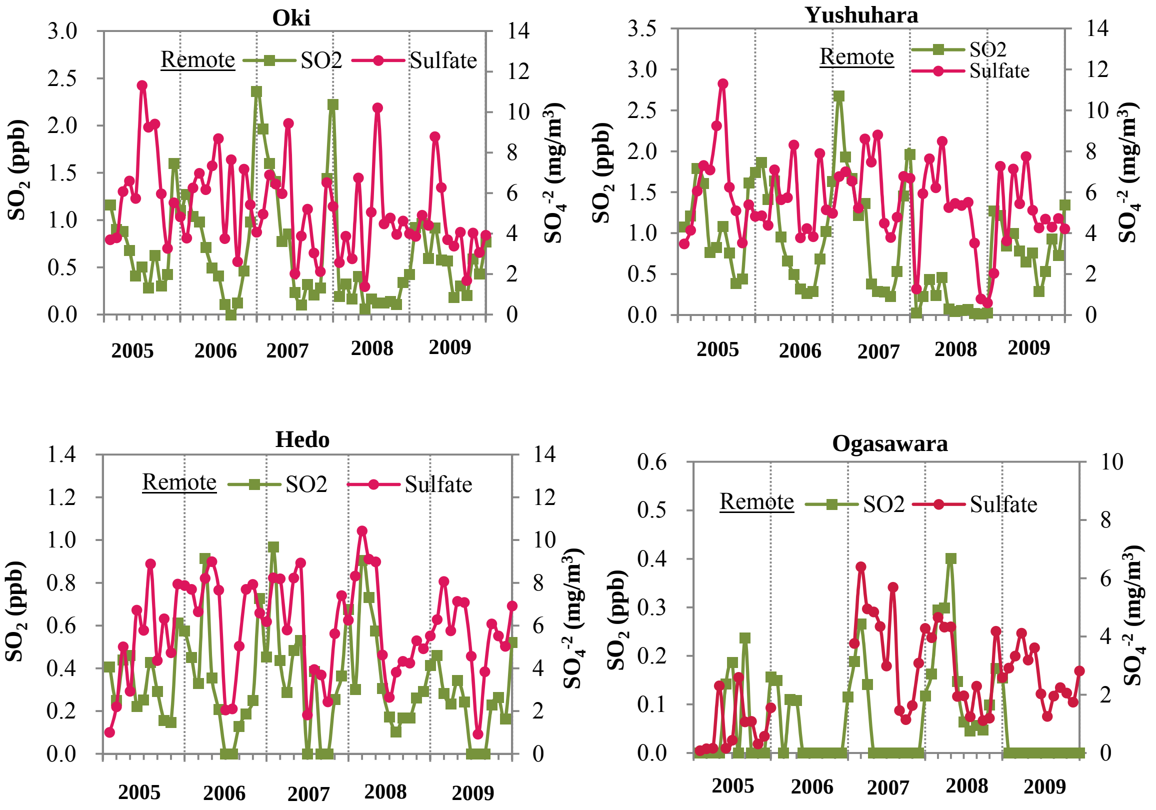


Figure 4.4.5. *(continued).*

HNO$_3$–NO$_3^-$ concentrations

The HNO$_3$ and NO$_3^-$ are fluctuating while seasonal changes. Only at Primorskaya, the rural site shows clear cyclic pattern as the component reached the highest peak in the winter and the minimal values during the mid-summer. The nitrate components are detectably very low at Terelj, Irkutsk, Listvyanka and Primorskaya monitoring sites in certain years (Figure 4.4.6.). Hence, they seem to have no relationship with large existing amount of HNO$_3$. Generally, the NO$_3^-$ concentrations were higher than HNO$_3$ by approximately 3–4 times.

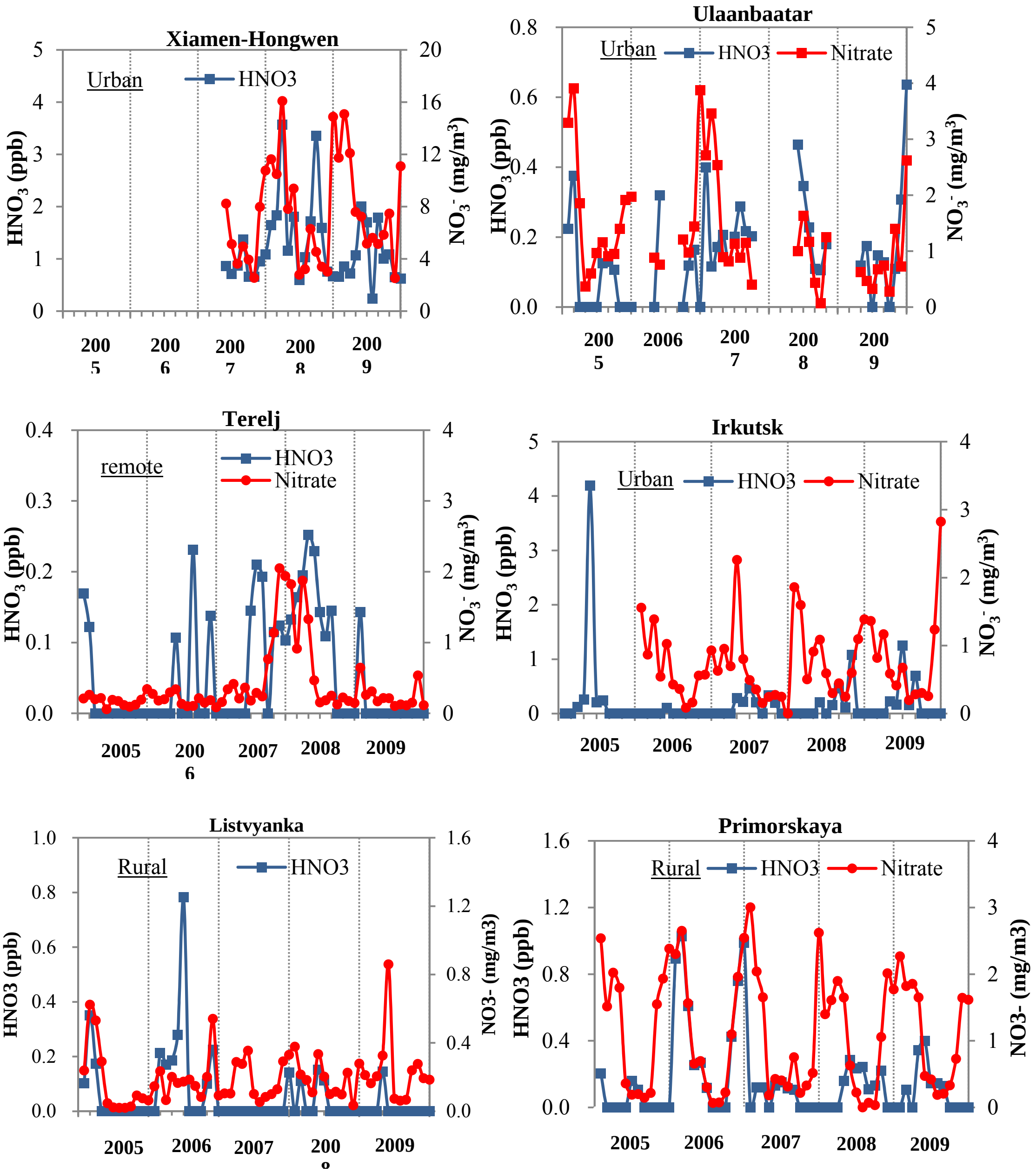


Figure 4.4.6. Seasonal characteristics of HNO_3 and nitrate concentrations at urban, rural and remote monitoring sites in China, Mongolia and Russia.

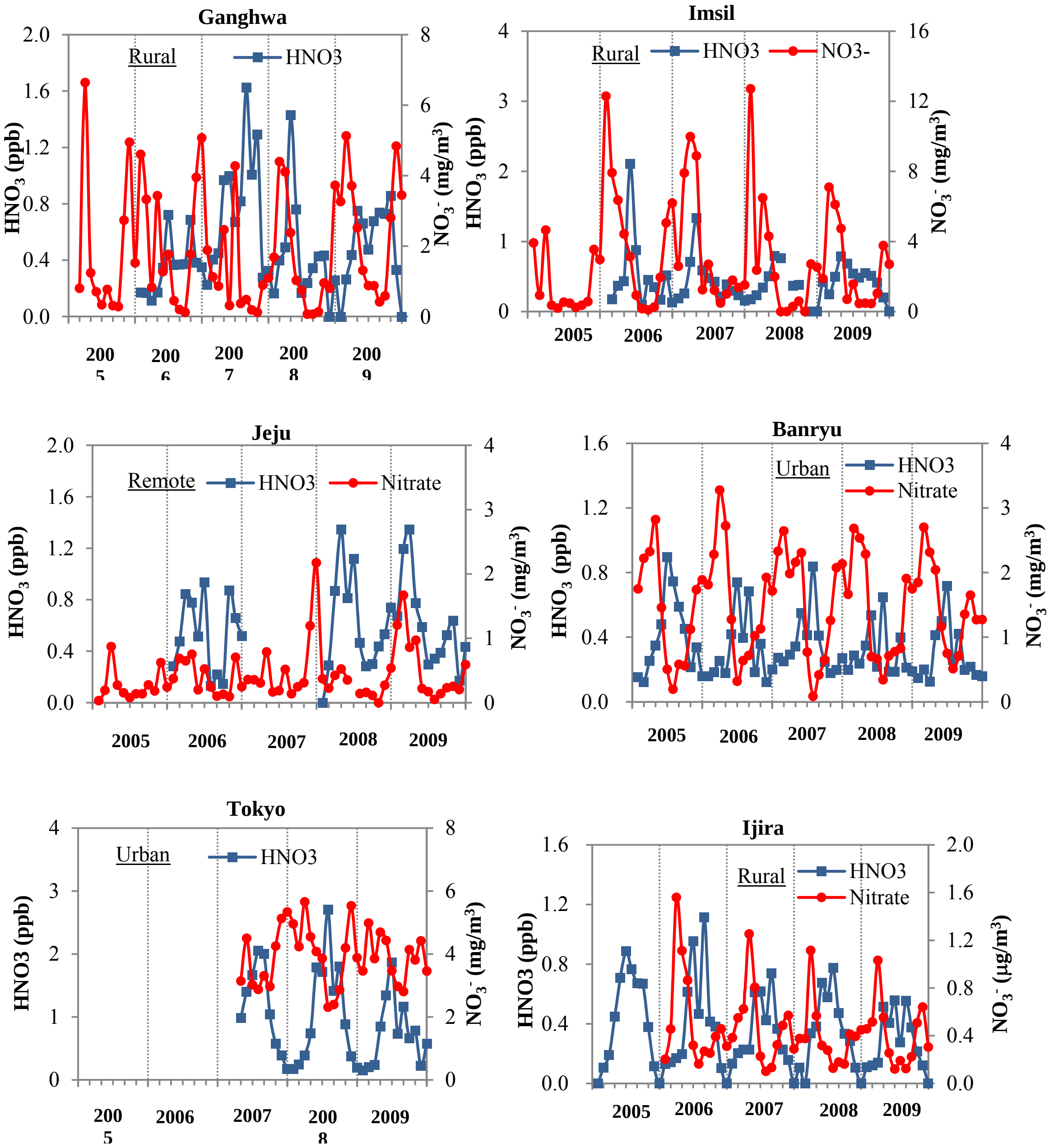


Figure 4.4.7. Seasonal characteristics of HNO$_3$ and nitrate concentrations at urban, rural and remote monitoring sites in the Republic of Korea and Japan.

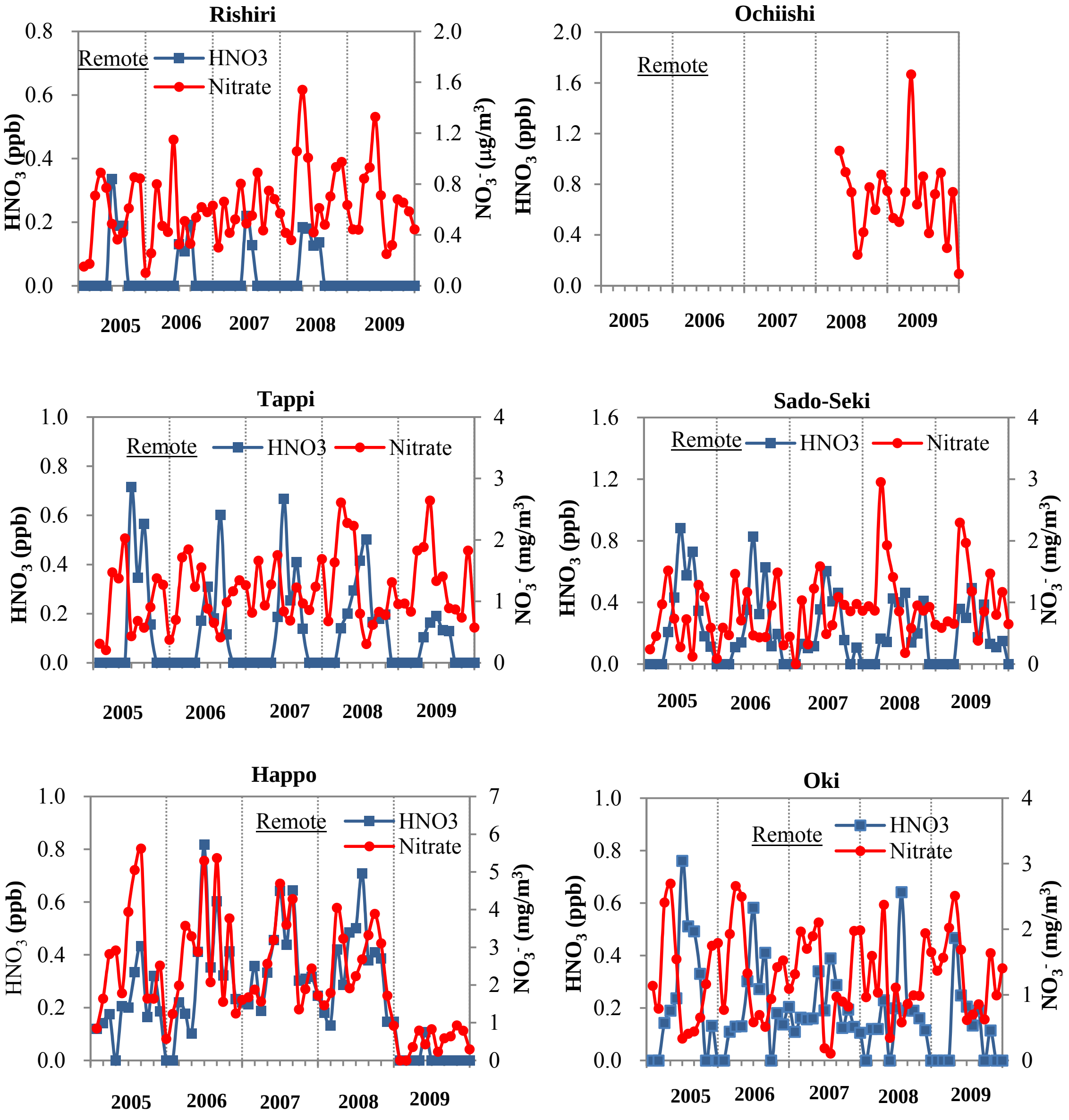


Figure 4.4.7. *(continued).*

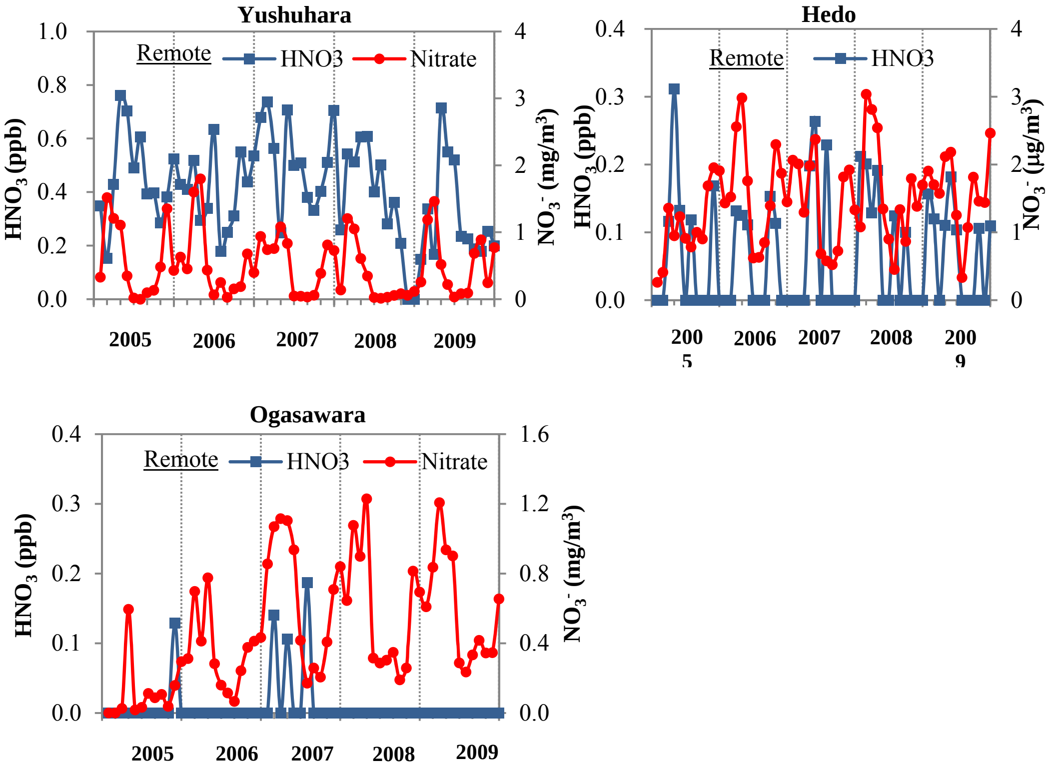


Figure 4.4.7. *(continued).*

NH_3–NH_4^+ concentrations

There is a clear pattern of seasonal variation between NH_3 and NH_4^+ at most of northern inland Asia, except Russia (at Listvyanka, Primorskaya and Mondy) (Figure 4.4.8). The concentrations of NH_3 were high in the winter and decreased to very low values in the summer while the concentrations of NH_4^+ showed in the opposite trend. High release of NH_3 during the warm season was due to agricultural activities. For high NH_4^+ in the winter was due to slow biological reaction. It is also observed that the amounts of NH_3 and NH_4^+ release are not much different among the urban, rural and remote sites.

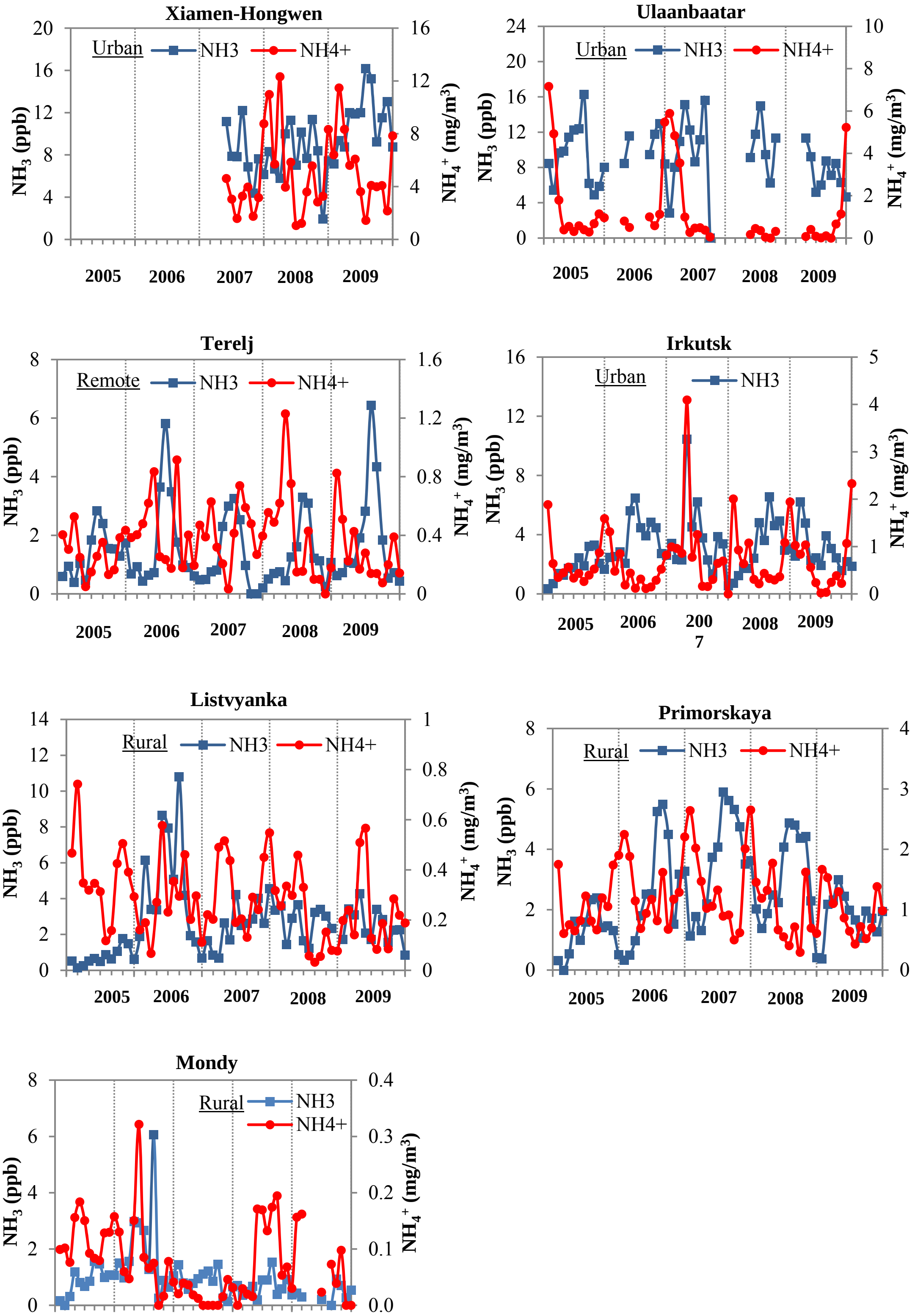


Figure 4.4.8. Seasonal characteristics of NH_3 and ammonium concentrations at urban, rural and remote monitoring sites in China, Mongolia and Russia.

It is interesting to see an opposing trend of seasonal variation of NH_3 and NH_4^+ concentrations at the monitoring sites in the pacific island from the inland sites. The observation shows that both chemical species, NH_3 and NH_4^+ are high in the winter and low in the summer. In terms of quantities, both components at all monitoring sites in the Republic of Korea and Japan are not significantly distinguishable among the urban, rural and remotes sites (Figures 4.4.9 and 4.4.10).

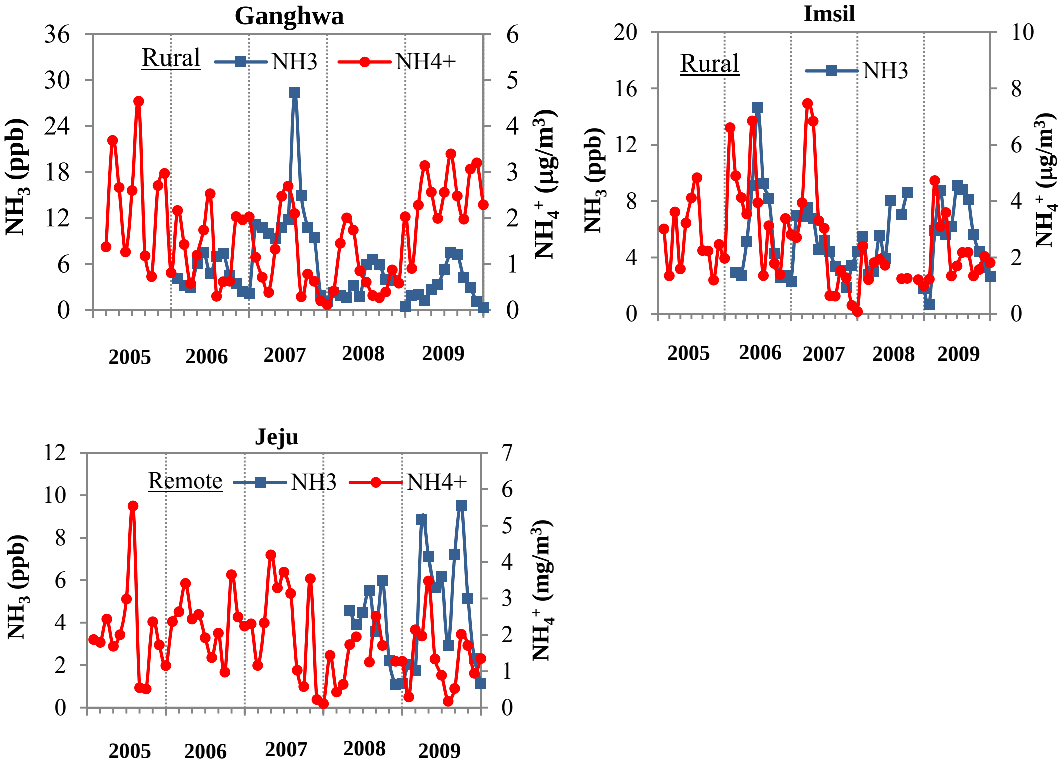


Figure 4.4.9. Seasonal characteristics of NH_3 and ammonium concentrations at urban, rural and remote monitoring sites in the Republic of Korea.

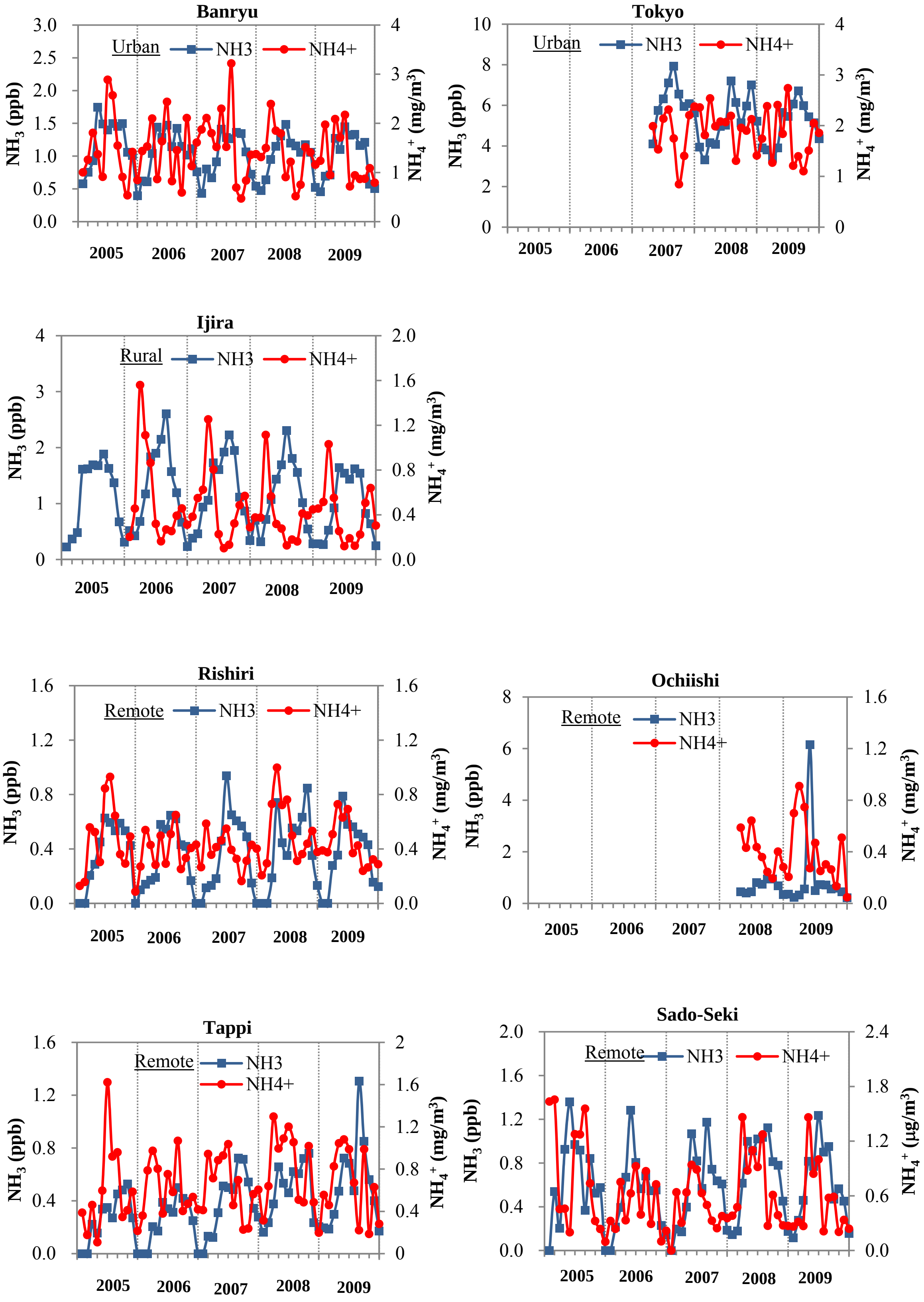


Figure 4.4.10. Seasonal characteristics of NH$_3$ and ammonium concentrations at urban, rural and remote monitoring sites in Japan.

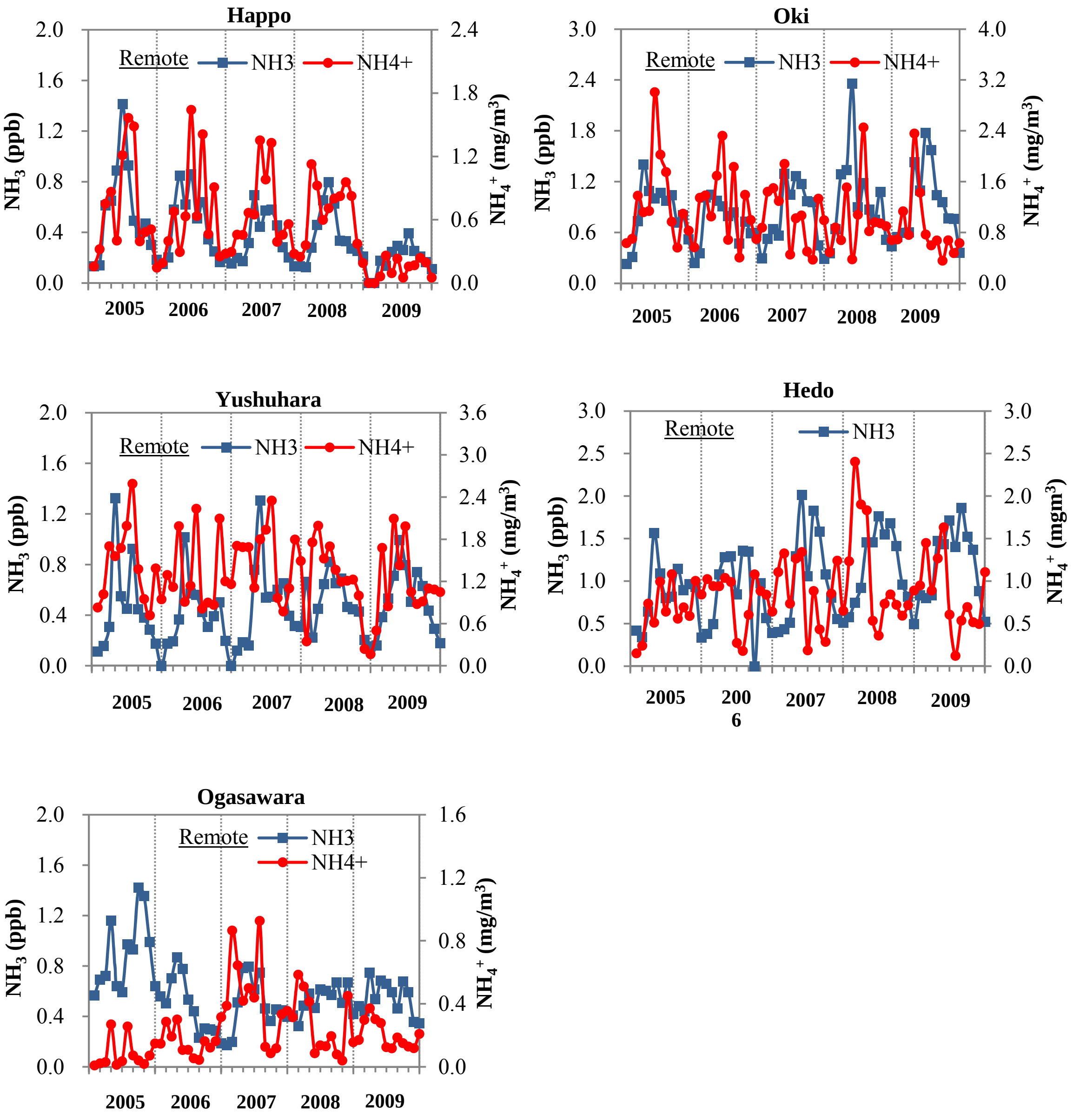


Figure 4.4.10. *(continued).*

NO_2, NOx. O_3, PM_{10} concentrations

Figure 4.4.11 shows annual variations of NO_x or NO_2 concentration at EANET sites in China, the Republic of Korea and Japan from 2000-2009. Concentration levels at Chinese sites have remained high level though out these period. However, NO_2 concentrations at Xiang Zhou and Hongwen showed decreasing trends especially since 2006. No specific trends in Japan was observed, but the average NO_x concentration at air quality monitoring stations in Japan has been reported to decrease.

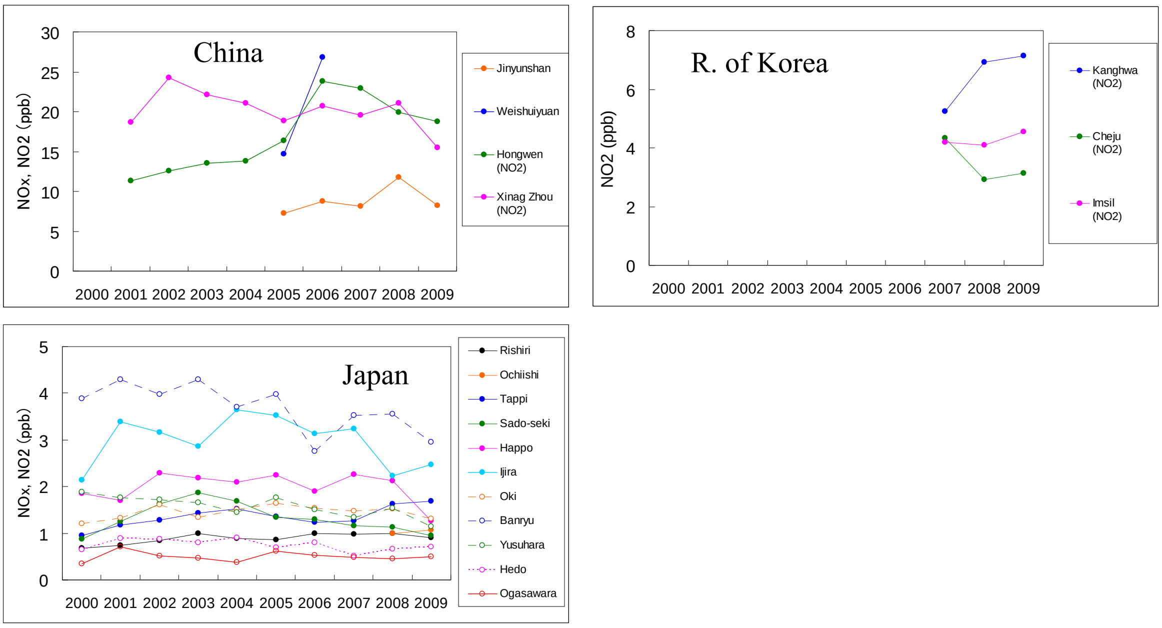
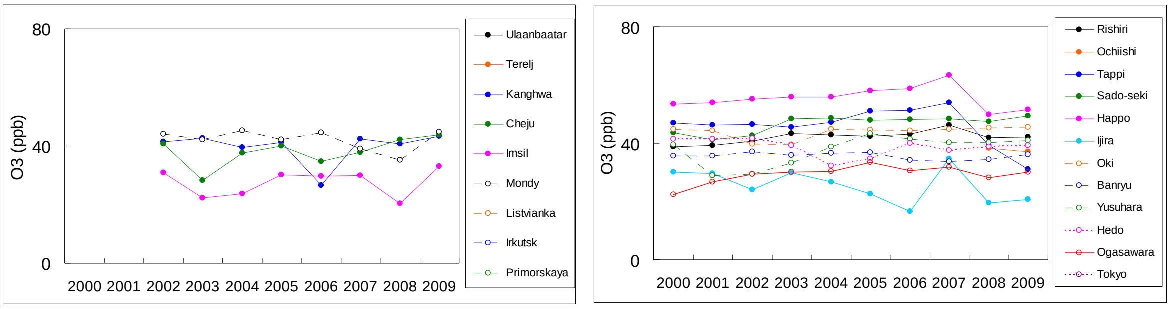


Figure 4.4.11.Annual variations of NOx or NO_2 concentration in China, the Republic of Korea and Japan from 2000 -2009.

Figure 4.4.12 shows annual variations of O_3 concentration at EANET sites in the Republic of Korea and Japan from 2000-2009, and Figure 4.4.13 shows comparison of 5 year average monthly O_3 concentrations between the former (1999-2004) and the latter (2005-2009) 5 years at Japanese EANET sites. The common seasonal trend was observed in Japanese sites. Namely, higher in spring, Lower in summer, Slightly higher in autumn. As previously mentioned, higher O_3 conc. was observed at the Happo (mountain) site because of effect from the stratosphere in the alpine and partially long range transportation of air pollutants. In the pacific ocean region, O_3 conc. in summer was very low because of the clean Pacific air mass. Briefly, increasing trend of O_3 concentrations was observed at many Japanese sites.


Mongolia, R. of Korea, Russia

Japan

Figure 4.4.12. Annual variations of O_3 concentration in Russia, Mongolia, the Republic of Korea, Russia and Japan from 2001-2009.

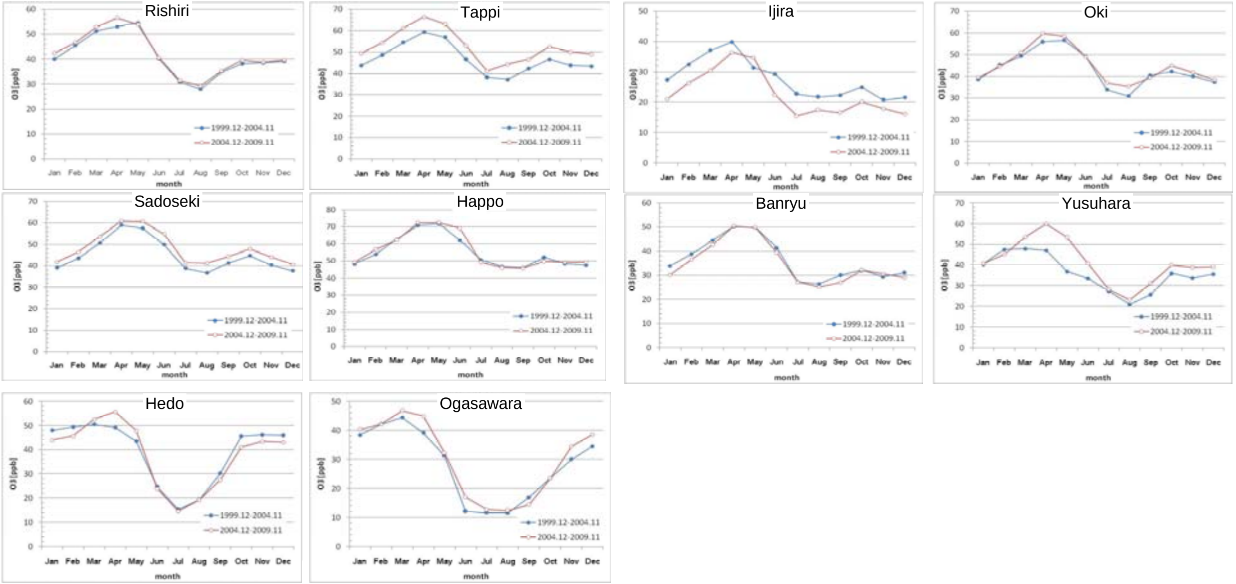
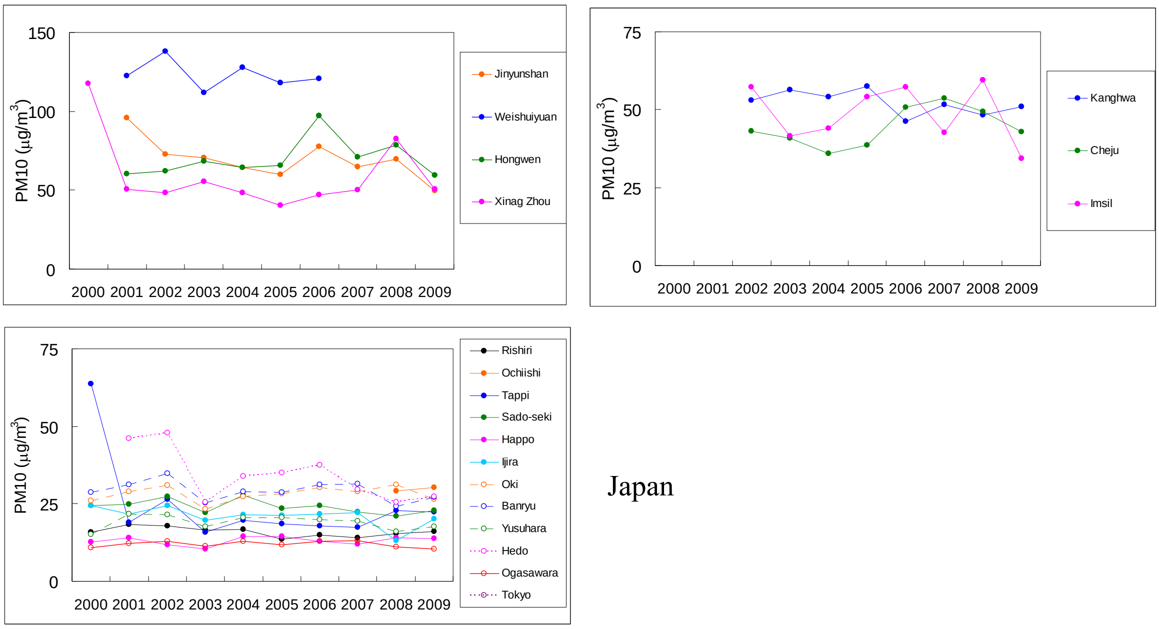


Figure 4.4.13. Comparison of 5 year average monthly O$_3$ conc. between the former (1999-2004) and the latter (2005-2009) 5 years at Japanese EANET sites.

Except Ijira (rural) and Banryu (urban), the average monthly O$_3$ conc. in the latter 5 years (2005-2009) was higher than those in the former 5 years (2000-2004). Increasing in spring was remarkable at Sea of Japan (Rishiri, Tappi and Oki) and Pacific (Yusuhara, Hedo, Ogasawara) sites. Seasonal trend did not change between the former and the latter 5 years.


China

R. of Korea

Japan

Figure 4.4.14. Annual variations of PM$_{10}$ concentration in China, the Republic of Korea and Japan from 2001-2009.

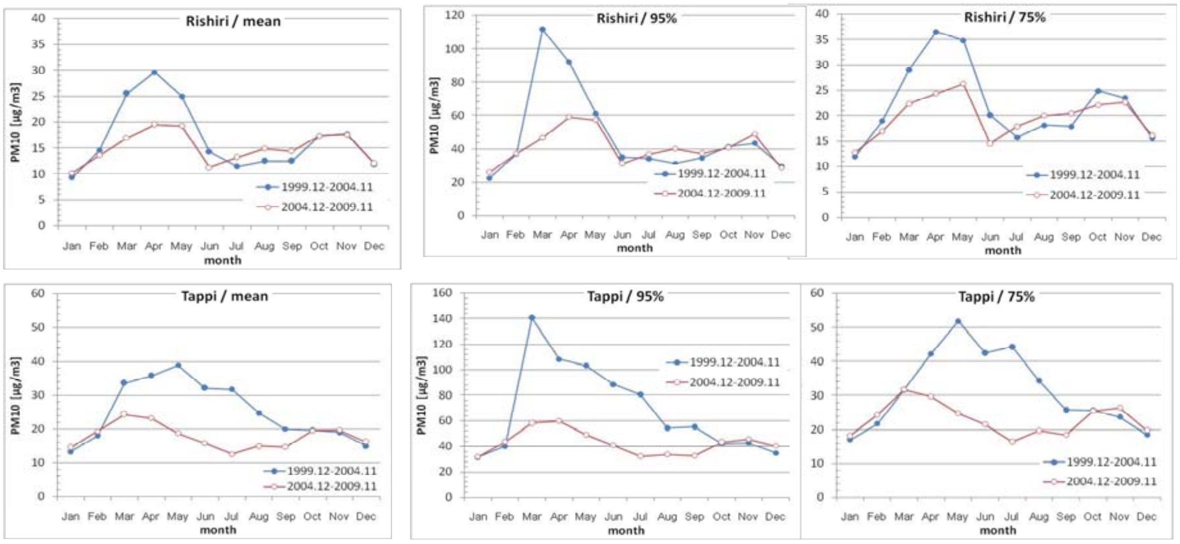


Figure 4.4.15. Comparison of 5 year monthly average, 95 and 75 percentile PM_{10} conc. between the former (1999-2004) and the latter (2005-2009) 5 years in Japanese EANET sites.

Figure 4.4.14 shows annual variations of PM_{10} concentration at EANET sites in China, the Republic of Korea and Japan from 2001-2009, and Figure 4.4.15 shows comparison of 5 year average monthly average, 95 and 75 percentile PM_{10} concentrations between the former (1999-2004) and the latter (2005-2009) 5 years at Japanese EANET sites. Concentration levels of PM_{10} at Chinese sites have remained high level though out these period. However, PM_{10} concentrations at all Chinese sites showed decreasing trends especially since 2006. This observation may imply recent air quality improvement.

At most of Japanese sites, the average monthly PM_{10} concentrations in the latter 5 years (2005-2009) were lower than those in the former 5 years (2000-2004). Decrease in high percentile concentrations in spring season is mainly attributable to monthly and annual average conc.

4.4.2 Seasonal characteristic in the Southeast Asia regions

SO_2 and SO_4^{2-} concentrations

All urban sites in EANET Southeast Asia region show significant improvement of SO_2 and SO_4^{2-} emission reduction during 2005-2009. This could have been linked to the pollution reduction policy applied in most of the cities in Southeast Asia. A significant reduction of SO_2 can be seen by comparing the average measured values in 2005 with 2009 as follow: Bangkok site 7.3/1.9; Hanoi site 2.4/1.3; Hoa Binh site 2.7/0.9; Metro Manila site 2.7/0.9 (in 2008); Petaling Jaya site 3.8/1.8 and Jakarta site 11.0/8.3. The related chemical species, i.e. SO_4^{2-} are decreasing also during 2005–

2009 in approximately the same magnitude as SO_2 for all the urban sites, except Bangkok site. The reduction trend of SO_4^{2-} at Bangkok is not clearly distinguished (Figure 4.4.16).

Concentrations of SO_2 and SO_4^{2-} at the rural and remote sites are likely to be seasonal independency. Since climatology in Southeast Asia is invariant, explicitly the countries near equator, i.e. Malaysia and Indonesia. Therefore, it is presumably that fluctuations in ambient concentrations are derived from mobile and industrial emissions. The effect of sea salt sulphate is likely to be small (the calculated results based on Cl^- concentrations are not shown in this chapter).

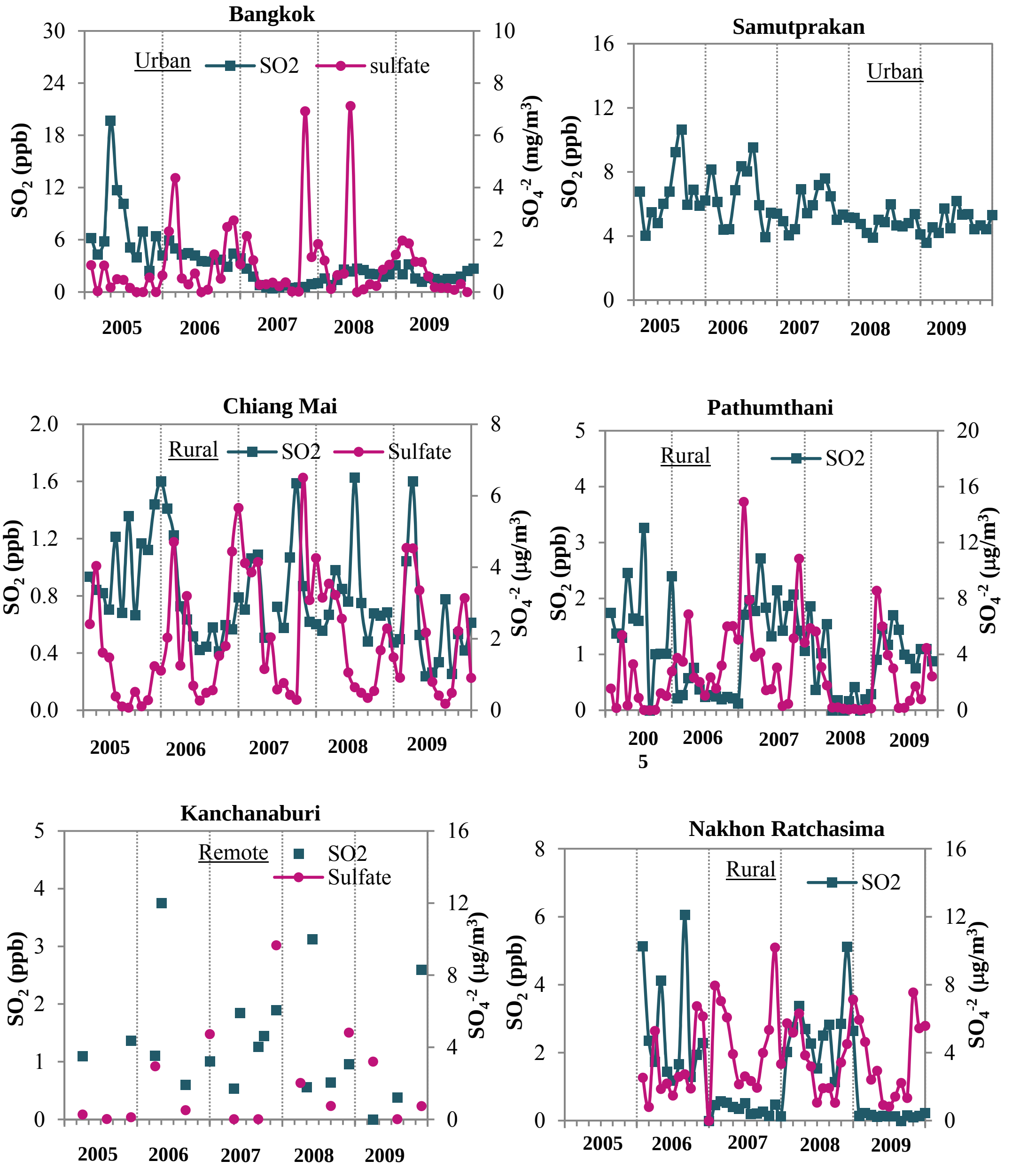


Figure 4.4.16. Seasonal characteristics of SO_2 and SO_4^{2-} concentrations at urban and rural monitoring sites in Thailand, Viet Nam, Malaysia, Indonesia and Philippines.

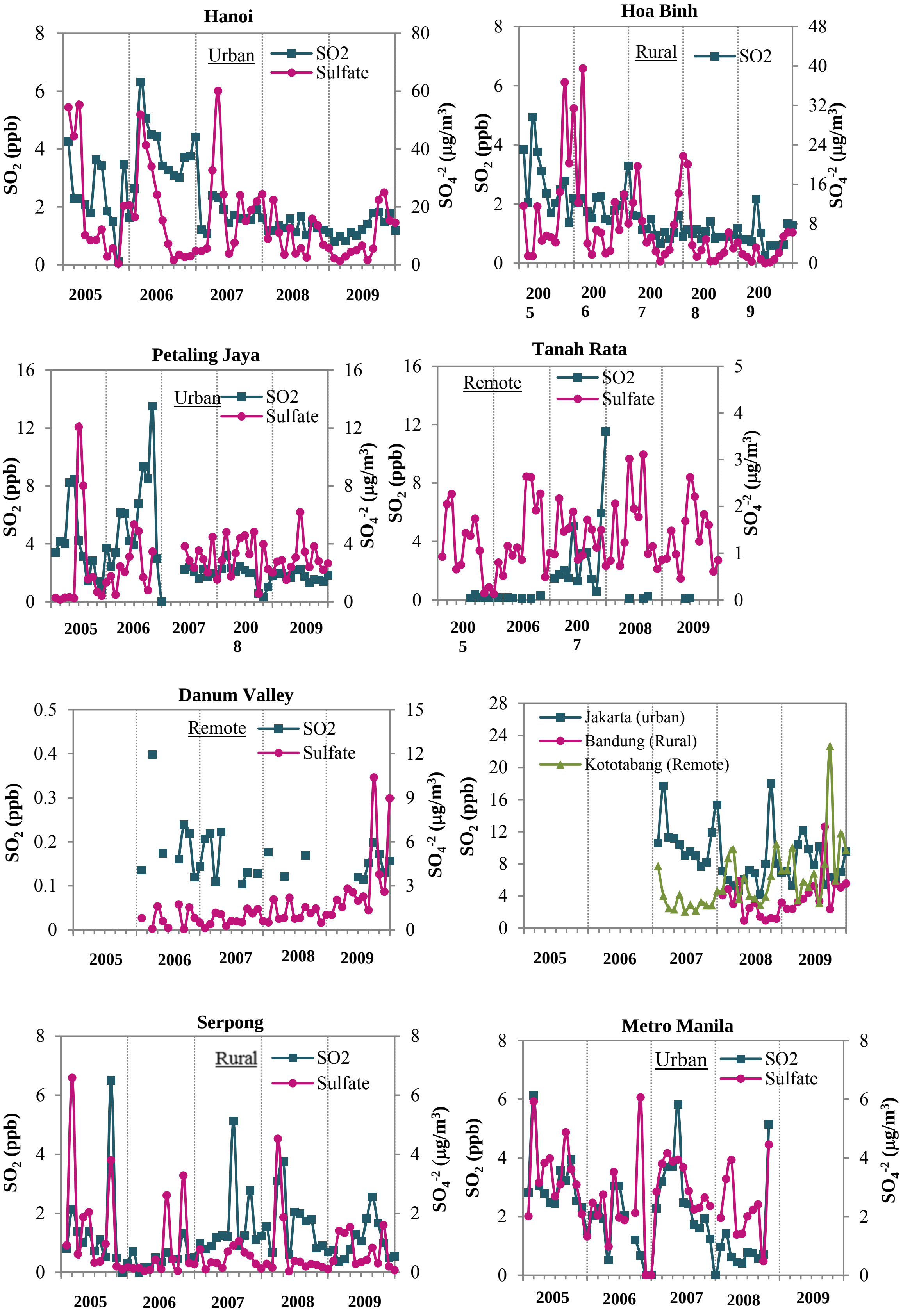


Figure 4.4.16. *(continued).*

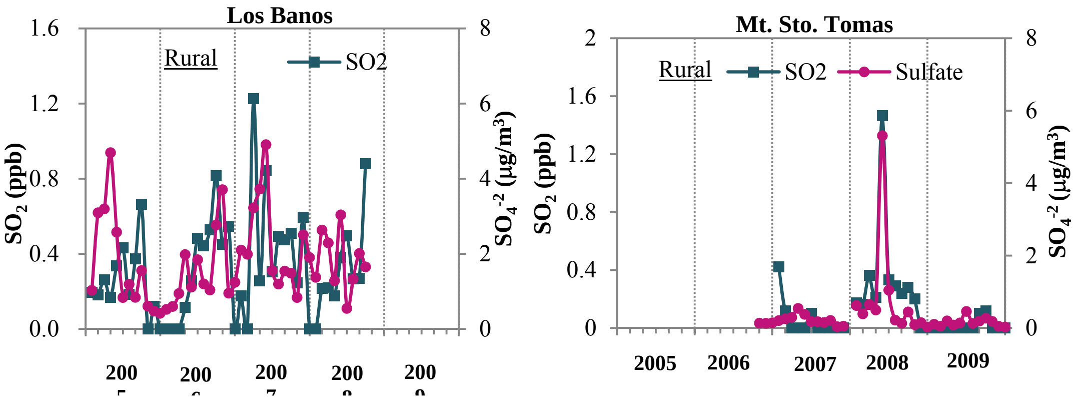


Figure 4.4.16. *(continued).*

HNO_3 and NO_3^- concentrations

The concentrations of HNO_3 and NO_3^- at the monitoring site located inland (Thailand and Vietnam) are more affected by seasonal variation than the sites located close to the Pacific Ocean. This is due to the inland climate that can be distinguished to dry and wet seasons. The observed concentrations can be seen low during the hot and wet season (around the mid-year) and high during the cool and dry season (around end of the year). The low concentrations are believed to cause by heavy monsoon rain washed out. For the countries close to the Pacific Ocean, they are under the influence of rained forest climatology (Malaysia, Indonesia and the Philippines) in which the ambient temperature are not varied appreciably plus long monthly rain falls. The seasonal fluctuations in concentration are not observed clearly. It should be noted that the HNO_3 and NO_3^- at the remote site in Mt. Sto. Tomas was very high in 2008. For the remote site in Kanchanaburi, the concentrations were monitored only three times a year (in March, July and November) for its practicality to move a monitoring mobile to the site located in high mountain area.

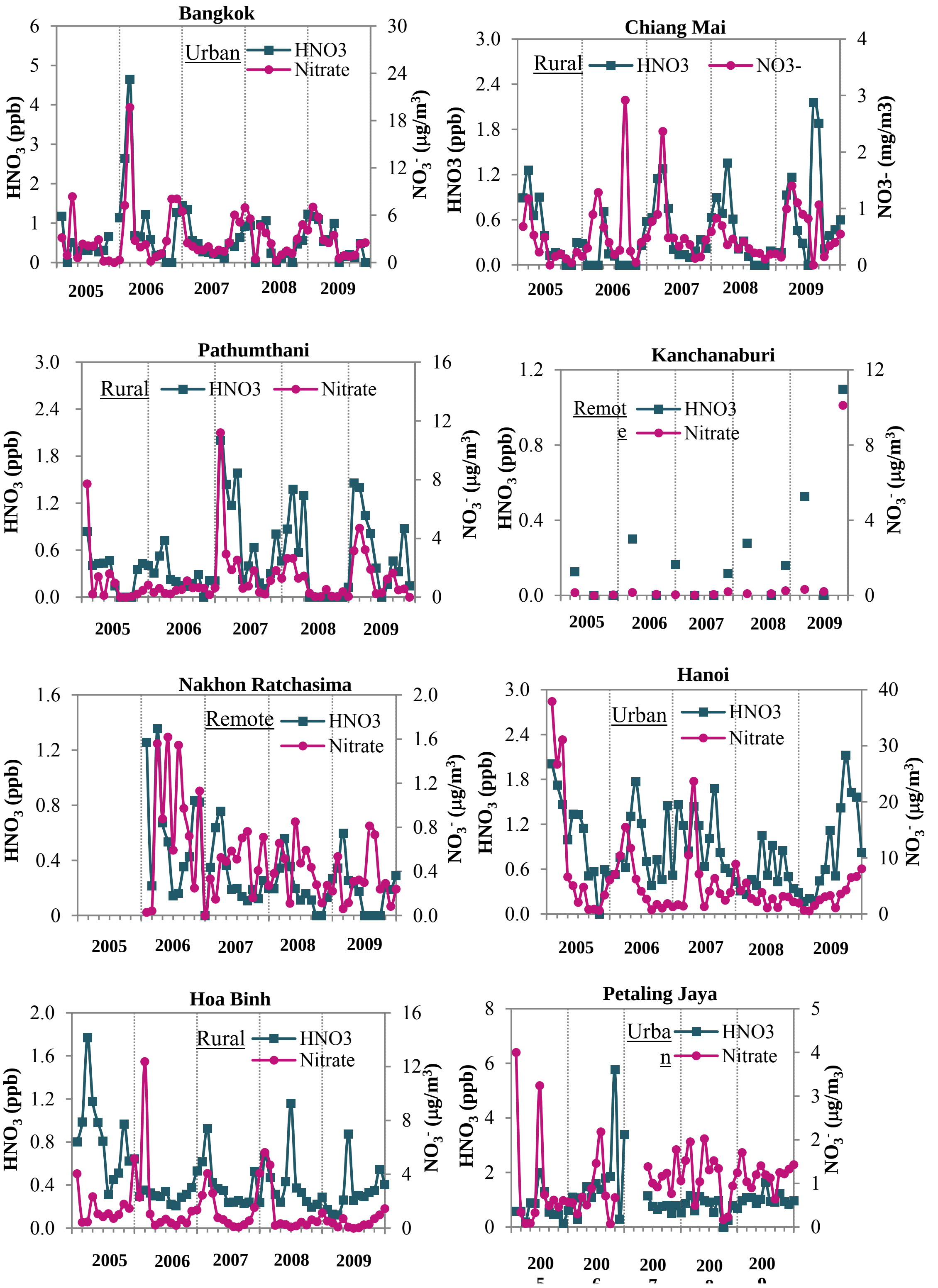


Figure 4.4.17. Seasonal characteristics of HNO_3 and NO_3^- concentrations at rural and urban monitoring sites in Thailand, Viet Nam, Malaysia, Indonesia and Philippines.

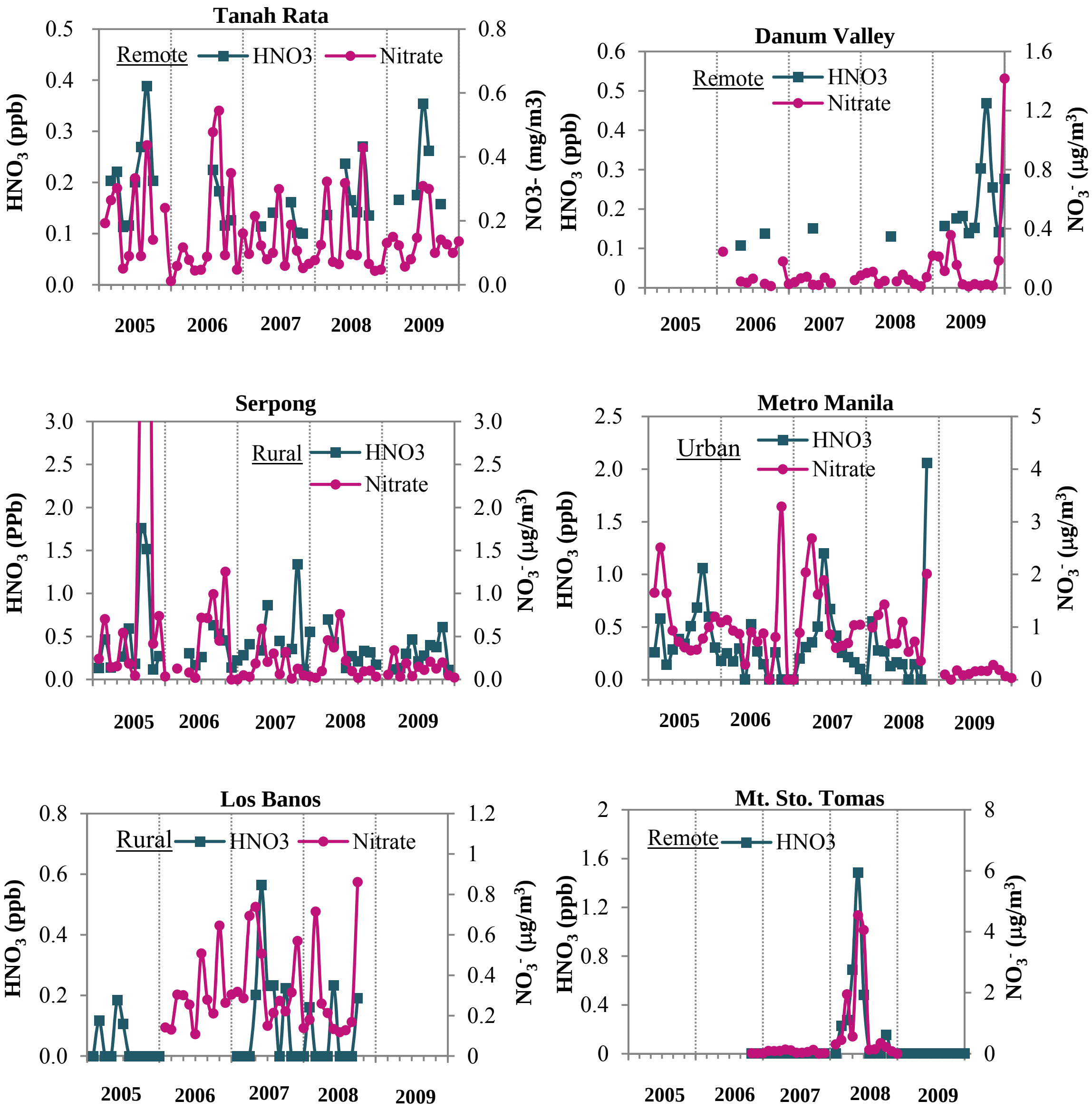


Figure 4.4.17. *(continued).*

NH_3–NH_4^+ concentrations

The observed concentration of NH_3 and NH_4^+ at urban, rural and remote sites in Thailand followed seasonal variation pattern influencing by tropical climate rather that under the influence of agricultural cultivation cycles (Figure 4.4.18.). That is high concentrations found in dry season and very low in wet seasonal though the cultivations are carried out throughout the year because of the all year round of warm climate. It is observed that at the remote site (Nakhon Ratchasima), NH_3 concentration is very low comparing with NH_4^- concentration. This indicates less impact of urban pollutant crated by photochemical reaction.

The monitoring sites at Hanoi urban site and Hoa Binh rural site in Vietnam also show seasonal variation cycle in 2005 and 2006. However, the concentration of NH_3 in 2007-2009 remains constantly high throughout the year while the concentration of NH_4^+ is noticeably low. This indicates an expansion of city communities in the recent years and consequently the effects of photochemical reaction become evident.

For the sites located near the Pacific Ocean, seasonal variations of chemical concentrations cyclic pattern although are not clearly observed as in the case of Thailand and Vietnam but can be seen at Petaling Jaya urban site, Tanah Rata rural site and Serpong rural site. The concentrations of NH_3 and NH_4^+ found high during the midyear and low toward the end of the year. The cycle pattern is differed from the inland Southeast Asia in which the heavy rain fall occurs in June and July. Because of the typical climatology of the region lies about the equator, the heavy rain fall in Malaysia and Indonesia occurs in November and December. It should be noted that the chemical concentration at Danum Valley remote site has increased substantially toward the end of year 2009.

The concentrations of NH_3 and NH_4^+observed at Metro Manila urban site, Los Banos rural site and Mt. Sto. Tomas remote site in the Philippines are generally not seasonal variably during 2005–2009. Only in the year 2008, NH_3 and NH_4^+ are observed to be exceptionally high at certain time of the year.

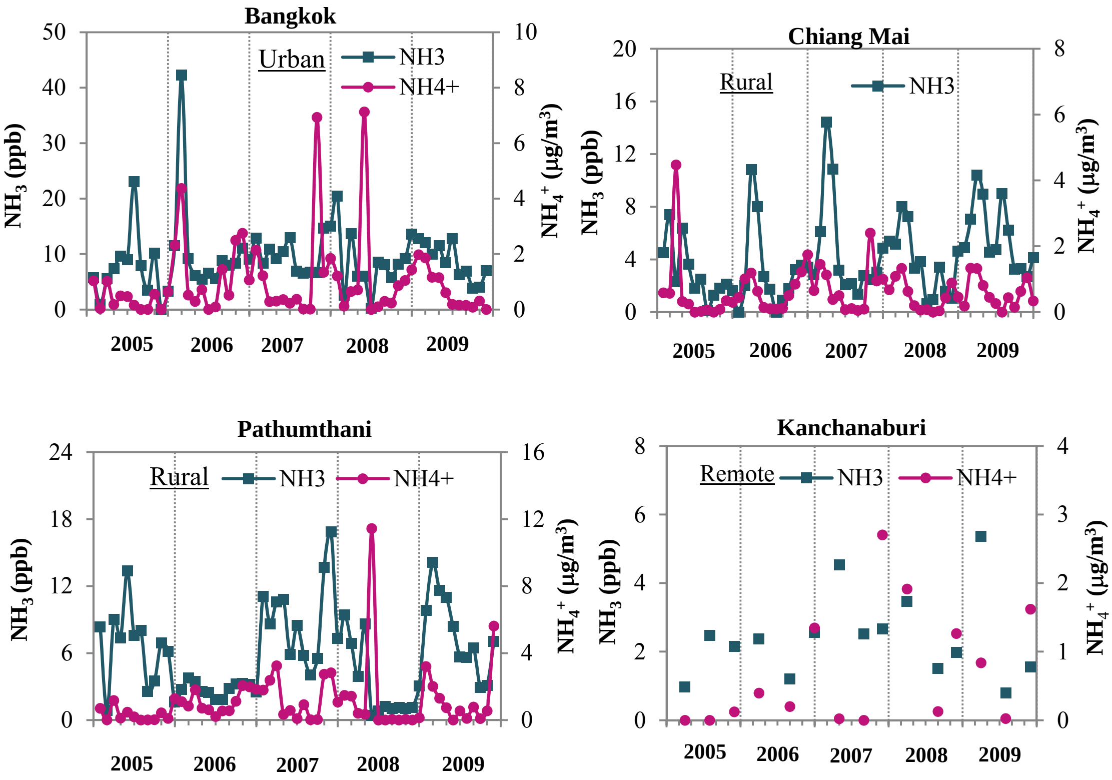


Figure 4.4.18. Seasonal characteristics of NH_3 and NH_4^+ concentrations at urban, rural and remote monitoring sites in Thailand, Viet Nam, Malaysia, Indonesia and Philippines.

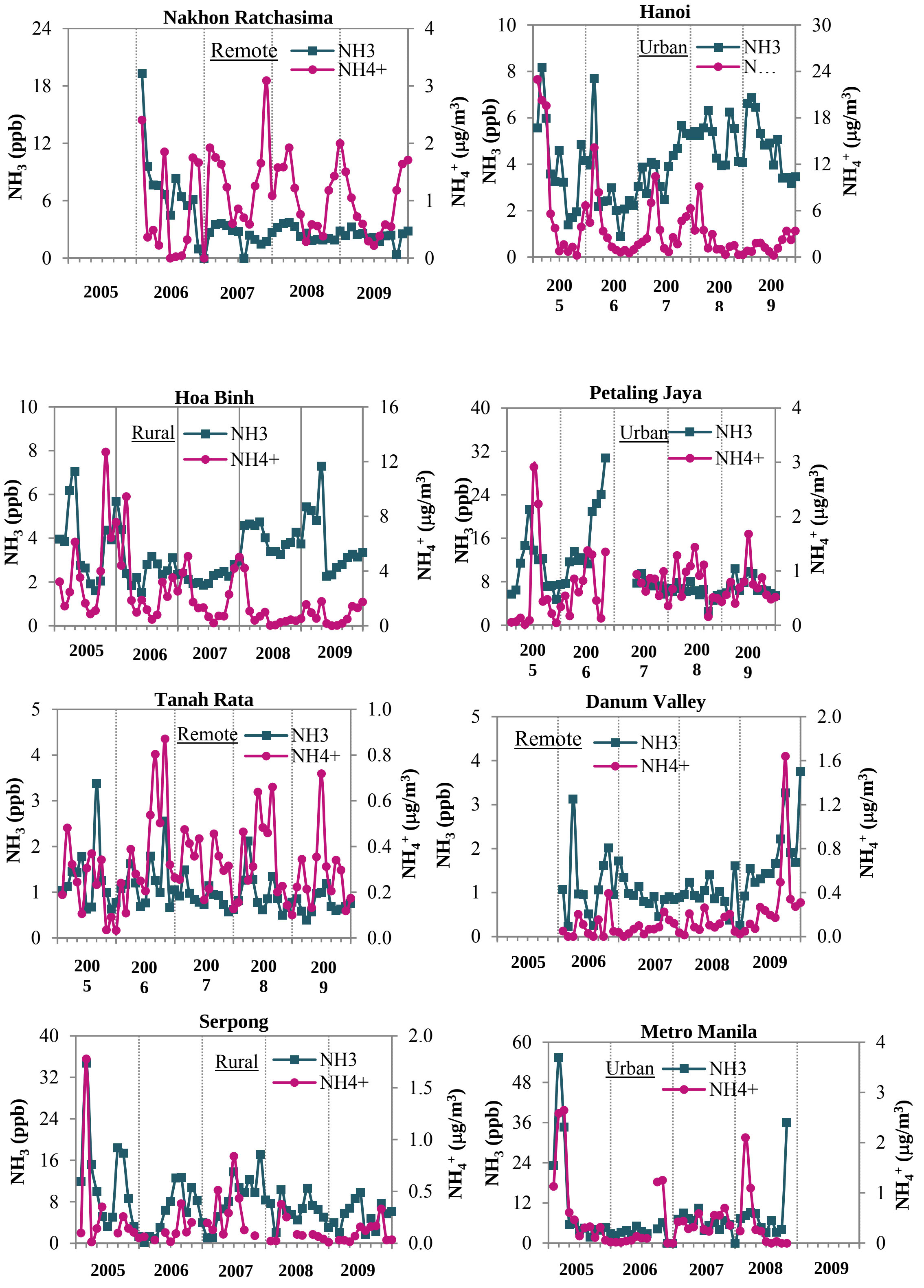


Figure 4.4.18. *(continued)*.

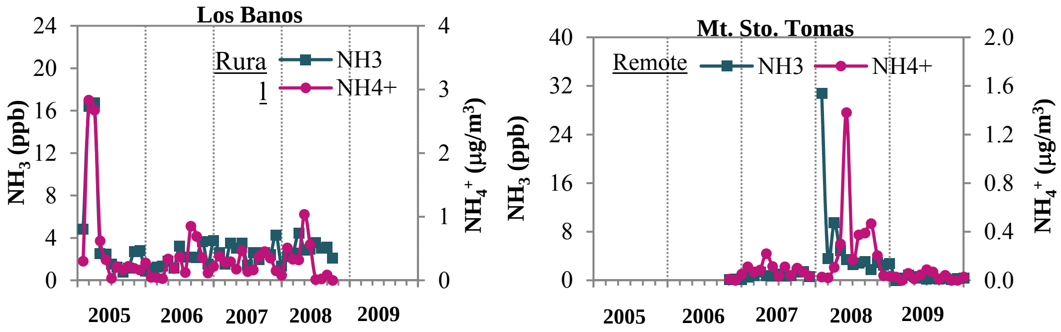


Figure 4.4.18. *(continued).*

NO$_2$, NOx. O$_3$, PM$_{10}$ concentrations

Figures 4.4.19 and 4.4.20 show annual variations of NOx, NO$_2$, O$_3$ and PM$_{10}$ concentration in Indonesia and Thailand from 2001-2009. Some sites in Thailand showed increasing trend, but the other data did not show significant trend. Variations in PM concentration at specific period should be noted because severe adverse atmospheric effects have been reported during haze event of dry season.

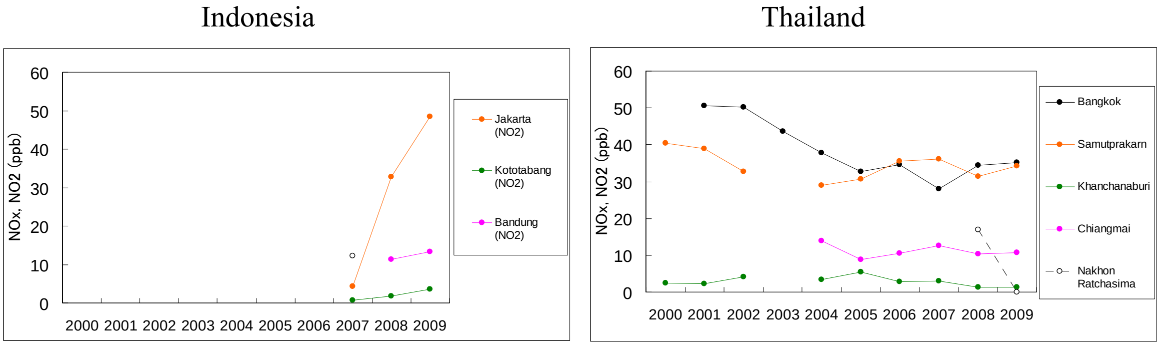


Figure 4.4.19. Annual variations of NOx or NO$_2$ concentration in Indonesia and Thailand from 2001-2009.

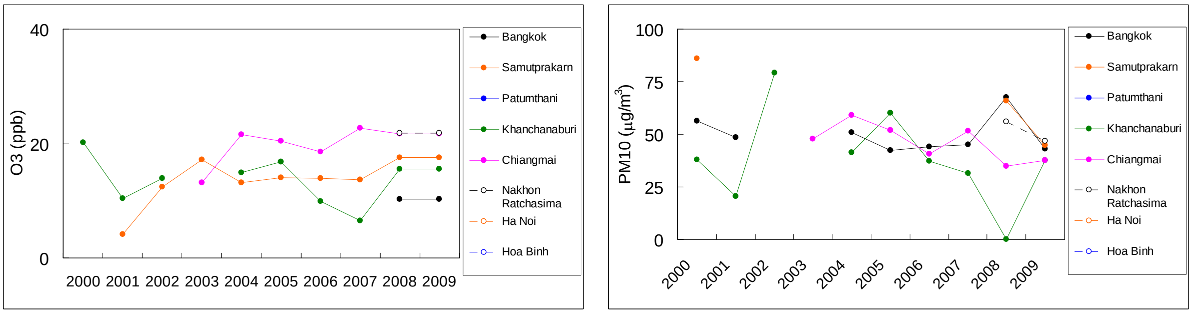


Figure 4.4.20. Annual variations of O$_3$ or PM$_{10}$ concentration in Thailand from 2001-2009.

4.5 Trends of amount of dry deposition and total deposition

By using the air concentration data of sulphur and nitrogen compounds and meteorological parameters measured in the EANET stations in Japan during 2003-2007, dry deposition amounts were calculated for each site. 5-year mean annual dry deposition amounts for sulphur and nitrogen compounds at Japanese EANET sites are shown in Figure 4.5.1, and they were in the range of 5-37 mmol.m^2 yr^{-1} and 7-50 mmol.m^2 yr^{-1}, respectively. SO_2 was the largest contribute or to dry deposition at each site. The contribution of seasalt SO_4^{2-} was relatively small, but it was found to be higher, especially, at coastal sites, such as Tappi, Sado-seki, Oki, and Hedo. The dry deposition amounts of HNO_3 and NH_4^+ dominated in the dry deposition of nitrogen compounds at most sites and accounted for approximately 20-50% and 10-40%, respectively.

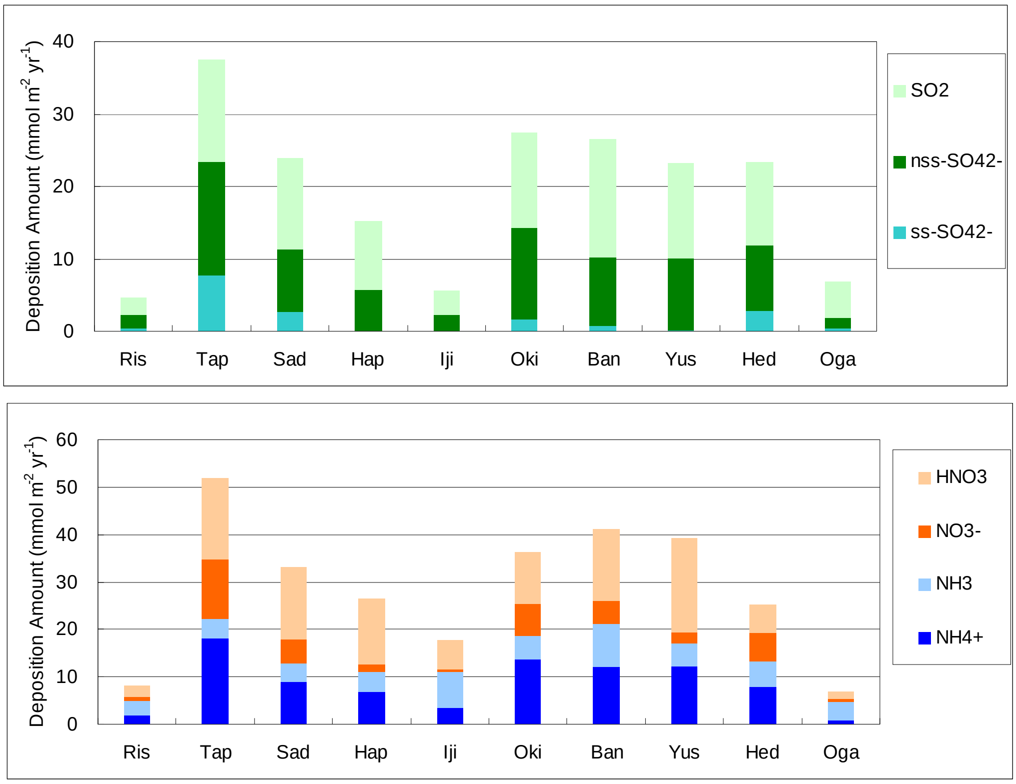


Figure 4.5.1. 5-year mean annual dry deposition amount of sulphur and nitrogen compounds at Japanese EANET sites during 2003 and 2007.

Note: Ris: Rishiri, Tap: Tappi, Sad: Sado-seki, Hap: Happo, Iji: Ijira, Oki: Oki, Ban: Banryu, Yus: Yusuhara, Hed: Hedo, Oga: Ogasawara

The regional characteristics of dry deposition are similar among sulphur and nitrogen compounds. They were higher in the Sea of Japan side (Tappi and Sado-seki) and in the Western Japan (Oki, Banryu, Yusuhara, and Hedo) and lower at Rishiri, Ijira, and Ogasawara. At Tappi and Sado-seki which categorized into remote site, the atmospheric concentrations were not significantly higher, whereas it is likely that higher V_d caused higher dry deposition.

Figure 4.5.2 shows time variations of annual dry and wet deposition for sulphur and nitrogen compounds from 2003 to 2007. 5-year average of total atmospheric deposition of sulphur and nitrogen compounds was in the range of 28 − 77 mmol.m^{-2}yr^{-1}and 22 − 130 mmol.m^{-2}yr^{-1}, respectively

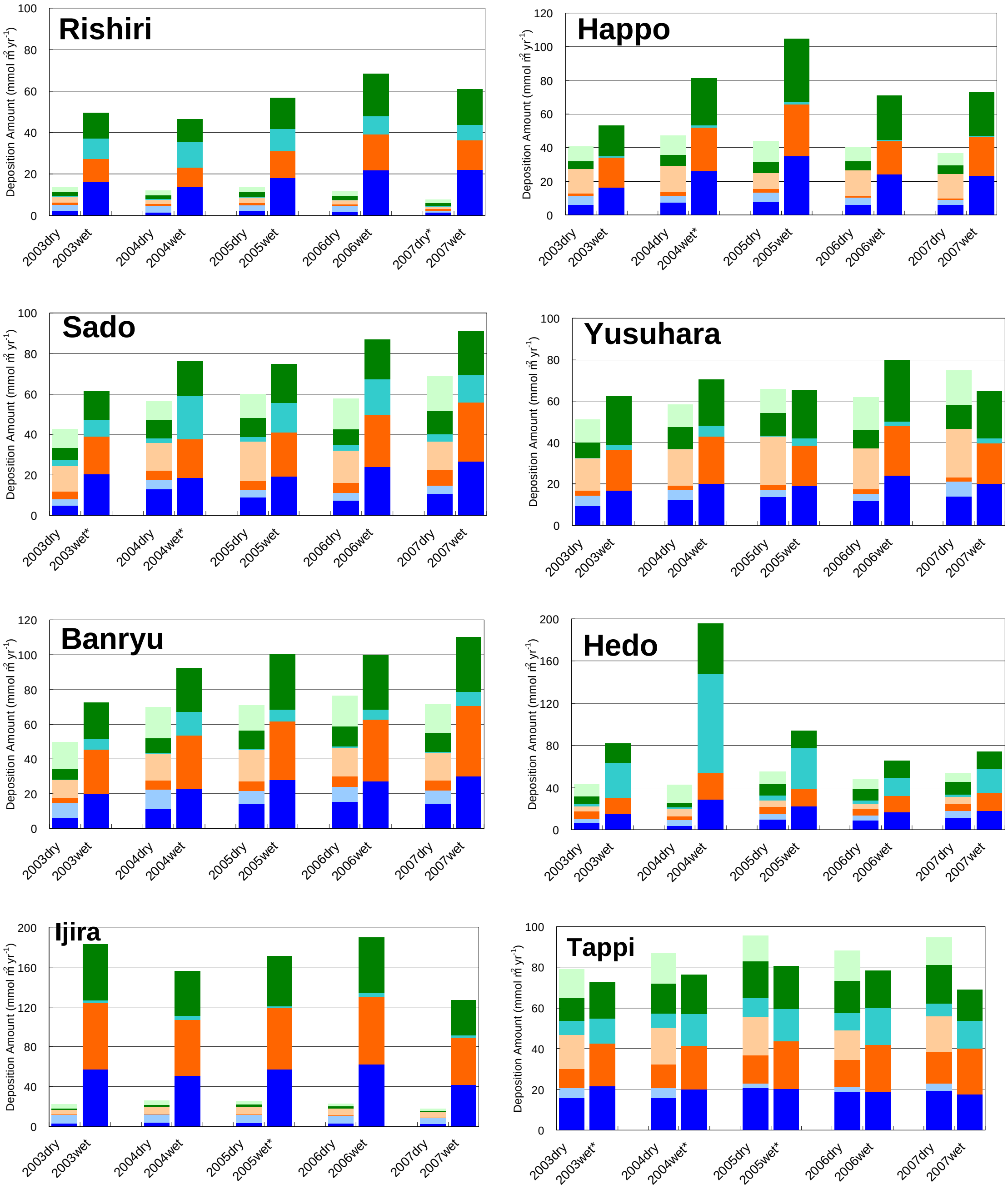


Figure 4.5.2.Time variations of annual dry and wet deposition for sulphur and nitrogen compounds from 2003 to 2007.

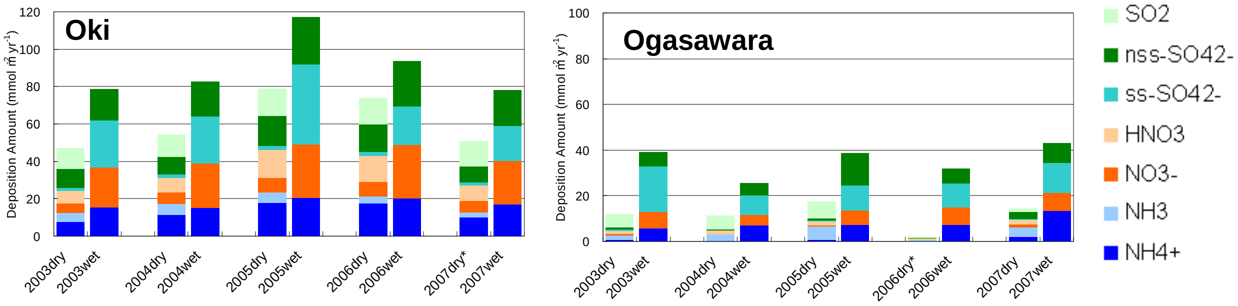


Figure 4.5.2. *(continued).*

The total atmospheric deposition of sulphur compounds was higher in the Sea of Japan side (Tappi and Sado-seki) and in the Western Japan (Oki, Banryu, Yusuhara, and Hedo) and lower at Rishiri and Ogasawara. At Banryu and Yusuhara, the SO_2 concentration was especially higher in autumn and winter, which resulted in higher mean annual concentrations. A winter monsoon dominantly comes from Eurasia to the Japan islands, and the SO_2 concentrations during 2003 and 2007 at 4 EANET sites in China, which are located in rural and urban sites and upwind of Japan, were 20 – 30 times higher than those at the EANET sites in Japan. Therefore, one of the reasons for the high total deposition amounts in Japan may be long-range transboundary air pollution.

Higher total atmospheric deposition amounts of nitrogen compounds were observed at Ijira and Banryu, which are classified as rural and urban sites, respectively. On the other hand, total nitrogen depositions were lower at Rishiri and Ogasawara, which are located far from major emission sources of nitrogen oxides. These observations suggest that the total atmospheric deposition of nitrogen compound is influenced by local emission sources rather than long-range transboundary air pollution. Since the atmospheric lifetime of nitrogen compounds is generally shorter than that of sulphur compounds, the regional characteristics of the total deposition amounts for nitrogen compounds would be dominated by the local emission sources.

The contribution of dry depositions to the total depositions for sulphur and nitrogen compounds also showed regional characteristics; namely, they were higher at Tappi and lower at Rishiri, Ijira, and Ogasawara. At Tappi, the contributions for sulphur and nitrogen compounds were 55 and 56%, respectively, because of the high dry deposition amount caused by the high V_d, as reported above. On the other hand, the contributions at Ijira were 10 and 13%, respectively. Because the nss-SO_4^{2-}, NO_3^-, and NH_4^+ concentrations in precipitation and precipitation amounts at Ijira were higher than those at other sites, the wet deposition amount at Ijira was dominant and, thus, the contribution of dry deposition amount was low. At Rishiri and Ogasawara, the contribution was approximately 20 – 30% because of the low dry deposition amounts caused by low atmospheric concentrations.

The annual variation, which is defined as the correlation between the 3-year moving averages of dry, wet, and total deposition amounts, and the measurement year remarkably increased at some sites, especially, on the Sea of Japan. At Rishiri, Sado-seki, and Banryu, the wet deposition amounts of nitrogen compounds increased significantly, and, thus, the total deposition amounts at Rishiri and Sado-seki also increased. In addition, the yearly fluctuation of the dry deposition amount of sulphur compounds at Sado-seki and Yusuhara significantly increased.

4.6 Overall analysis on atmospheric deposition

To characterize the atmospheric deposition in Japanese EANET sites, total atmospheric depositions of sulphur and nitrogen compounds in this study were compared with those estimated by the CASTNET (Clean Air Status and Trends Network) in the U.S.A. Figure 4.6.1 and 4.6.2 show the statistical values of dry, wet, and total deposition amounts for sulphur and nitrogen compounds in 3 networks, respectively. For CASTNET data, the dry deposition amount of NH_3 was not estimated in CASTNET and was not included in Figure 4.6.2. For EMEP data in this study, dry and wet deposition amounts at each grid covering 27 EMEP countries were estimated by the unified EMEP models

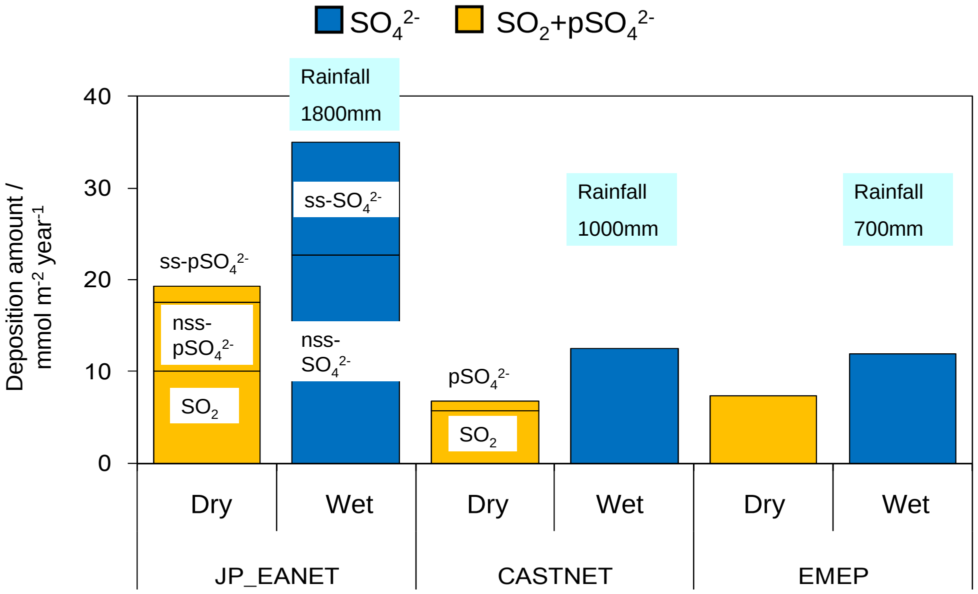


Figure 4.6.1. Comparison of dry, wet, and total deposition amounts of sulphur compounds among Japanese EANET sites, CASTNET in US and EMEP in Europe. *(Average of 2003-2007).*

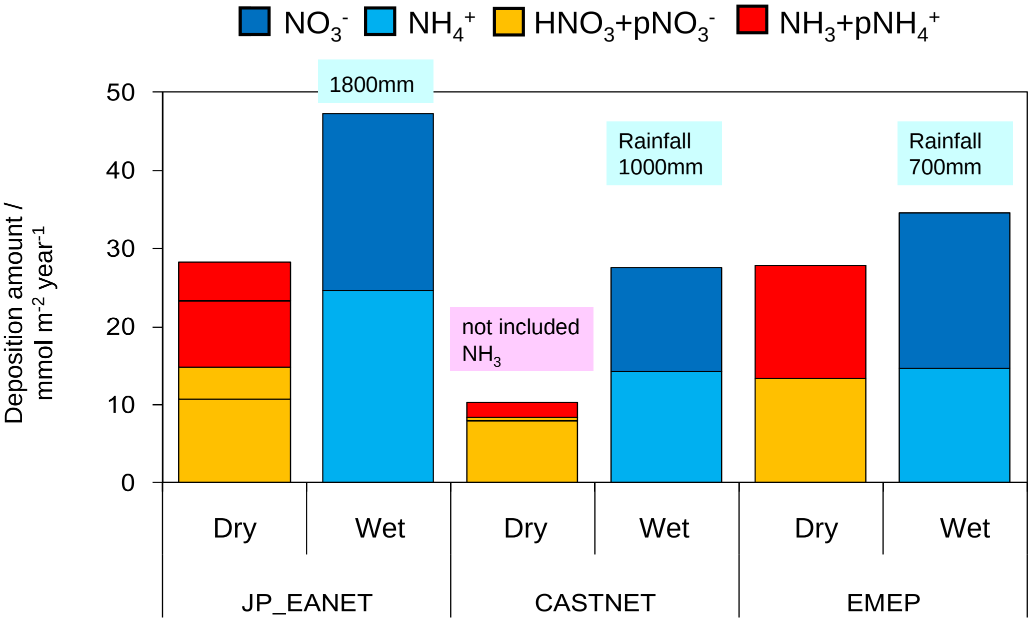


Figure 4.6.2. Comparison of dry, wet, and total deposition amounts of nitrogen compounds among Japanese EANET sites, CASTNET in US and EMEP in Europe. *(Average of 2003-2007).*

The 5 year average values for the total deposition amounts of sulphur and nitrogen compounds in Japan were higher than those in the other networks. Especially notable, the dry, wet, and total deposition amounts of sulphur compounds in Japan were approximately 3 times higher than those in the other networks. Since most of the EANET sites in Japan were located in a remote area, the deposition amounts were expected to be lower than those of EMEP, including data in urban areas. Although the total deposition amounts in the eastern region of CASTNET had higher values (average: 9.2 kgS ha^{-1}) because of the site locations close to the SO_2 emission sources, the deposition amounts in Japan were higher than those of CASTNET.

The reasons for the high deposition amounts are the high precipitation amounts and high nss-SO_4^{2-} concentrations of wet deposition in Japan. Other study found that the particulate SO_4^{2-} concentration by filter-pack was higher at sites toward the West of Japan, in closer to Asian continent. Also it was suggested that the outflow from Asian continental influenced the particulate SO_4^{2-} concentration throughout Japan from model result and the SO_2 concentration was influenced by the local emission. Therefore, the higher deposition amounts of sulphur compounds in Japan were possibly caused by the long-range transboundary air pollution and natural emission sources, such as volcanic eruptions in Japan.

For nitrogen compounds, the average value of the total deposition amounts in Japan was higher than that from other networks. Although the average and median values of dry deposition amounts were the same level in Japan and EMEP (approximately 4 kgN ha^{-1}), the values of wet deposition amounts in Japan were higher because of high precipitation, as reported above. Dry deposition amounts in CASTNET were lower in 3 networks because of the lack of dry deposition amounts of NH_3. The emission trends of sulphur and nitrogen compounds would be associated with the deposition amounts in these networks. In CASTNET, the SO_2 and NOx emissions from the electric power industry were reduced from 1990 to 2007, and, thus, the total deposition amounts of sulphur and nitrogen compounds declined. In EMEP, although the inter-annual variability in emissions has been small over the last decade, the total sulphur and NOx emissions decreased from 2006 to 2007 in the 27 EMEP countries. Therefore, the total deposition amounts of oxidized sulphur and nitrogen compounds and reduced nitrogen compounds decreased from 2006 to 2007. On the other hand, the total deposition amounts at some sites in Japan increased remarkably, as reported above. The increasing SO_2 and NOx emissions in East Asia would cause high atmospheric deposition, and, thus, the total deposition amounts in Japan will be higher than those in other networks.

4.7 Conclusions

Most of EANET monitoring sites in Northeast Asia as well as in Southeast Asia show stabilizing trend of gaseous (SO_2, HNO_3 and NH_3) and aerosol (SO_4^{2-}, NO_3^- and NH_4^+) concentrations during 2005-2009. Some urban sites in Southeast Asia showed significant improvement in SO_2 reduction.

Five years analyses (2005-2009) of gaseous and aerosol concentrations indicated specific spatial characteristics of chemical distribution among EANET observing sites. All high concentrations of NH_3 were observed in urban, rural and remote areas while SO_2 concentrations measured at the same sites were lower and significantly very low for HNO_3. This phenomenon was unexpectedly to occur in the city where NH_3 would generally be found in agricultural areas.

The aerosol concentrations of sulphate (SO_4^{2-}) were comparably higher than NO_3^- and NH_4^+ at all urban, rural and remote sites in Northeast Asia. Sulphate is a secondary chemical species and hence, the observed large values of SO_4^- had been traded off with low measured values of SO_2 concentration at the same sites. On the other hand, high sulphate can be resulted by a secondary reaction of gaseous sulphur components during a long range transport. For another secondary acid component, HNO_3, the concentrations were detectably very low at all the Northeast Asia monitoring sites.

All measured concentrations of aerosols (SO_4^{2-}, NO_3^- and NH_4^+) in Southeast Asia sites were reportedly very low while in some urban sites, the measured values of SO_2 and NH_3 were significantly high. This may indicate a low effect of a long range transport of this secondary species.

Temporal analyses (2005-2009) of chemical distribution in Northeast Asia show distinctive seasonal characteristics. A cyclic pattern of SO_2 and SO_4^{2-} concentrations had been seen clearly at most of the urban, rural and remote sites. The lowest concentration was found in the mid-summer while the highest value lied in the winter. This indicates high energy utilization during the low temperature climate. Concentrations of HNO_3 and NO_3^- were fluctuating with seasonal changes.

Temporal analyses (2005-2009) of chemical distribution in Southeast Asia showed similar trend in which atmospheric chemical concentration verily with season. NH_3 concentration was found substantially high at all monitoring sites (urban, rural and remote) when compared with SO_2. Their monthly average concentrations are generally 3 times higher than the measured SO_2 values. A large release of ammonia observed at all the EANET sites indicates large effects of Nitrogen constituent emissions from agricultural activities. In case of aerosol components, SO_4^{2-} is proportionately much greater than other two acid components, i.e. NO_3^- and NH_4^+.

All urban sites in EANET Southeast Asia countries show significantly improvement of SO_2 and SO_4^{2-} emission reduction during 2005-2009. Concentrations of SO_2 and SO_4^{2-} at the rural and remote sites are likely to be seasonal independency.

The 5 year average values for the total deposition amounts of sulphur and nitrogen compounds in Japan were reportedly high which due to high precipitation and nss-SO_4^{2-} concentrations. It was possibly caused by the long-range transboundary air pollution and natural emission sources, such as volcanic eruptions in Japan.

Currently, all EANET sites, except Japan have not been facilitated with required meteorological instruments for dry deposition evaluations. It is recommended that EANET countries should have the meteorological instruments installed within assigned time frame.

4.8 References

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Chapter 5. Impacts on Ecosystems in East Asia

5.1.Introduction

The determination of impacts of acid deposition on ecosystems remains to be a vital mandate of the EANET. Participating countries in the network have established ecological monitoring plots in strategic locations within the country following the EANET prescriptions and guidelines. The ecological monitoring activities of the EANET participating countries are ultimately designed to assess the impacts of acid deposition on the soil, forest vegetation, inland water such as lake and river, and other terrestrial ecosystems. The initial data gathering was intended to establish baseline information on the monitoring sites. Subsequent observations at regular intervals of time were made to detect changes in the ecosystems being monitored. Such should allow for the early detection of impacts on the terrestrial and inland water ecosystems. In the case of forest vegetation, the primary interest is to recognize early signs of forest decline attributable to acid deposition. The same is true for soil and inland water acidification. A standard methodology was developed for the monitoring of acid deposition impacts on the said ecosystems for adoption in all EANET member countries to allow for comparison of results at a later period of time.

The countries that participated in the EANET activities on ecological monitoring were China, Indonesia, Japan, Korea, Lao PDR, Malaysia, Mongolia, Philippines, Thailand, Russia and Vietnam (Figure 5.1.1). There were seven countries which did monitoring of the forest soil during the period under review namely China, Indonesia, Japan, Korea, Philippines, Russia and Thailand. There were also seven countries which established and monitored for impacts on forest vegetation in the region. These were China, Japan, Korea, Mongolia, Philippines, Russia and Thailand. Finally, all countries with the exception of Korea did ecological monitoring for inland aquatic ecosystems.

The different parameters measured from soil, inland aquatic bodies and forests are presented in Table 5.1.1. Some parameters are common among the three sampling sites, although there are also distinct measurements that are unique to each location. Soil and the inland aquatic ecosystem were sampled for elements and ions, and their physical characteristics. On the other hand, forest measurements focused on observing for declines in tree, forest growth, and understory vegetation.

This chapter describes the features of the different ecological monitoring sites in the participating countries of the EANET. It also discusses the trends in the observations made on the soil, forest vegetation, and inland aquatic ecosystems in the said stations from 2000 to 2009. A section in this chapter will also report on the progress and results of the catchment scale analysis of the effects of acid deposition undertaken by the Network Center and scientists of the EANET community in several locations within the East Asian Region.

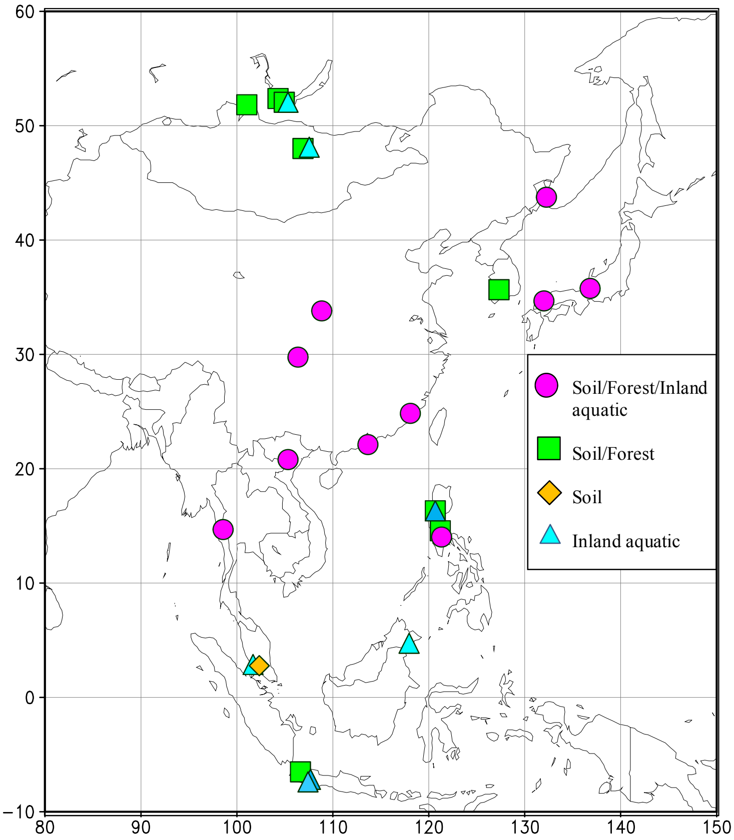

Figure 5.1.1. Location of the monitoring sites of different participating countries.

Table 5.1.1. Basic survey parameters for soil, forest vegetation and inland waters.

Item	Parameters
Soil	- pH (H_2O), pH (KCl), Exchangeable base cations (Na, K, Ca, and Mg), Exchangeable acidity, Effective cation exchange capacity (ECEC), Carbonate content (if pH > 7) - Exchangeable Al and H, Total C content, Total N content (optional) - Available P, Sulfate (voluntary) - Physical properties (Fine earth bulk density, and Penetration resistance)(optional)

Forest	- General description of the forest
	  - description of trees
	  - understory vegetation
	- Observation of tree decline
	- Photographic record of tree decline
	- Estimation of decline causes (optional)

Inland Aquatic	- Temperature, pH, electrical conductivity, alkalinity - SO_4^{2-}, NO^{3-}, NO^{2-}, PO_4^{3-}, Cl^- - NH_4^+, K^+,Mg^{2+}, Ca^{2+}, Na^+, - Total Al^{3+}, inorganic Al^{3+} - DOC, COD, transparency, water color

5.2.Soil features and trends

5.2.1. Soil features in EANET sites

Researchers and scientists are familiar with the surface or top soil–the dark layer which is also known as the O horizon. Going down the lower layer yields surprising colors and structures of the soil. The subsoil gives hints to the history of the soil and past environmental conditions. It also shows the conditions that plant roots must explore and how this will affect the land use. Soil resources are always changing and with this, pore spaces, water and gas contents, and the electron exchange environment are dynamically changing every moment. Within this continuing changing environment, the roots of the trees must develop growth and survival solutions or mechanisms especially in areas where acid deposition is prevalent.

Seven countries implemented soil monitoring for acid deposition effects namely China, Indonesia, Japan, Korea, Philippines, Russia and Thailand. Table 5.2.1 shows the location of the soil monitoring sites, soil types and the initial soil analyses at the 0-10 cm soil layer.

China has four monitoring sites, namely Jinyunshan, Jiwozi, Xiaoping and Zhuxiandong which listed Acidic-Udic Argosols, Brown soil, Red soil and Ochinic Udic Ferrosols, respectively, as their soil types. Based on their soil analysis, only the brown soil (Jiwozi) is considered as moderately acidic while the rest are extremely acidic with soil pH below 4.5. This acidic property could also be described by the soil type which is Red soil, Acidic–Udic Argosols and Ochinic Udic Ferrosols which also suggests high iron content which is one major characteristic of an acidic soil. Udic implies humid climate or moist soil condition wherein the soil is not dry in any part for as long as 90 cumulative days. The base cation Ca is very low to medium, Mg is very low to low and K is low to high. Another characteristic that makes these soils acidic is the occurrence of high amount of exchangeable Al in Jinyunshan, Xiaoping and Zhuxiandong.

Indonesia’s monitoring site is in Bogor near Serpong and has typic Dystrudepts as soil type which connotes soil with low base saturation (Table 5.2.1) and with udic moisture regime suggesting moist or humid climate and belonging to the order Inceptisols or embryonic soil with few diagnostic feature, meaning younger soil. The pH is extremely acidic with very low levels of Ca, Mg and K and high amount of exchangeable aluminum.

Japan has four monitoring sites located in Banryu, Iwami, Ijira and Yamato with the corresponding soil types of Cambisols, Acrisols, Dystric Cambisol and Andosols, respectively. The pH of these monitoring sites ranges from extremely acidic to very strongly acidic (pH 4.19-4,74) while the Ca, Mg and K content range from low to very low which is characteristic feature of acidic soils aside from the very high level of exchangeable aluminum, as discussed earlier. Cambisols have a mineral horizon that has a texture of loamy very fine sand, contains weatherable minerals, and is characterized by the alternation or removal of mineral material. It lacks cementation or induration and has too few evidences of illumination to meet the requirements of argillic or spodic horizon. Acrisols is characterized as those soils undergoing extreme weathering while Andosols are from volcanic ejecta, dominated by allophane or Al-humic complexes.

Mt. Naejang is the monitoring site for Korea and is characterized by very strongly acidic soil, very low Ca and Mg and low K contents. The exchangeable Al is very high with low effective CEC.

The Philippines has four monitoring sites located in Quezon, Laguna, Quezon City and Benguet. Only the Dystric Nitosol from Quezon and Eutric Cambisol from Laguna were the soil types identified. The soil pH from Quezon is extremely acidic (pH 4.10), Ca is very low, Mg and K are considered low while the exchangeable Al is very high. These characteristic feature fits well with the soil type which is Dystric Nitosol which connotes low base saturation and strongly weathered soils but productive. The other soil type, Eutric Cambisol means high base saturation with cambic horizon. The high base saturation is shown in the soil properties of the three other monitoring sites with medium to high Ca level, high to very high Mg, medium to very high K and moderately acidic.

Irkutsk, the monitoring site in Russia, has two soil types, Eutric Regosols and Calcaric Luvisols. The soil properties suggest slightly acidic to neutral pH, high to very high Ca, high Mg and low to medium K. The soil type eutric connotes high base saturation as shown in Table 5.2.1. Calcaric refers to presence of lime or calcium carbonate as shown by high amount of Ca in the soil while Luvisols means a soil layer or horizon in which material carried from an overlying layer has been precipitated from solution or deposited from suspension or a layer of accumulation.

Thailand has two monitoring sites, namely Vachiralongkorn Dam with soil type Ferric Acrisol and Vachiralongkorn Puye with Luvisols as the soil type. These two sites represent soils with different sensitivities to acid deposition. Ferric Acrisol denotes high iron content and high weathering although values of the soil analyses are high (slightly acidic soil pH, high Ca and Mg) as compared to Luvisols with extremely acidic soil pH, medium Ca and Mg contents, very low amount of K and high exchangeable aluminum. Classifying or identifying the soil type is very important since these could be used as a guide in determining the chemical or physical properties of a given soil or area and in assessing its land use.

Table 5.2.1. Location, soil type and initial soil chemical properties of EANET soil monitoring sites.

Country	Location	Soil Type	Initial soil chemical analysis at 0-10 cm soil depth							
			soil pH		Ca	Mg	K	Exch Al	ECEC	BS (%)
			H2O	KCl	cmole(+)/kg soil					
China	Jinyunshan (2000)	Acidic-Udic Argosols	3.79	3.29	0.57	0.15	0.11	7.10	10.52	8.34
	Jiwozi (2001)	Brown soil	5.95	5.19	8.58	0.68	0.86	0.01	10.30	99.20
	Xiaoping (2003)	Red soil	4.41	3.82	0.05	0.08	0.19	4.79	6.01	6.60
	Zhuxiandong (2003)	Ochinic Udic Ferrosols	4.03	3.58	4.13	1.37	1.74	3.65	12.01	66.00
Indonesia	Bogor (Serpong) (2001)	Typic Dystrudepts	4.30	3.88	0.93	0.31	0.07	3.34	4.81	26.79
Japan	Banryu-2	Cambisols	4.74	3.75	0.42	0.33	0.22	3.16	4.58	22.10
	Iwami (2001)	Acrisols	4.26	3.38	0.42	0.48	0.29	7.30	9.58	13.6
	Ijira (2001)	Dystric Cambisol	4.19	3.32	0.32	0.77	0.11	6.37	8.82	16.41
	Yamato (2006)	Andosols	4.39	3.93	0.18	0.20	0.24	0.51	1.25	51.84
Korea	Mt. Naejang (2001)		5.04	3.91	0.79	0.29	0.14	3.21	5.27	25.69
Philippines	Quezon (2000)	Dystric Nitosol	4.10	4.00	0.60	0.70	0.40	3.90	7.30	41.20
	Laguna (2002)	Eutric Cambisol	5.42	4.77	17.70	15.94	2.80	0.23	39.71	98.87
	Quezon City (2007)		5.66	4.39	6.90	3.65	0.24	0.09	14.15	41.20
	Mt. Sto. Tomas, Benguet (2008)		5.36	5.05	7.98	0.94	0.24	1.62	11.50	83.17
Russia	Irkutsk (2001)	Eutric Regosols	6.74	5.43	19.57	4.16	0.43	0.10	24.46	99.32
	Irkutsk (2001)	Calcaric Luvisols	6.39	5.54	24.37	6.34	0.22	0.10	31.13	99.91
Thailand	Vachiralongkorn Dam	Ferric Acrisols	6.12	5.55	13.13	55.61	0.06	0.02	69.52	99.25
	Vachiralongkorn Puye	Luvisols	4.27	4.13	5.04	1.59	0.09	1.30	8.35	78.60

5.2.2. Trend of soil chemical properties

5.2.2.1. Regional variation in soil chemical properties

Soil monitoring has been conducted several times for 10 years (2000-2009) in 20 sites including around 40 plots and 200 subplots (Table 5.2.2). Mean value of soil chemical properties for 10 years had a large variation between the sites (i.e. regional variation). For example, the values in 0-10 cm varied from 4.0 to 6.8 in pH (H_2O), 3.4 to 5.8 in pH (KCl), and 0.05 to 26.0 $\mathrm{cmol_c\ kg^{-1}}$ in Ex-Ca^{2+}. Especially, a coefficient of variance (C.V.) in this regional variation was 18% in pH (H_2O) for 0-10 cm. This was much larger than average of a variation between 2 or 3 plots in the same site (4%), a variation between 5 subplots in the same plot (3%), a variation between the repeat analyses in the same samples (1%) and a variation between the year in the same subplots (i.e. temporal variation; 4%). Similar tendency was seen among other soil properties.

A map shows the regional variation in the ratio between exchangeable base cations (Ca^{2+}, Mg^{2+}, Na^+, K^+) and acid cations (H^+, Al^{3+}), which were adsorbed in the negative charge on a surface of clay minerals or organic components of soil (Figure 5.2.1). The ratio indicates the buffering capacity of the soil against atmospheric acid or base deposition. Base cations were dominant in the soil of the monitoring site in Russia, Jiwozi, Vachiralongkorn (Kanchanaburi), Boneco (Mt. Sto. Tomas), La Mesa (Metro Manila) and so on, whereas acid cations largely exceeded the base cations in large part of the monitoring site. In general, these regional variations are mainly derived from a difference of soil types decided by geological, climate, topographic and biological factors for long time scale.

In all the temporal-spatial variation with the exception of Russia, pH (H_2O) in 0-10 and 10-20 cm was well correlated with Ex-Ca^{2+} (Figure 5.2.2a). The relationship indicates that base cation concentrations are crucial for soil pH in the area, except in Russia. In general, high Ex-Ca^{2+} concentration and pH were observed on basic rock (e.g. Vachiralongkorn). Buffering capacity against acid deposition should be much higher in the area. Meanwhile, pH (H_2O) controlled by the base cations strongly regulates the solubility of the Aluminum ions (Al^{3+}) which is harmful to plant roots (Ulrich and Summer, 1991). In the soils of the participating countries, over 70% samples showed less than pH 5.4 in which Aluminum exists as Al^{3+} (Figure 5.2.2b). The pH decline may lead to increase in Al^{3+} in many parts of the monitoring sites, although an increasing rate may be depend on the site characteristics.

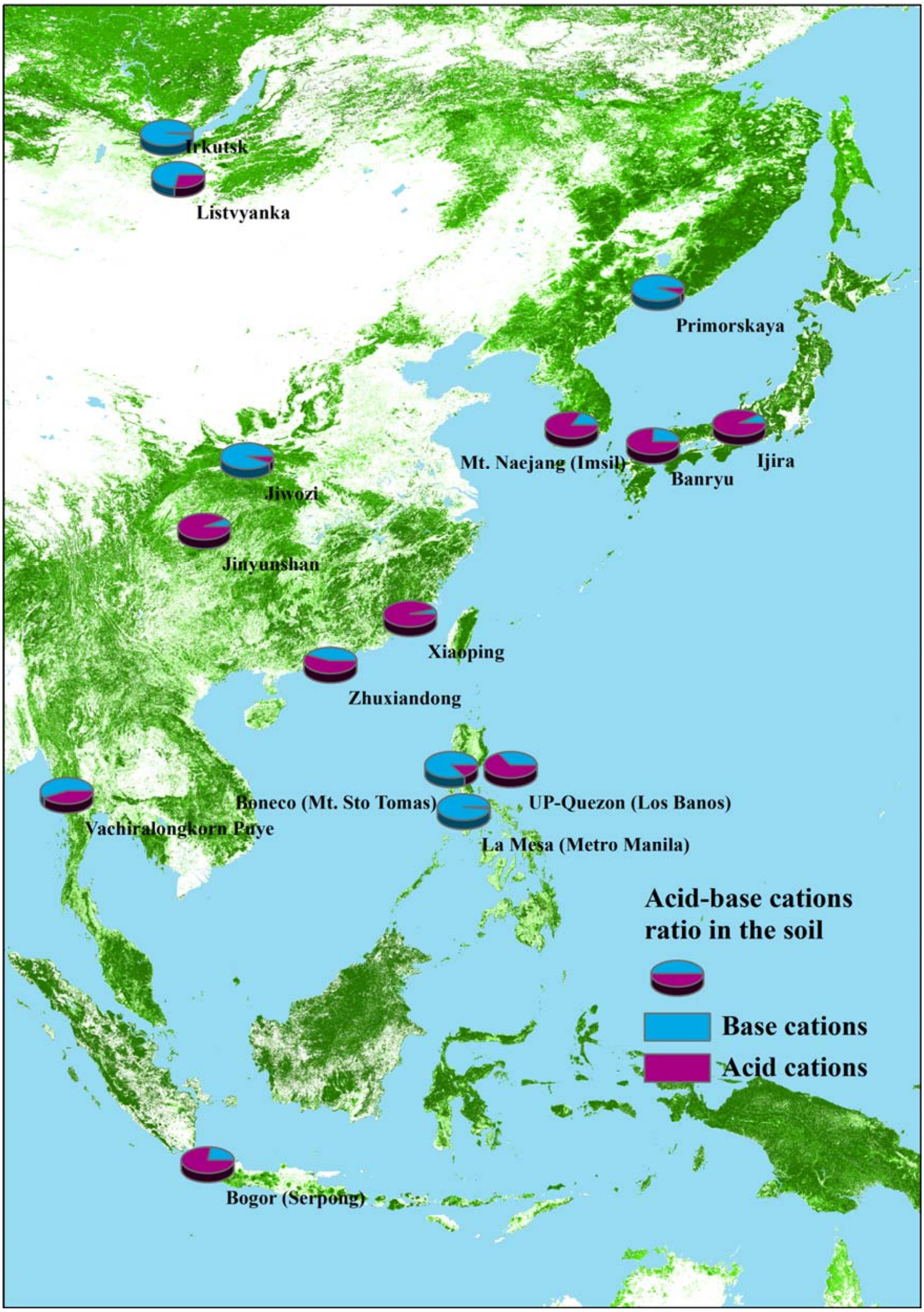

Figure 5.2.1. Ratio between base cations ($Ca^{2+} + Mg^{2+} + Na^{+} + K^{+}$) and acid cations ($H^{+}+ Al^{3+}$) on a negative charge of 0-10 cm soils. Only representative site in each area were displayed. Dense of green color shows tree cover. The value is average in 2000-2009.

Table 5.2.2. Mean value of the soil chemical properties for 2000-2009 in each monitoring site.

Country	Site name (Site for wet and dry)	survey year	Plot	Depth	pH(H_2O)	pH(KCl)	Ca^{2+}	Mg^{2+}	K	Al	ECEC	BS
				(cm)			(cmolc kg-1)					(%)
China	Jinyunshan	2000, 2003, 2006, 2009	1	0-10[*1]	4.0	3.4	0.39	0.09	0.09	6.91	8.1	7.3
				10-20[*2]	4.2	3.6	0.25	0.06	0.07	5.79	6.6	6.2
			2	0-10[*1]	4.1	3.5	0.33	0.06	0.08	5.38	6.6	7.5
				10-20[*2]	4.5	3.7	0.19	0.06	0.08	3.99	4.9	11.8
	Jiwoji	2003, 2006, 2009	1	0-10	6.1	5.2	6.91	0.81	0.61	0.11	9.1	93.5
				10-20	6.3	5.3	6.94	0.82	0.66	0.06	9.0	94.3
			2	0-10	6.3	5.1	6.08	0.81	0.57	0.11	8.2	93.7
				10-20	6.3	5.1	5.77	0.73	0.41	0.08	7.6	93.4
	Xiaoping	2003, 2006, 2009	1	0-10	4.4	3.8	0.05	0.07	0.19	6.14	7.6	4.6
				10-20	4.5	3.9	0.04	0.06	0.17	4.79	6.1	5.0
			2	0-10	4.5	3.8	0.07	0.09	0.20	4.52	5.5	8.3
				10-20	4.6	3.9	0.02	0.05	0.17	4.21	4.9	5.8
	Zhuxiandong	2003, 2006, 2009	1	0-10	4.3	3.7	1.36	0.44	0.53	0.82	6.0	31.5
				10-20	4.3	3.8	1.07	0.35	0.47	0.70	5.2	29.0
			2	0-10	4.3	3.7	1.38	0.47	0.60	0.69	5.3	35.3
				10-20	4.4	3.8	1.37	0.52	0.63	0.63	6.4	36.1
			3	0-10	4.2	3.6	1.53	0.52	0.67	1.11	7.0	30.5
				10-20	4.2	3.6	1.03	0.33	0.55	1.13	6.2	26.2
Indonesia	Bogor (Serpong)	2001, 2007, 2009	1	0-10	4.3	3.8	0.93	0.29	0.08	4.09	5.9	22.6
				10-20	4.3	3.8	0.91	0.32	0.06	3.64	5.3	25.3
Japan	Banryu	2001, 2006	1	0-10	4.8	3.8	0.49	0.38	0.21	3.15	4.7	23.7
				10-20	4.9	3.9	0.22	0.27	0.17	3.02	4.1	17.2
			2	0-10	4.8	3.8	0.49	0.40	0.25	3.10	4.8	24.5
				10-20	4.9	3.9	0.25	0.27	0.20	2.88	4.1	18.6
	Iwami (Banryu)	2001, 2006	1	0-10	4.4	3.6	0.47	0.47	0.31	5.89	8.1	16.1
				10-20	4.5	3.7	0.18	0.30	0.23	5.58	7.1	10.6
			2	0-10	4.3	3.4	0.43	0.43	0.26	8.20	10.6	11.7
				10-20	4.5	3.6	0.17	0.26	0.19	7.17	8.7	7.9
	Ijira	2000, 2006	1	0-10[*3]	4.2	3.4	0.27	0.35	0.10	8.00	9.1	9.1
				10-20[*4]	4.2	3.5	0.13	0.26	0.08	6.83	7.6	7.2
			2	0-10[*3]	4.3	3.5	0.31	0.26	0.21	7.13	9.0	9.2
				10-20[*4]	4.5	3.7	0.06	0.06	0.16	4.93	5.9	5.0
	Yamato (Ijira)	2006	1	0-10	4.3	3.9	0.21	0.21	0.26	0.55	1.3	52.2
				10-20	4.3	3.9	0.08	0.11	0.18	0.44	0.9	44.2
			2	0-10	4.5	4.0	0.14	0.19	0.22	0.46	1.1	51.4
				10-20	4.5	3.9	0.07	0.13	0.17	0.35	0.8	49.1
Korea	Mt. Naejang (Imsil)	2001, 2004	1	0-10	4.9	3.9	0.54	0.18	0.14	3.45	5.4	17.3
				10-20	4.9	4.0	0.26	0.12	0.13	3.18	4.4	12.9
			2	0-10	5.0	4.0	0.56	0.21	0.10	3.25	5.5	18.0
				10-20	4.9	4.1	0.19	0.15	0.08	2.96	4.2	12.2
Phillipine	La Mesa Dam Watershed (Metro Manila)	2007	1	0-10	5.8	4.4	10.75	6.29	0.29	0.07	21.2	98.6
				10-20	5.6	4.4	10.68	5.36	0.29	0.03	19.9	98.4
			2	0-10	5.4	4.3	10.50	6.16	0.27	0.14	19.8	97.9
				10-20	5.5	4.2	10.26	6.42	0.23	0.05	19.4	98.4
	Mt. Makiling (Los Banos)	2001, 2005, 2008	1	0-10	5.6	4.8	13.47	13.64	3.97	0.15	33.5	98.4
				10-20	5.2	4.4	10.54	12.74	3.60	1.28	30.3	93.8
			2	0-10	6.2	5.3	20.67	12.62	4.59	0.00	40.8	99.4
				10-20	6.0	5.0	17.22	12.07	4.11	0.02	36.2	99.1
	Up Quezon (Los Banos)	2001, 2005, 2008	1	0-10	4.6	4.1	3.54	2.88	1.03	3.33	11.8	43.7
				10-20	4.5	4.0	2.84	2.58	0.87	3.54	10.7	36.7
			2	0-10	4.6	4.1	4.87	2.94	1.06	4.75	14.8	38.3
				10-20	4.5	4.0	3.66	2.57	0.87	4.54	12.7	33.5
	Boneco LTER (Mt. Sto Tomas)	2008	1	0-10	5.3	5.0	8.47	1.03	0.25	1.35	11.8	86.2
				10-20	5.1	4.9	6.88	0.78	0.20	1.49	10.1	81.4
			2	0-10	5.4	5.1	7.49	0.85	0.23	1.89	11.2	80.2
				10-20	5.2	4.9	5.60	0.72	0.19	2.00	9.2	74.0

Table 5.2.2. *(continued)*

Russia	Irkutsk	2001	1	0-10[*5]	6.5	4.4	14.64	3.20	0.24	0.23	18.5	98.3
				10-20[*6]	6.5	4.2	12.46	2.89	0.27	0.19	15.9	98.5
			2	0-10[*5]	6.4	5.5	24.37	6.34	0.22	0.10	-	-
				10-20[*6]	6.2	5.0	18.95	5.26	0.20	0.29	-	-
	Bolshie koty (Listvyanka)	2000	1	0-10[*7]	6.7	5.9	30.29	3.91	0.46	0.04	34.8	99.7
				10-20[*8]	6.8	5.8	21.58	3.62	0.26	0.06	25.7	99.5
			2	0-10[*7]	4.8	3.9	23.06	6.75	1.59	0.55	32.6	96.5
				10-20[*8]	4.8	3.7	4.55	1.16	0.13	3.15	9.2	63.4
	Pereem-naya river (Listvyanka)	2005	1	0-10	4.2	2.8	10.30	1.84	0.99	3.50	18.5	71.2
				10-20	4.6	4.2	0.47	0.24	0.12	15.23	18.5	4.8
			2	0-10	4.5	3.4	8.40	2.49	0.81	3.17	16.6	70.0
				10-20	5.0	4.0	6.70	1.62	0.29	3.79	13.8	61.5
	Primorskaya	2006	1	0-10	4.7	2.9	20.00	4.90	0.88	-	27.6	93.9
				10-20	4.9	3.1	8.58	3.77	0.38	-	16.4	78.5
			2	0-10	5.6	3.9	30.49	5.92	0.87	-	38.5	97.1
				10-20	5.1	4.2	7.23	2.44	0.49	-	12.5	82.9
Thailand	Vachiralongkorn Dam (Kanchanaburi)	2006, 2008	1	0-10	6.8	5.8	23.26	4.57	0.04	0.19	28.5	98.8
				10-20	6.8	5.6	17.74	3.52	0.04	0.16	21.8	98.8
			2	0-10	6.7	5.7	26.00	4.92	0.03	0.13	31.4	99.3
				10-20	6.7	5.6	22.09	3.90	0.03	0.13	26.4	99.2
	Vachiralongkorn Puye (Kanchanaburi)	2006, 2008	1	0-10	5.5	4.4	3.22	1.10	0.02	0.47	5.3	82.5
				10-20	5.3	4.0	1.76	0.64	0.02	1.26	4.3	57.5

[*1]: 1-7 cm, 3-12 cm, and 5-18 cm (A layer) in 2000 and 2003; [*2]: 7-21 cm, 12-24 cm and 12-41 (B layer) in 2000 and 2003; [*3]: 0-5, 0-6 and 0-7 cm (A_1 layer) in 2000; [*4]: 10-15, 11-16, 12-17 (A_2 layer) in 2000; [*5]: 0-10, 0-15, 1-10 and 1-9 cm; [*6]: 10-30, 10-35, 15-33 and 9-33 cm; [*7]: 0-10, 0-7, 0-8, 1-7, and 1-8 cm; [*8]: 10-18, 11-19 and 13-20 cm, BS: Base saturation

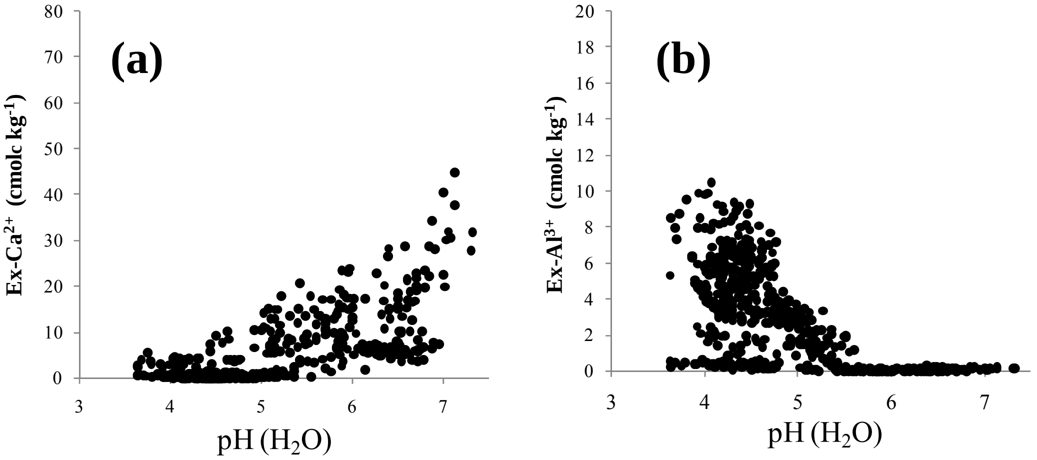

Figure 5.2.2. Scatter plot between pH and Ex-Ca^{2+} (a), and between pH and Ex-Al^{3+} (b) in 0-10 and 10-20 cm from 2000 to 2009, in all monitoring site except Russia.

5.2.2.2.Temporal trend of soil chemical properties in 2000–2009

Figures 5.2.3, 5.2.4 and 5.2.5 show the temporal trend in pH (H_2O), base saturation and exchangeable acidity in 0-10 cm, respectively. It is discussed the temporal variation in some monitoring sites where soil survey had been conducted twice or more.

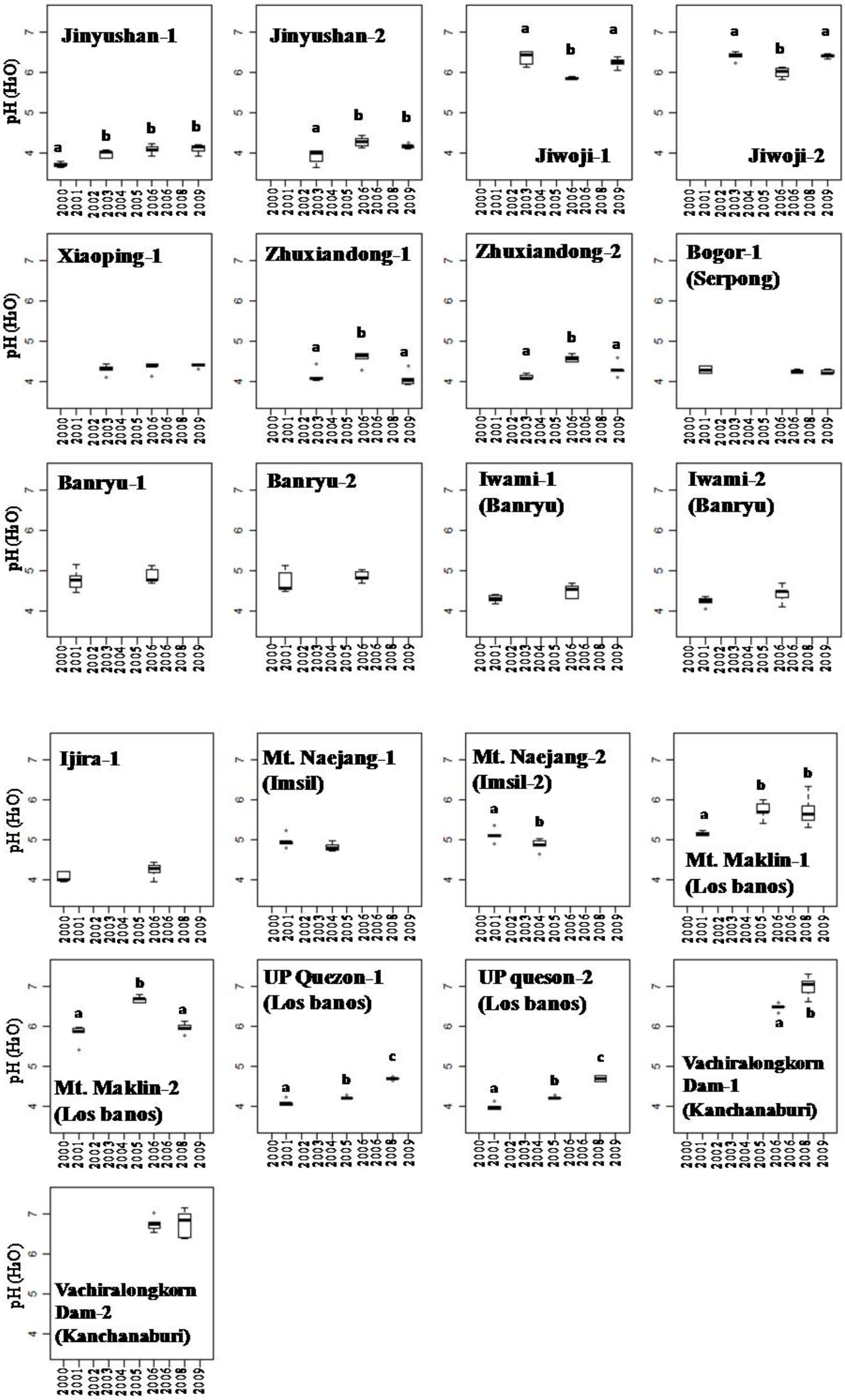

Figure 5.2.3. Temporal variation for 2000-2009 in soil pH in 0-10 cm depth of each plot (n=5)

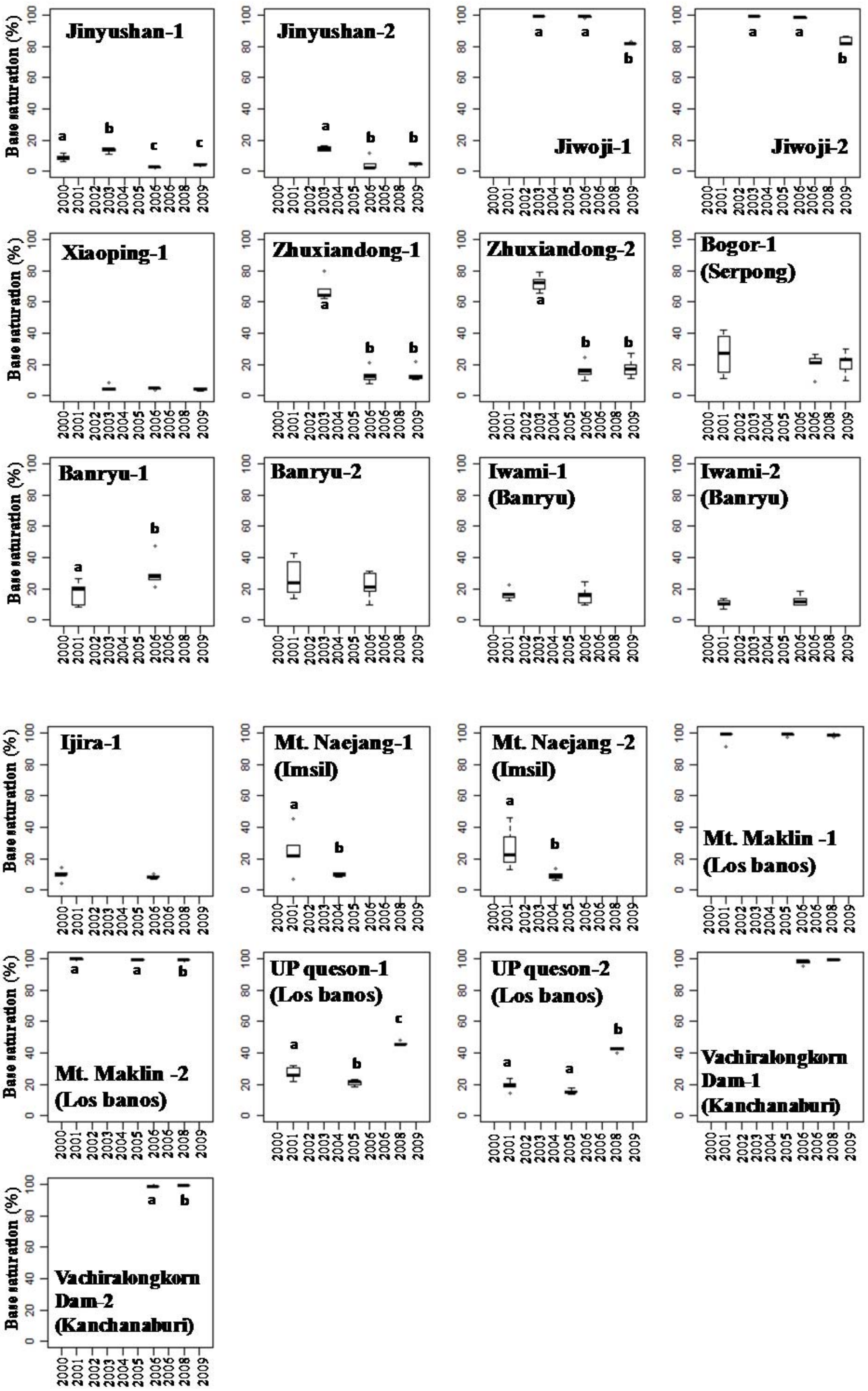

Figure 5.2.4. Temporal variation for 2000-2009 in base saturation in 0-10 cm depth of each plot (n=5).

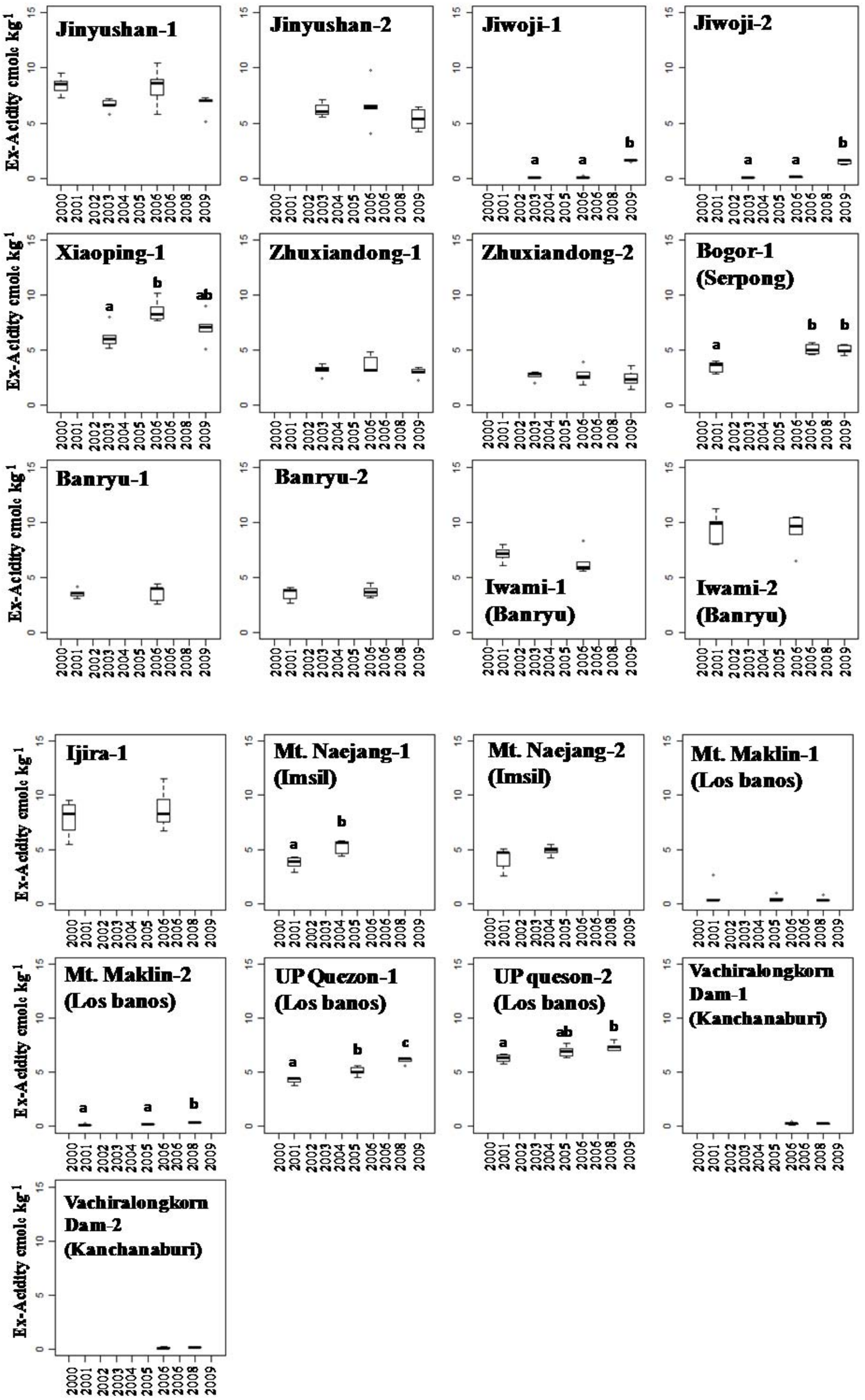

Figure 5.2.5. Temporal variation for 2000-2009 in exchangeable acidity in 0-10 cm depth of each plot (n=5).

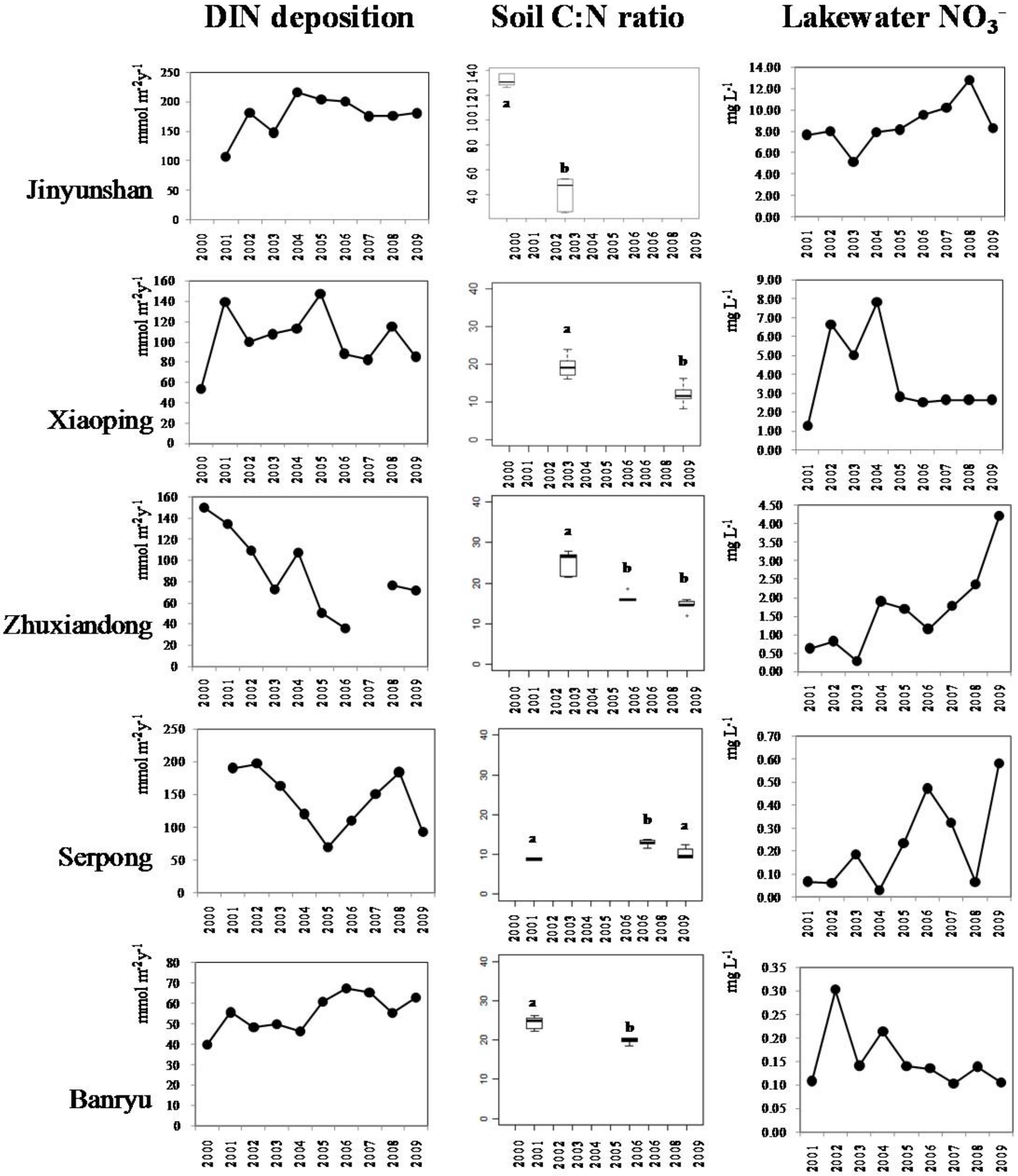

Figure 5.2.6. Comparison between DIN (NO_3^- + NH_4^+) deposition, C/N in surface soil and NO_3^- concentration in inland water of 5 monitoring sites. *In Serpong area, Bogor for soil and Patengang Lake for inland water were compared. *In Zhuxiandong, the sampling point for inland water was changed from lake to stream in 2004.*

In some monitoring sites, it was found an increasing trend in pH (H_2O) in 0-10 cm (Figure 5.2.3). During the period, pH continuously increased in Jinyunshan, Mt. Makiling-1 (Los Baños), UP-Quezon (Los Baños) and Vachiralongkorn Dam-1 (Kanchanaburi), whereas pH decreased only in Mt. Naejang (Imsil). On the other hand, base saturation (Figure 5.2.4) significantly decreased in Jinyunshan from 2003 to 2006, Jiwozi from 2006 to 2009, Zhuxiandong from 2003 to 2006 and Mt. Naejang (Imsil) from 2001 to 2004, whereas base saturations increased in Banryu-1. In UP-Quezon

(Los Baños), base saturation significantly decreased and increased from 2000 to 2005 and from 2005 to 2009, respectively. Exchangeable acidity (Figure 5.2.5) significantly increased in Jiwozi from 2006 to 2009, Xiaoping from 2002 to 2005, Bogor (Serpong) from 2001 to 2006, Mt. Naejang (Imsil) from 2000 to 2004 and UP-Quezon Land Grant (Los Baños) from 2000 to 2009. It was not observed a decreasing trend in exchangeable acidity in any site.

Unlike the regional variation (Figure 5.2.2), pH, acid and base cations did not harmonically change in the temporal variation in many plots because other factors controlling pH (e.g. organic acid) might also change. Because the factors controlling pH may be too complex to clarify in this temporal variation, it was mainly discussed the change of base and acid cations status. It is hypothesized that these temporal trends were caused by nutrient imbalances in the forest ecosystems derived from atmospheric deposition or change of forest conditions, although the materials for clarifying the trend were very limited in the monitoring data.

1) Symptom of "nitrogen saturation" in the forest ecosystems

In several monitoring sites, it is compared temporal trend of soil C/N (total C / total N) to DIN (NO_3^-+ NH_4^+) wet deposition and NO_3^- concentration in the lake or stream water (Figure 5.2.6). Although total C and N concentration in soils were not mandatory items for the regular monitoring, some laboratories have analyzed it optionally. In forest ecosystems, the C/N of the forest leaf litter or top mineral soil is a good indicator of N status related to nitrate leaching. Nitrate leaching increases with increasing N deposition in many European forests where low C/N of forest leaf litter was observed (e.g. Gundersen *et al.*, 1998; Park and Matzner, 2006). This status in a forest is called "nitrogen saturation" (Aber *et al.*, 1989). A terrestrial eutrophication, soil acidification and fresh water eutrophication occur in stages, by "too much" of N input. It is observed a significant decreasing in soil C/N in Jinyunshan, Xiaoping, Zhuxiandong and Banryu (Figure 5.2.6). Meanwhile, in Jinyunshan, Zhuxiandong and Serpong, NO_3^- concentration in the lake increased in 2000-2009. With too much NO_3^- in soil solution, which is indicated by low C/N, it may flow into the stream and lead to NO_3^- increase in the lake. It is assumed that a decreasing in base saturation in the soil in these areas (Figure 5.2.5) might be due to a leaching of base cations accompanied with the NO_3^-(Binkley and Richter, 1987). Increase in N deposition in 2000-2009 in Jinyunshan and Serpong, and high DIN deposition until 2000 in Zhuxiandong (Figure 5.2.6) may lead to this nitrogen saturation in the forest. In Banryu Lake, a decrease in soil C/N also suggests the possible increase in NO_3^- concentration in the future, although the concentration was still low in 2009. The tendency of nitrogen saturation had been reported in other study sites of subtropical China (Chen and Mulder 2006, Fang *et al*. 2008 and Fang *et al*. 2009).

2) Effect of forest rehabilitation or degradation

Forest conditions (e.g. growth or degradation) also may affect the soil chemical properties. In UP-Quezon (Los Baños), forest rehabilitation was observed because young trees (DBH < 5cm; Figure 5.2.7) were dominant in 2002 whereas middle size trees (DBH > 7.5) increased in 2008. In the period, basal area, which indicates the biomass in the forest, increased from 8.8 to 13.2 m^2 ha^{-1}. It is assumed that an increasing in the forest biomass leads to a development of soil organic matter in surface soil by acceleration of the litterfall. Soil organic matter may cause 1) increase in base cations by mineralization of it, and 2) increase in ECEC by negative charge on the surface of organic matter (Oades, 1984). Therefore, in UP-Quezon (Los Baños), an increase in base saturation (Figure 5.2.4) with exchangeable acidity (Figure 5.2.5) might be caused by this forest growth with developing organic matter. In Jinyunshan, basal area increased from 2003 to 2006, whereas it decreased from 2006 to 2009. In a personal communication, tree disease or pestilence had occurred in the site from 2006 to 2009, which was supported by a decrease in young trees in the period (DBH < 5 cm; Figure 5.2.7). It is assumed that organic layer was decreased by this degradation of the forest. Therefore, base cations (Figure 5.2.4) might decrease because mineralization also declined by low resources derived from organic matter. It is speculated that pH increased with the decrease in the organic acid in the forest floor (Figure 5.2.3). Meanwhile, in strongly weathered soil in tropical area, too rapid growth of forest (e.g. fast wood plantation) may result in pH decline due to translocation of the base cations from soil to plant biomass (e.g. Binkley and Giardina, 1997; McGrath *et al.*, 2001; Sanchez, 1985; Yamashita *et al.*, 2008). Soil chemical properties in each monitoring site can easily change by complex effects derived from forest conditions.

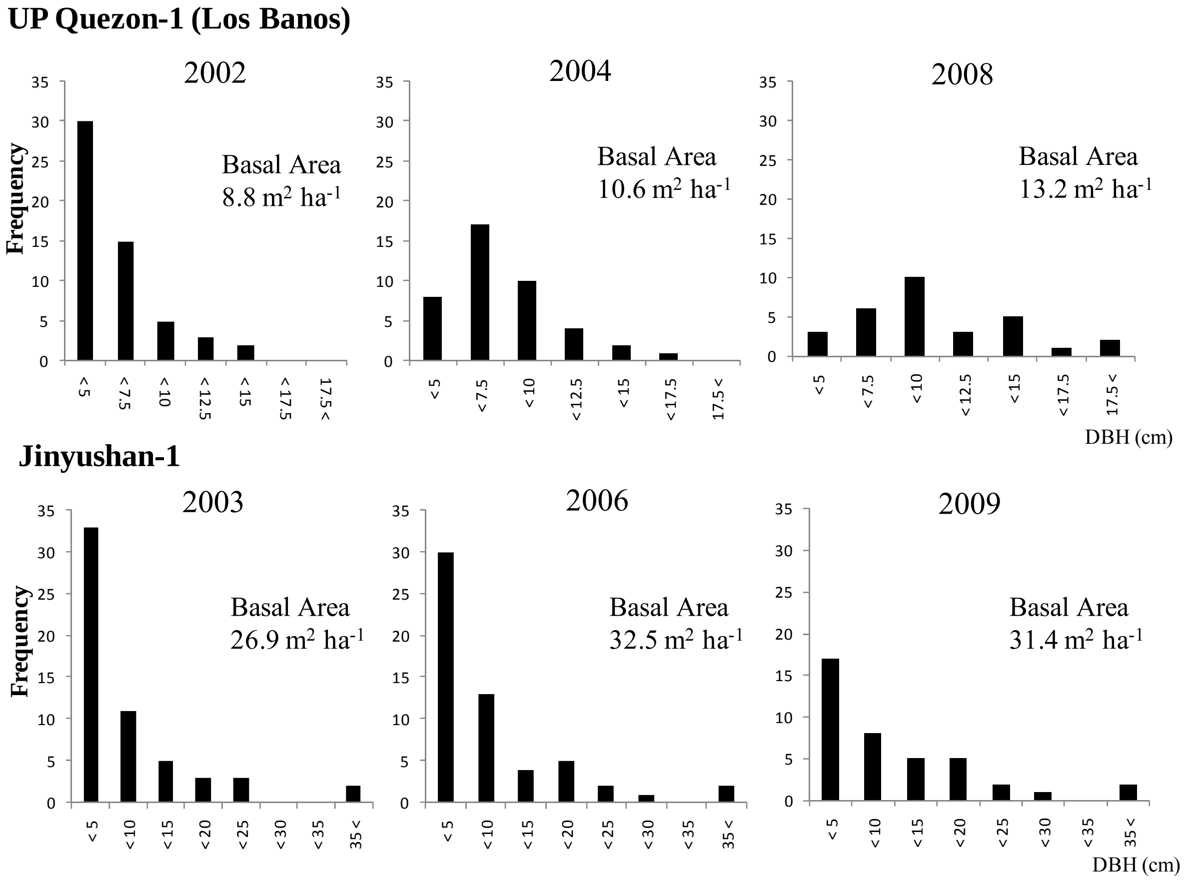

Figure 5.2.7.Temporal change of frequency distribution of DBH in small circle plot (200 m^2).

3) Effect of seasonal variation in dry forest

In addition to the inter-annual variation described above, a seasonal variation was also observed in Vachiralongkorn (Kanchanaburi) plots in 2003 and 2006 (Table 5.2.3). Soil pH (H_2O) significantly increased from the beginning of the wet season to the beginning of the dry season in some plots in 2003. It is assumed that the seasonal variation was derived from strong phenology (biological seasonality) of defoliation in Tropical dry forest, which is the dominant forest in Thailand, and represents more than 40% in tropical forest area (Murphy and Lugo, 1986). Because forest floor is quickly accumulated by defoliation during the dry season and is decomposed during the wet season, a lot of base cations derived from forest floor may flow into the surface soil and increase pH (H_2O) in the soil (Yamashita *et al*., 2010).

Table 5.2.3. Seasonal differences of soil pH in 2003 and 2006 in Vachiralongkorn (Kanchanaburi) site, soil depth are 0-20 cm in 2003 and 0-10 cm in 2006.

year	Plot	pH(H2O)			pH(KCl)		
		Apr.	Dec.	p	Apr.	Dec.	p
2003	Dam1	5.8	6.4	<0.05	5.7	5.8	
	Dam2	6.0	6.3		5.7	5.7	
	Puye	4.6	5.3	<0.05	4.4	4.3	
2006	Dam1	6.4	6.5		5.4	5.4	
	Dam2	6.5	6.7		5.3	5.6	
	Puye	5.2	5.4		4.1	4.2	

4) Interactions between soil, forest, inland water and atmospheric deposition

In conclusion, an increase in NO_3^- concentration in inland water (lake or stream) and a decrease in soil C/N in Jinyunshan, Zhuxiandong and Bogor (Serpong) might indicate the acceleration of the N flux in the forest ecosystems and the NO_3^- leaching into the stream, which is called nitrogen saturation. High N deposition in the area may lead to this nitrogen saturation in the forest. As a result, in Jinyunshan and Zhuxiandong, base saturation in the soil might decrease by leaching of base cations with NO_3^-, whereas soil pH did not harmonically connect to this mechanism because soil pH might be controlled by other factors like an organic acid. Meanwhile, in UP-Quezon (Los Baños), forest rehabilitation may lead to the increase in soil pH by growth of soil organic matter. This suggests that forest conditions (growth or degradation) are also key factors controlling soil chemical properties. In addition, for the comprehensive assessment of temporal variation, a seasonal variation in soil chemical properties should be also considered in the forest which has strong phenology.

5.3.Vegetation features and trends

5.3.1. Vegetation features in EANET sites

The forest vegetation monitoring plots varied in terms of species composition, from monocultures (pure stands) to mixed stands dominated by a single species to highly mixed stands where no species dominate. The pure stands are mostly conifers like the *Pinus elliottii, Pinus massoniana*, and *Chamaecyparis obtusa*. There are also pure stands of *Michelia macclurei* and *Acacia confusa*. Pure stands comprised 33% of the plots established (Figure 5.3.1). The rest are mixed stands. Of the 13 forest vegetation monitoring plots in China, six (46%) are pure stands. The single plots in Mongolia and Korea are both pure stands too. All the forest vegetation monitoring plots in the Philippines and Japan are mixed stands. Four plots in Russia are mixed stands as well and the remaining one in Mondy, a pure forest. The single plot in Thailand is a mixed stand as well.

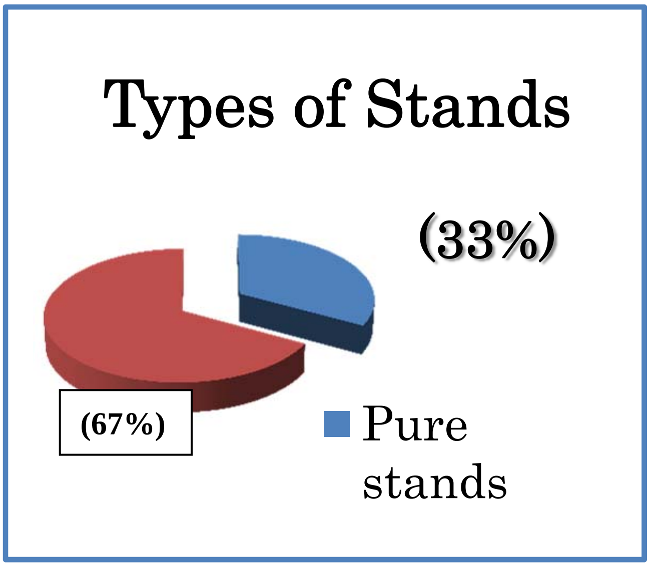

Figure 5.3.1. Types of forests monitored for acid deposition impacts in the East Asian region.

The mixed stands have highly variable species composition. As expected, the more temperate the conditions, the lesser the number of species found. Species increase in number as conditions become more tropical in nature. From a low of two species in Preemnaya, Russia, there can be as many as 57 species found in the UP Los Baños Land Grant in the Philippines. The smallest number of trees in a monitoring plot is found in Fernland, China. There are economically and commercially important tree species in some of the monitoring plots like those of the dipterocarps in Thailand (*Dipterocarpus turbinatus*) and the Philippines (*Parashorea malaanonan, Hopea foxworthyii,* and *Shorea contorta*).

The vegetation distribution in the EANET sites consists of tropical and subtropical moist broadleaf forest to tropical and subtropical dry broadleaf forest to temperate broadleaf and mixed forest (Olson *et al.*, 2001). The type of forest ranged from natural forest to plantation forests.

Sites in China and Japan have some very distinct species dominance which are useful in evaluating the possible changes in the species composition. The genus *Castanopsis,* for instance, is a keystone species in its respective ecosystem. This species has been monitored in China at the Jinyunshan Plot No. 1. A keystone species is a species that plays a critical role in maintaining the structure of an ecological community and exerts impact on the community that is greater than what would be expected based on its relative abundance or total biomass. Hence, the ecosystem may experience a dramatic shift if the keystone species is removed, even though that species was a small part of the ecosystem.

On the other hand, *Fokienia hodginsii* is a near threatened species according to the IUCN Red List. The tree does not tolerate shade, and requires mild climate and abundant rainfall to grow best. *Acacia confusa*, a perennial tree native to Southeast Asia is planted as plantation crop in China. It is considered an invasive species in Hawaii. *Michelia macclurei* is a broadleaf evergreen species. A number of other *Pinus* species were also monitored in China.

5.3.2. Forest vegetation trends

5.3.2.1.Tree decline and tree growth

Overall vitality decline of *Pinus armandi* Fanch was observed in Jiwozi (Dabagou, China, since 2003 and was reported to be due to dense planting, while *Pinus massoniana* in Zhuxiandong-1 since 2003 was due to pest infestation. Decline in *Acacia confusa in* Zhuxiandong-2 was also reported while not noted in *A.confusa* in Zhuxiandong-1**.**

In Primorskaya, Russia, all monitored trees showed symptoms of decline and was reported to be caused by pest infestation in 2006. Likewise *Chamaecyparis obtusa* were reported to have occasional pest and disease infestation that caused reduced vitality in the stand in Lake Ijira, Japan.

In the Philippines, Thailand and Indonesia, tree decline assessment were not reported during 2000-2009, probably due to the dense canopy cover of the forest. However, in 2008, tree decline was reported in Philippines and was noted to be due to termite infestation.

Increase in DBH and height had been noted when compared between the 2000-2004 (PRSAD 1) and 2005-2009 (PRSAD 2) periods, except average diameter reduction at Mt. Makiling in the Philippines (Figure 5.3.2 and 5.3.3). The decline in diameter of trees in the stand could be due to the death of some large canopy trees in the area.

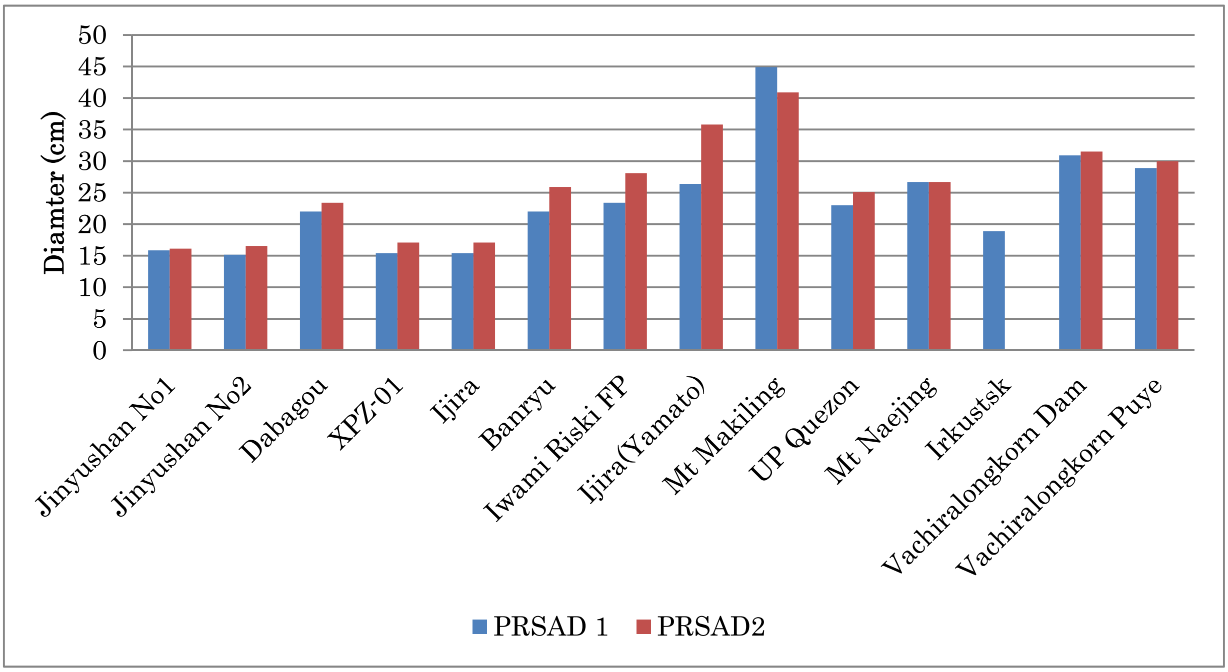

Figure 5.3.2. Changes in average diameter at breast height (DBH) between PRSAD1 and PRSAD2.

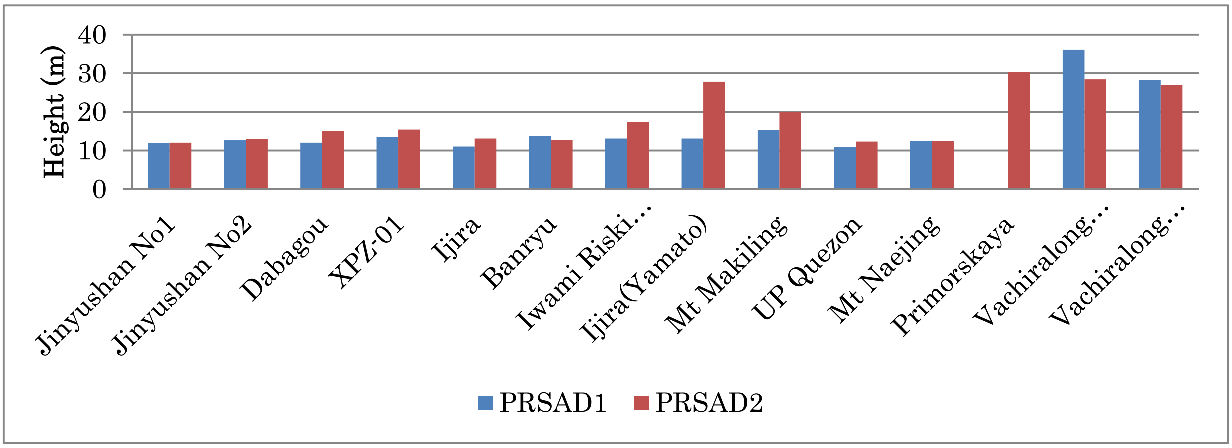

Figure 5.3.3. Changes in average height between PRSAD 1 and PRSAD2.

Pest populations are frequently regulated below their potential levels of abundance by natural enemies, host resistance, or other biological interactions. However, if these regulating processes operate imperfectly, or are intolerant to variations in pest density, then it may be observed periodic outbreaks of the pest (Berryman, 1982). A number of sites had reported incidence of pest outbreaks during the monitoring period, but did not indicate if such was a seasonal or a continuous outbreak.

It was reported that growth rate reduction reached an average of 17% compared with the healthy one in sugar maple in Canada (Duchesne L *et al*, 2002). Although multiple causal agents have been identified to cause the decline in sugar maple, atmospheric acid deposition (N and S) and soil exchangeable acidity were positively associated with stand decline rate, and negatively with the average tree basal area increment trend. The appearance of the forest decline phenomenon in Québec can be attributed, at least partially, to soil acidification and acid deposition levels.

Castanopsis is a keystone species, hence, it is good to monitor the species as decline in the same will affect the composition and functions of the ecosystem. It was reported that about 40 understory species is found in the Jinyunshan Plot No 1.

No incidence of decline is reported for *Fokienia hodginsii* which suggests that this species is not affected by the acid deposition thus far. It is important to continue monitoring this species because of its near threatened status in the IUCN Red List.

The temporal changes in soil pH may indicate the deposition load in the soil. But reduced decomposition rates due to increased soil moisture resulting from increased rainfall may also reduce the soil pH. No report is available on leaf litter trends to determine if leaf litter is increasing. If crown density is reduced, then the leaf litter must increase making it important to determine the decomposition rates.

5.3.2.2.Changes in understory plant species

The number of understory species increased between PRSAD1 and PRSAD2 except in Vachiralongkorn Puye (Figure 5.3.4). The increase in recruitment between these two periods indicated that the overall forest functions and structure are maintained. Most recruitments are generalist.

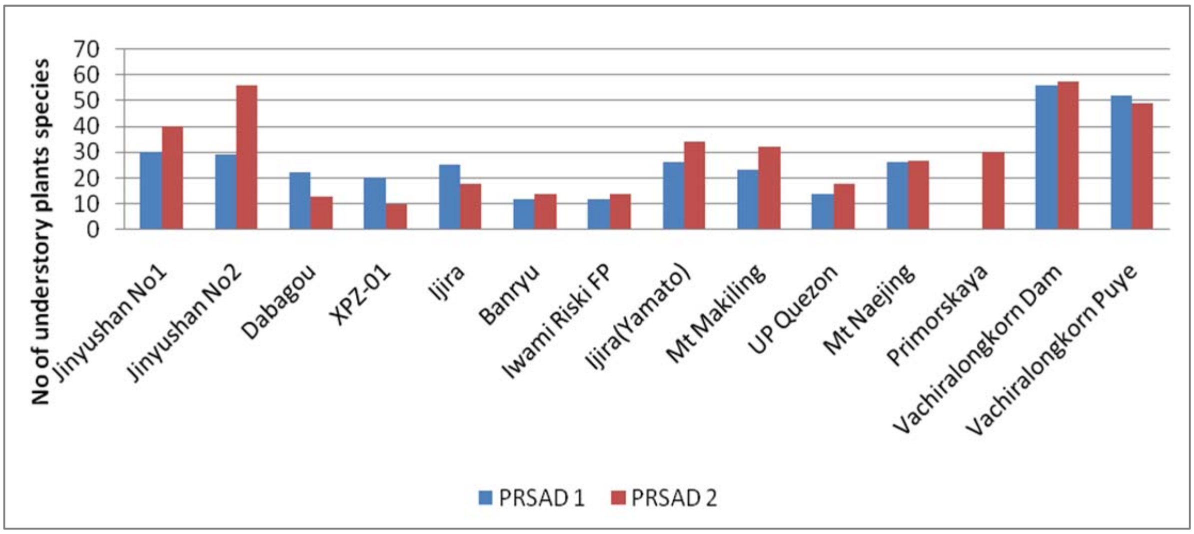

Figure 5.3.4. Changes in numbers of understory plants species (new recruitment).

Overall, there is no decline in growth and new recruitments in the EANET ecological monitoring sites between the two reporting periods. The decline symptoms observed were often temporary in nature and were caused by biotic causal agents.

5.4. Inland water and trend

5.4.1. Features on inland aquatic environment in EANET sites

The EANET has 18 sites (as of the year 2009) for monitoring on inland aquatic environment, namely four sites in China, two sites each in Indonesia, Japan, Malaysia, Philippines, and Russia, and one site each in Lao PDR, Mongolia, Thailand and Vietnam. The name, start year, and monitoring interval are shown in Table 5.4.1. Six rivers and twelve lakes are selected as the monitoring sites. Among the lakes, six artificial lakes (reservoirs/dam) and six natural lakes are included. In this section, characterization of inland aquatic environment in EANET sites is

Table 5.4.1. Outline of inland aquatic environment monitoring of EANET (EANET, 2009).

Country	Name of site	Distance from the site	Nearest deposition monitoring site	Start year	Interval
China	Jinyunshan Lake (Chongqing)	-	Rural	2001	4times/yr.
	Xiaoping Dam (Xiamen)	-	Remote	2001	4times/yr.
	Jiwozi River (Xi'an)	-	Remote	2001	4times/yr.
	Zhuxiandong Stream (Zhuhai)	-	Urban	2004	4times/yr.
Indonesia	Patengang Lake (Bandung)	47km	Urban	2001	5times/yr.
	Gunung Lake (Sukabumi)	-	-	2008	5times/yr
Japan	Ijira Lake (Gifu Pref.)	1.1km	Rural/Ecolog. (Ijira)	2001	4times/yr.
	Banryu Lake (Shimane Pref.)	0.5km	Urban/Ecolog. (Banryu)	2001	4times/yr.
Lao PDR	Nam Hum Lake (Vientiane province)	-	Vientiane	2009	2times/yr.
Malaysia	Semenyih Dam	26km	Urban (Petaling Jaya)	2005	3times/yr.
	Tembaling River (Sabah)	-	Remote (Danum Valley)	2007	4times/yr.
Mongolia	Terelj River	-	Remote (Terelj)	2002	6times/yr.
Philippines	Pandin Lake (Los Banos)	30km	Rural (	2004	4times/yr.
	Ambulalakao Lake	-	Remote (Mt. Sto. Tomas)	2005	2times/yr.
Russia	Pereemnaya River (Listvyanka)	48km	Rural (Listvyanka)	2004	4times/yr.
	Komarovka River (Primorskaya)	0.5km	Rural (Primorskaya)	2005	5times/yr.
Thailand	Vachiralongkorn Dam (Kanchanaburi Province)	-	Remote	2002	4times/yr.
Vietnam	Hoa Binh Reservoir	-	Rural	2001	4times/yr.

discussed with their general properties and water chemistry.

5.4.1.1. General properties of sites

Inland water can be considered as an output from its watershed. Its chemistry may be decided primarily based on biogeochemical processes in the watershed. Moreover, in case of lakes, their size, including the area, water depth, water volume, and other conditions such as residential time of the water may affect the water chemistry. Thus, general properties of sites are important to interpret the inland water chemistry. *The Technical Manual for Monitoring on Inland Aquatic Environment* (EANET 2000) requested surveyors to compile the general properties of the monitoring sites. However, the information recorded for the sites are still inadequate. Based on the available data, it was tried to provide an overview of the EANET monitoring sites.

1) Artificial lakes

Artificial lakes were constructed as dam or reservoir. However, their characteristics, such as area, water depth, water volume, and watershed area, were very varied probably depending on their purposes. For example, the area of Vachiralongkorn Dam, which was constructed in a main river for generating electricity, was quite large, 370,000 times larger than that of Jinyunshan Lake, which was constructed for irrigating agricultural fields. Similarly, the watershed of Hoa Binh Reservoir for electricity was 10,000 times larger than that of Ijira Lake built for irrigation. Maximum and minimum values of the available information were shown in Table 5.4.2.

Table 5.4.2. Maximum and minimum values of the major properties of artificial lakes in the EANET monitoring sites on inland aquatic environment.

Property	Maximum	Minimum	No data
Area	3720 km^2 Vachiralongkorn Dam in Thailand	0.00999 km^2 Jinyunshan Lake in China	
Shore line length	16.7 km Hoa Binh Reservoir in Vietnam	100 m Xiaoping Dam in China	Vachiralongkorn Dam in Thailand
Water depth	153.21 m Vachiralongkorn Dam in Thailand	5.4 m Ijira Lake in Japan	
Water volume	6.7276 km^3 Vachiralongkorn Dam in Thailand	60,000 m^3 Jinyunshan Lake in China	Semenyih Dam in Malaysia
Watershed area:	51,700 km^2 Hoa Binh Reservoir in Vietnam	5.4 km^2 Ijira Lake in Japan	Jinyunshan Lake in China and Semenyih Dam in Malaysia
Annual water level fluctuation	9 m average, Vachiralongkorn Dam in Thailand	0.22m Ijira Lake in Japan	Xiaoping Dam in China and Hoa Binh Reservoir in Vietnam
Residence time of water	more than 1 year Semenyih Dam in Malaysia	15 days Xiaoping Dam in China	Jinyunshan Lake in China
Altitude	595 m Xiaoping Dam in China	23 m Hoa Binh Reservoir in Vietnam	Jinyunshan Lake in China
Precipitation (annual)	2500 mm $year^{-1}$ Xiaoping Dam in China	1198.9 mm $year^{-1}$ Jinyunshan Lake in China	Semenyih Dam in Malaysia and Hoa Binh Reservoir in Vietnam
Lake Utilization	Most of lakes are for Irrigation, next is for Electric power then for Drinking water and Fishing		

Note: Original information can be found in the annual Data Report of EANET.

2) Natural lakes

The information on the natural lakes was very limited. Even for the precipitation, only the data on three sites were submitted. The area, water depth, and water volume were varied. Although the watershed area was not clear for these sites, the difference of the water volume between maximum and minimum suggested also variations of the watershed area. Maximum and minimum values of the available information are shown in Table 5.4.3.

Table 5.4.3. Maximum and minimum values of the major properties of natural lakes in the *EANET monitoring sites on inland aquatic environment.*

Property	Maximum	Minimum	No data
Area	0.65 km^2 Patengang Lake in Indonesia	0.006087 km^2 Ambulalakao Lake in Philippines	Nam Hum Lake in Lao PDR
Water depth	61.75 m Pandin Lake in Philippines	1.5 m Ambulalakao Lake in Philippines	Nam Hum Lake in Lao PDR
Water volume	4.98×10^6 m^3 Patengang Lake in Indonesia	6600 m^3 Pandin Lake - Philippines	Gunung Lake in Indonesia and Nam Hum Lake in Lao PDR
Altitude	2400 m Ambulalakao Lake in Philippines	25m Banryu Lake in Japan	
Precipitation (annual)	Patengang Lake in Indonesia: 2357 – 2686 mm year^{-1} Gunung Lake in Indonesia: 1611 – 4311 mm year^{-1} Banryu Lake: 1,749.5mm year^{-1}		Nam Hum Lake in Lao PDR and 2 sites in Philippines
Lake Utilization	Most of lakes are for Irrigation, the others are for Tourism / Sightseeing and Power.		

Note: Original information can be found in the annual Data Report of EANET.

3) Rivers

Features of the watershed may be important factors to interpret the water chemistry, especially for rivers. However, important properties, such as water discharge, vegetation, soil type, geology etc. have not been compiled for many sites. The watershed areas of these sites vary from 360 km^2 to 1,490 km^2 (in Russian Federation sites). The annual precipitation changes from 240.6 mm yr^{-1} (Terelj River in Mongolia) to 1,996.5 mm yr^{-1} (Zhuxiandong Stream in China).

Some important properties have not been compiled in several sites. Efforts should be made to record the important information on the monitoring sites for effective interpretation of the monitoring data in the future.

5.4.1.2. Characteristics of the monitoring sites

In order to characterize the sites, the Piper tri-linear diagram was applied for the total average values of the respective sites (Figure 5.4.1). The tri-linear diagram can show balances among the major parameters, which is often used for characterization of inland waters. The center diamond plot, the left triangle plot and the right triangle plot show a balance between anions and cations, balance among cations, and balance among anions, respectively.

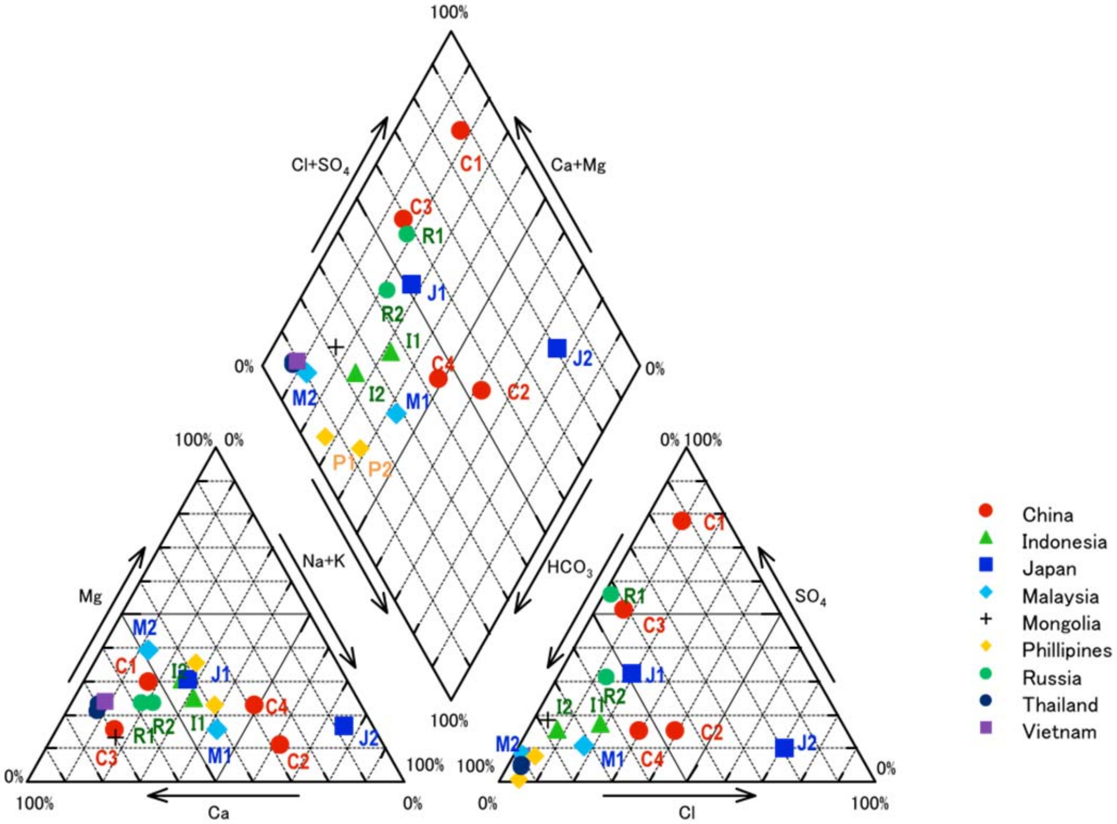

Figure 5.4.1. Piper trilinear diagram of the inland water chemistry from the EANET sites. The total mean for the monitoring period was applied for the plot. HCO_3^- was based on alkalinity. C1, Jinyunshan Lake; C2, Xiaoping Dam; C3, Jiwozi River; C4, Zhuxiandong Stream; I1, Patengang Lake; I2, Gunung Lake; J1, Ijira Lake; J2, Banryu Lake; M1, Semenih Dam; M2, Tembaling River; P1, Panding Lake; P2, Ambulalakao Lake; R1, Pereemnaya River; R2, Komarovka River. Marks for Mongolia, Thailand, and Vietnam showed the data from Terelj River, Vachiralongkorn Dam and Hoa Binh Reservoir, respectively.

Jinyunshan Lake (shown as C1) showed peculiar positions in the center and right plots. The water of Jinyunshan Lake was rich in SO_4^{2-} and Ca^{2+} but poor in alkalinity. Actually, as shown in Appendix Table 5.1, the concentration of SO_4^{2-} was among the highest in the sites and alkalinity was the second lowest. The pH was the lowest. The water may be sensitive to acid deposition, although the clear reason for the sensitivity has not been reported yet. Jiwozi River (as C3), Pereemnaya River (as R1), and Ijira Lake (as J1) were plotted near Jinyunshan Lake in the center plot.

Banryu Lake (shown as J2) showed different peculiar positions in three plots. The water in Banryu Lake was rich in Cl^- and Na^+. As shown in Table 5.4.3, Banryu Lake is located at 25 m above sea level. Actually, the lake was very close to the seashore. It was therefore suggested that the lake is affected by sea salt. Xiaoping Dam was plotted near Banryu Lake in the center plot.

Vachiralongkorn Dam, Hoa Binh Reservoir and Tembaling River (as M2) were plotted in the left corner in the center plot. The water of these lakes/rivers may be rich in Ca^{2+} and alkalinity. Although information on surface geology is not clear in most sites, it has been well known that Vachiralongkorn Dam was located in a limestone area. The water chemistry may reflect the geology of the watershed.

5.4.2. Trend of inland water

Based on accumulated data in the EANET sites above, trend analysis was conducted for major parameters. The data obtained by quarterly sampling were analyzed through the non-parametric Seasonal Mann-Kendall method (Hirsch *et al.*, 1982). In several sites, the data in 2000 were also used for the analysis in addition to the data shown in Table 5.1.1. Results of the trend analysis are shown in Table 5.4.4. The data in Gunung Lake, Tembaling River, Nam Hum Lake, Ambulalakao Lake were not analyzed yet since there were not enough data sets to do this.

Table 5.4.4. Results of seasonal Mann-Kendall trend analysis for inland water of the EANET sites.

Country	site	pH	EC	Alkalinity	SO_4^{2-}	NO_3^-	Cl^-	NH_4^+	Na^+	K^+	Ca^{2+}	Mg^{2+}
China	Jinyunsyan Lake	-3.34 ***	4.34 ***	-2.69 **	3.44 ***	2.50 *	2.04 *	0.10	1.73	2.72 **	3.81 ***	2.67 **
	Xiaopin Dam	-2.35 *	0.06	-0.91	2.40 *	0.21	4.59 ***	-0.37	-0.58	2.60 **	1.88	-1.74
	Jiwozi River	-2.11 *	2.92 **	3.46 ***	3.68 ***	0.87	-0.22	1.90	-1.62	1.08	-1.19	-2.17 *
	Zhuxiandong Stream	0.39	-1.88	-0.38	-2.27 *	-0.09	-0.28	1.16	-2.19 *	-1.01	-2.88 **	-0.50
Indonesia	Patengang Lake	-2.36 *	-1.79	-1.40	2.35 *	1.23	-3.98 ***	-0.11	-2.57 *	1.74	0.06	-1.62
Japan	Ijira Lake	1.49	0.65	3.19 **	1.06	-3.01 **	-3.71 ***	-2.00 *	-2.55 *	-3.01 **	-1.02	-2.36 *
	Kamagatani River flowing river to Ijira Lake	3.05 **	1.79	3.81 ***	1.25	-1.39	-2.68 **	-3.41 ***	-0.40	-1.93	0.27	-1.21
	Kobara River flowing river to Ijira Lake	2.47 *	1.83	3.09 **	2.34 *	-1.70	-1.78	-2.74 **	0.32	-1.45	-0.44	-1.20
	Banryu Lake	-0.67	2.47 *	-0.72	1.17	2.60 **	2.41 *	0.45	3.98 ***	1.34	2.68 **	1.57
	Banryu Lake 3	-0.67	2.20 *	-0.49	1.57	1.86	2.07 *	-0.59	4.04 ***	1.39	2.25 *	0.67
Malaysia	Semenih Dam	-2.87 **	0.27	0.46	2.09 *	-1.59	-2.01 *	-1.88	-1.69	-0.94	-0.27	-0.78
Mongolia	Terelj River	0.60	1.53	2.69 **	1.37	-1.04	1.43	-2.75 **	1.42	-1.43	1.09	-0.27
Phillipines	Pandin Lake	-1.09	-0.89	-1.40	3.27 **	3.91 ***	2.08 *	0.73	-1.40	-1.49	0.40	0.30
Russia	Pereemnaya River	-0.94	4.46 ***	0.59	4.70 ***	2.63 **	-2.55 *	-0.29	-1.79	-3.38 ***	5.07 ***	-0.19
	Komarovka River	-0.37	0.49	-3.32 ***	2.92 **	0.49	1.35	1.46	1.35	0.25	-2.33 *	-1.22
Thailand	Vachiralongkorn Dam 1 Ban Pong Chang	0.87	-2.98 **	0.57	-2.07 *	1.81	-3.85 ***	1.58	-0.55	-1.80	-0.05	-0.50
	Vachiralongkorn Dam 2 Ban Pang Pueng	1.45	-3.39 ***	1.06	-1.34	2.44 *	-4.57 ***	-0.27	-1.46	-0.55	-1.30	-2.45 *
Virtnam	Hoa Binh Reservoir	0.36	1.61	-1.37	1.61	1.70	1.12	-3.02 **	2.41 *	0.00	1.61	-2.96 **

*Note: Value shows Z score, which indicates the increasing trend with positive values or decreasing trend with negative values. An asterisk shows significant levels; *, $p < 0.05$, **, $p < 0.01$, and ***, $p < 0.001$. Sampling frequency: 4 times a year, except for Terelj River (6 times a year) and Komorovka River (5 times a year).*

5.4.2.1. Sites in Indonesia, Malaysia, Thailand, and the Philippines

The monitoring was conducted in Patengan Lake (2000 to 2009) and Gunung Lake (2008 to 2009) in Indonesia, Semenyih Dam (2005 to 2009) and Tembaling River (2007 to 2009) in Malaysia, Vachilongkorn Dam (two stations, 2000 to 2009) in Thailand, and Pandin Lake (2004 to 2009) and Ambulalakao lake (2007-2009) in the Philippines. Changes in annual average values were shown in Figure 5.4.2 a) and b). Results of the trend analysis for the major parameters are as follows:

1) pH

   In Patengan Lake, the pH decreased significantly ($p < 0.05$). The pH in Semenyih Dam also decreased significantly ($p < 0.01$). In Pandin Lake and Vachiralongkorn Dam, the pH did not show any significant changes.

2) Electric Conductivity (EC)

   In Vachiralongkorn Dam, the ECdecreased significantly at Ban Pong Chang station ($p < 0.01$) and at Ban Pang Pueng station ($p < 0.001$), ranging from 9.10 to 19 $mS.m^{-1}$(average, 11.6 and SD, 2.01) and 9.15 – 15 (average, 10.73 and SD, 1.32), respectively. In Patengan Lake, Semenyih Dam and Pandin Lake, the EC did not show significant trend.

3) Alkalinity

   In Patengan Lake, Semenyih Dam, Vachiralongkorn Dam and Pandin Lake, alkalinity did not show significant trend.

4) Sulphate (SO_4^{2-})

   In Vachiralongkorn Dam, the concentration of SO_4^{2-} decreased significantly ($p < 0.05$) at Ban Pong Chang station, ranging from 0.98 to 31 $mg.l^{-1}$(average, 3.3 and SD, 5.37). The concentration increased significantly in Patengan Lake ($p < 0.05$), Semenyih Dam ($p < 0.05$) and Pandin Lake ($p < 0.01$), ranging from 1.8 to 12 (average, 6.0 and SD, 2.6), from 0.62 to 11.34 (average, 2.23 and SD, 3.17), and from 0.09 to 2.99 (average, 0.71 and SD, 0.57), respectively.

5) Nitrate (NO_3^-)

   At Ban Pong Chang station in Vachiralongkorn Dam, the concentration increased significantly ($p < 0.05$), ranging from 0.01 – 0.20 $mg.l^{-1}$(average, 0.10 and SD, 0.09). In Pandin Lake, the concentration increased siginificantly ($p < 0.001$), ranging from 0.07 to 7.91 (average, 2.3 and SD, 2.77). In Patengan Lake and Semenyih Dam, the NO_3^- concentration did not show significant trend.

6) Chloride (Cl^-)

   The Cl^- concentrations decreased significantly in PatenganLake($p < 0.001$), Semenyih Dam ($p < 0.05$) and both stations of Vachiralongkorn Dam($p < 0.001$).The concentration in Pandin Lake increased significantly ($p < 0.05$).

7) Amonium (NH_4^+)

   The concentration of NH_4^+did not show significant trend.

8) Sodium (Na^+)

   In PatenganLake, the concentration of Na^+ decreased significantly ($p < 0.05$). The concentrations in Semenyih Dam, Vachiralongkorn Dam and Pandin Lake did not show significant trend.

9) Potasium (K^+)

   The concentrations of K^+did not show significant trend.

10) Calcium (Ca^{2+})

    The concentration of Ca^{2+} did not show significant trend.

11) Magnesium (Mg^{2+})

    At Ban Pang Pueng station of Vachiralongkorn Dam, the concentration of Mg^{2+} decreased significantly ($p < 0.05$). In other sites, the concentration did not show significant trend.

5.4.2.2. Sites in China, Japan and Vietnam

The monitoring was conducted in Jinyunsyan Lake (2001 to 2009), Xiaoping Dam (2001 to 2009), Jiwozi River (2001 to 2009) and Zhuxiandong Stream (2004 to 2009) in China, in Ijira Lake and its two flowing rivers (2000 to 2009) and two stations of Banryu Lake (2000 to 2009) in Japan and in Hoa Binh Reservoir (2000 to 2009) in Vietnam. Changes in annual average values are shown in Figure 5.4.3 a) and b). Results of the trend analysis for the major parameters are as follows:

1) pH

   The pH decreased significantly in Jinyushan lake ($p < 0.001$), Xiaoping Dam ($p < 0.05$), Jiwozi River ($p < 0.05$). Inversely, the pH increased significantly in the two inflowing rivers to Ijira Lake, Kamagatani River ($p < 0.01$) and Kobora River($p < 0.05$). In other sites, the pH did not show significant trend.

2) EC

   EC increased significantly in Jinyushan Lake($p < 0.001$), Jiwozi River ($p < 0.01$), and the two stations of Banryu Lake ($p < 0.05$).

3) Alkalinity

   In JinyushanLake, alkalinity decreased significantly ($p < 0.01$), ranging from nd to 0.4 meq.l^{-1} (average, 0.1 and SD, 0.1). In Jiwozi River, Ijira Lake and its flowing rivers, alkalinity increased significantly.

4) Sulphate (SO_4^{2-})

   The concentration of SO_4^{2-}increased siginificantly in Jinyushan Lake ($p < 0.001$), Xiaoping Dam ($p < 0.05$), Jiwozi River ($p < 0.001$) and Kobora River, an inflowing river to Ijira Lake ($p < 0.05$). In Zhuxiandong Stream, the concentration decreased siginificantly ($p < 0.05$), ranging from 1.6 to 7.2 mg.l^{-1}(average, 3.9 and SD, 1.39).

5) Nitrate (NO_3^-)

   The NO_3^-concentration increased significantly in Jinyunshan Lake ($p < 0.05$) and Banryu Lake ($p < 0.01$), ranging from 3.1 to 17.1 mg.l^{-1} (average, 8.6 and SD, 3.2) and from nd to 0.61 (average, 0.13 and SD, 0.19), respectively.In Ijira Lake, the concentration decreased significantly ($p < 0.01$), ranging from 0.7 to 3.4 (average, 2 and SD, 0.6).

6) Chloride (Cl^-)

   The Cl^-concentrations increased significantly in Jinyunshan Lake ($p < 0.05$), Xiaoping Dam ($p < 0.001$), and two stations of Banryu Lake ($p < 0.05$). In Ijira Lake and its inflowing river, Kamagatani River, the concentration increased significantly ($p < 0.001$ and 0.01, respectively).

In Zhuxiandong Stream and Jiwozi River, the concentration was mostly stable and did not show significant trend.

7) Amonium (NH_4^+)

In Ijira Lake, its two inflowing rivers, and Hoa Binh Reservoir, the concentration of NH_4^+ decreased significantly.

8) Sodium (Na^+)

The concentrations of Na^+ in Zhuxiandong Stream and Ijira Lake decreased significantly ($p < 0.05$). In two stations of Banryu Lake, and Hoa Binh Reservoir, the concentrations increased significantly ($p < 0.001$ and 0.05, respectively).

9) Potasium (K^+)

The concentration of K^+increased significantly in Jinyushan Lake ($p < 0.01$) and Xiaoping Dam ($p < 0.01$).In Ijira Lake, the concentration decreased significantly ($p < 0.01$).

10) Calcium (Ca^{2+})

The concentration of Ca^{2+} decreased significantlyin Zhuxiandong Stream, ($p < 0.01$) and increased significantly in Jinyunshan Lake ($p < 0.001$), Banryu Lake ($p < 0.01$) and Banryu Lake 3 ($p < 0.05$).

11) Magnesium (Mg^{2+})

The concentration increased significantlyin Jinyunshan Lake ($p < 0.01$) and decreased in Jiwozi River ($p < 0.05$), Ijira Lake ($p < 0.05$) and Hoa Binh Reservoir ($p < 0.01$).

5.4.2.3. Sites in Mongolia and Russia

The monitoring was conducted in Terelj River (2002 to 2009) in Mongolia and Pereemnaya River (2004-2009) and Komarovka River (2005 -2009). Changes in annual average values were shown in Figure 5.4.4 a) and b). Results of the trend analysis for the major parameters are as follows:

1) pH

The pH did not show significant trend in Terelj River, Pereemnaya River and Komarovka River, ranging from 6.35 to 7.65 (average, 7.04 and SD, 0.27), from 6.19 to 7.12 (average, 6.71 and SD, 0.21), and from 6.8 to 7.51 (average, 7.16 and SD, 0.16), respectively.

2) EC

In Pereemnaya River, EC increased significantly ($p < 0.001$) ranging from 3.33 to 5.77 $mS.m^{-1}$ (average, 4.32 and SD, 0.69). The EC in Terelj River increased slightly.

3) Alkalinity .

The alkalinity decreased significantly in Komarovka River ($p < 0.01$), ranging from 0.29 to 0.57 $meq.l^{-1}$ (average, 0.41 and SD, 0.07), and increased significantly in Terelj River ($p < 0.01$) from 0.07 to 0.55 (average, 0.29 and SD, 0.1). The alkalinity was stable in Pereemnaya River and did not show significant trend.

4) Sulphate (SO_4^{2-})

The concentrations of SO_4^{2-} increased significantly in Pereemnaya River ($p < 0.001$) and Komarovka River ($p < 0.01$), ranging from 5.63 to 14.3 $mg.l^{-1}$(average, 8.82 and SD, 2.17) and from 7.13 to 13.9 (average, 11.08 and SD, 1.94), respectively. The concentration in Terelj River increased but not significantly.

5) Nitrate (NO_3^-)

The concentration of NO_3^- in Preemnaya River increased significantly ($p < 0.01$), ranging from 0.45 to 1.29 mg.l^{-1} (average, 0.84 and SD, 0.26). The concentrations in Komarovka River and Terelj River did not show significant trends.

6) Chloride (Cl^-)

The concentration of Cl^- increased significantly in Pereemnaya River ($p < 0.05$). The concentrations in Komarovka River and Terelj River were stable and did not show significant trends.

7) Amonium (NH_4^+)

The concentrations of NH_4^+decreased significantly in Terelj River ($p < 0.01$).

8) Sodium (Na^+)

The concentrations of Na^+ did not show significant trends in the three sites.

9) Potasium (K^+)

The K^+concentrationin Pereemnaya River decreased significantly ($p < 0.001$). Other two sites did not show significant trends.

10) Calcium (Ca^{2+})

The concentration of Ca^{2+} increased significantly in Pereemnaya River ($p < 0.001$) and decreased in Komarovka River ($p < 0.05$). The concentration in Terelj River did not show significant trend.

11) Magnesium (Mg^{2+})

The concentrations of Mg^{2+}in three sites were mostly stable and did not show significant trends.

5.4.2.4. Discussion

As described above, it was found that the pH of the inland water decreased significantly in five monitoring sites, namely, Jinyunshan Lake, Xiaoping Dam, Jiwozi River, Patengan Lake, and Semenyih Dam during the period from 2000 to 2009. In these acidified lakes/rivers, the concentrations of SO_4^{2-} increased significantly, too. Moreover, the concentration of NO_3^- increased significantly in Jinyunshan Lake. It may be possible that leaching of SO_4^{2-} and/or NO_3^- caused leaching of H^+, resulting in the acidification of the waters.

As described in Section 5.2.2 (Trend of soil chemical properties), the pH of the soil collected in Jinyunshan area increased through the years. Adsorption of anions to the soil may be generally regulated by soil pH. When soil pH is low and free H^+ exists in soil solution, adsorption of SO_4^{2-} is enhanced. Inversely, with increase of soil pH, adsorbed SO_4 become released with H^+ into soil water, and then probably to the lake/river water. Thus, the trend of soil chemical property corresponded well to the trend of lake water chemistry in the case of Jinyunshan Lake. Although the contribution of dry deposition is still uncertain, the wet deposition of non-sea salt (nss) SO_4^{2-} in Jinyunshan is the highest level among the EANET sites; e.g. 124 mmol m^{-2} yr^{-1} in 2009. It made possible the high S deposition affecting the soil and then inland water in Jinyunshan. Moreover, the wet deposition of dissolved inorganic nitrogen (DIN, NO_3^- + NH_4^+) was also the highest level, 181 mmol m^{-2} yr^{-1} (25 kg N ha^{-1}) in 2009, which exceeded the threshold value at 10 kg N ha^{-1}, for leaching of NO_3^- to the inland water (Kamisako *et al*., 2008). As discussed in section 5.2.2, carbon and nitrogen ratio (C/N) in soil was increased in Jinyunshan area, suggesting increase of nitrogen flux in the ecosystem. It was suggested that the leaching of NO_3^- into the lake water was a part of a "nitrogen saturation" attributed to high atmospheric DIN input.

The similar phenomena of acidification and nitrogen saturation were reported in the Lake Ijira catchment in Japan (Nakahara *et al*., 2010) as introduced in Chapter 6. Soil acidification was also observed there but leaching of SO_4^{2-} was observed in the period when the soil pH increased. It was also suggested that nitrogen saturation occurred in the Lake Ijira catchment. However, when we looked at the data from 2000 to 2009, it seemed that the water chemistry in the Lake Ijira catchment have been recovering in this period. The pH in the two inflowing rivers increased, alkalinity in the lake and rivers increased, and the NO_3^- concentration decreased in the lake. However, in case of the Ijira Lake, some meteorological events, such as cold summer and drought summer, may have triggered the imbalance of the ecosystem in this acid-sensitive catchment (Nakahara *et al*., 2010). Therefore, the trend should be carefully checked continuously. Actually, it was only in Kobora River that the concentration of $SO_4^{2-}$is increasing. In particular for the river, it may need to pay more attention in the coming years, whether this is some symptoms for the next imbalance.

The accumulated data for the last years gave us many suggestions on possible acidification or nitrogen saturation of the ecosystems in several EANET sites. It should be carefully discuss relationship among components of the ecosystems, including soil, vegetation, inland water, and atmosphere.

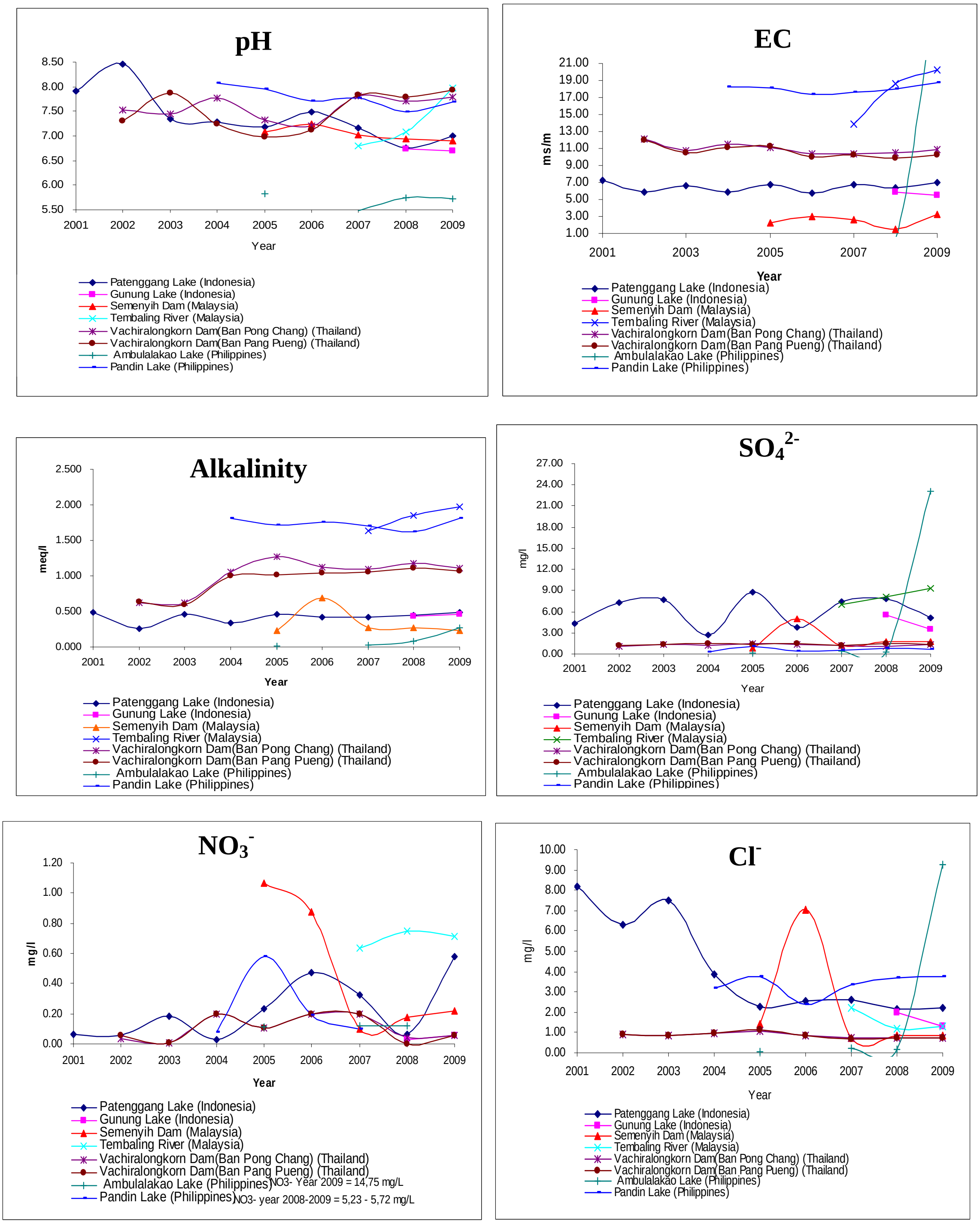

Figure 5.4.2 a). Changes in annual average values of pH, EC, Alkalinity, SO_4^{2-}, NO_3^- and Cl^- in inland water at monitoring sites in Indonesia, Malaysia, Philippines and Thailand.

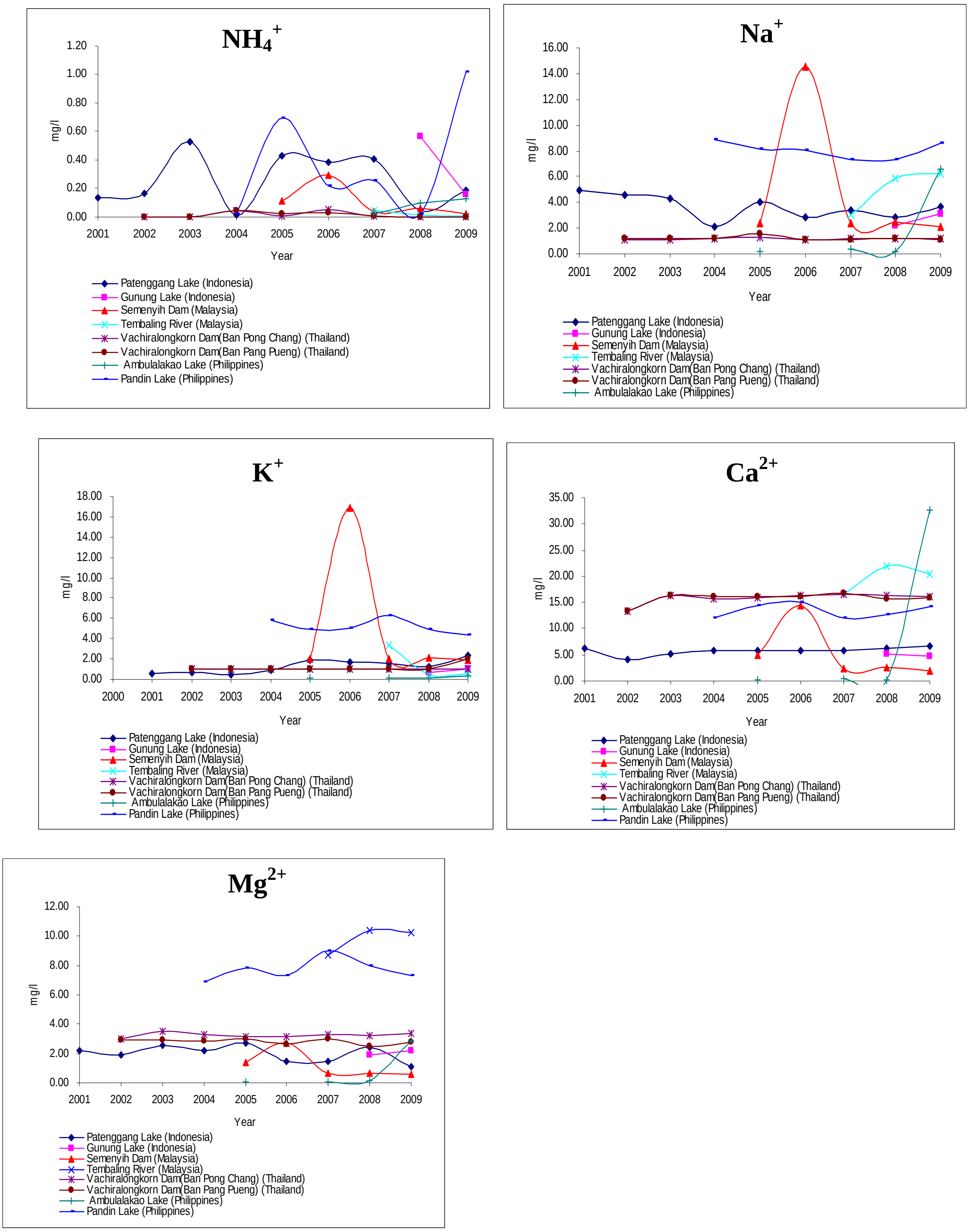

Figure 5.4.2 b). Changes in annual average values of NH_4^+, Na^+, K^+, $Ca^{2+}$and Mg^{2+} in inland water at monitoring sites in Indonesia, Malaysia, Philippines and Thailand.

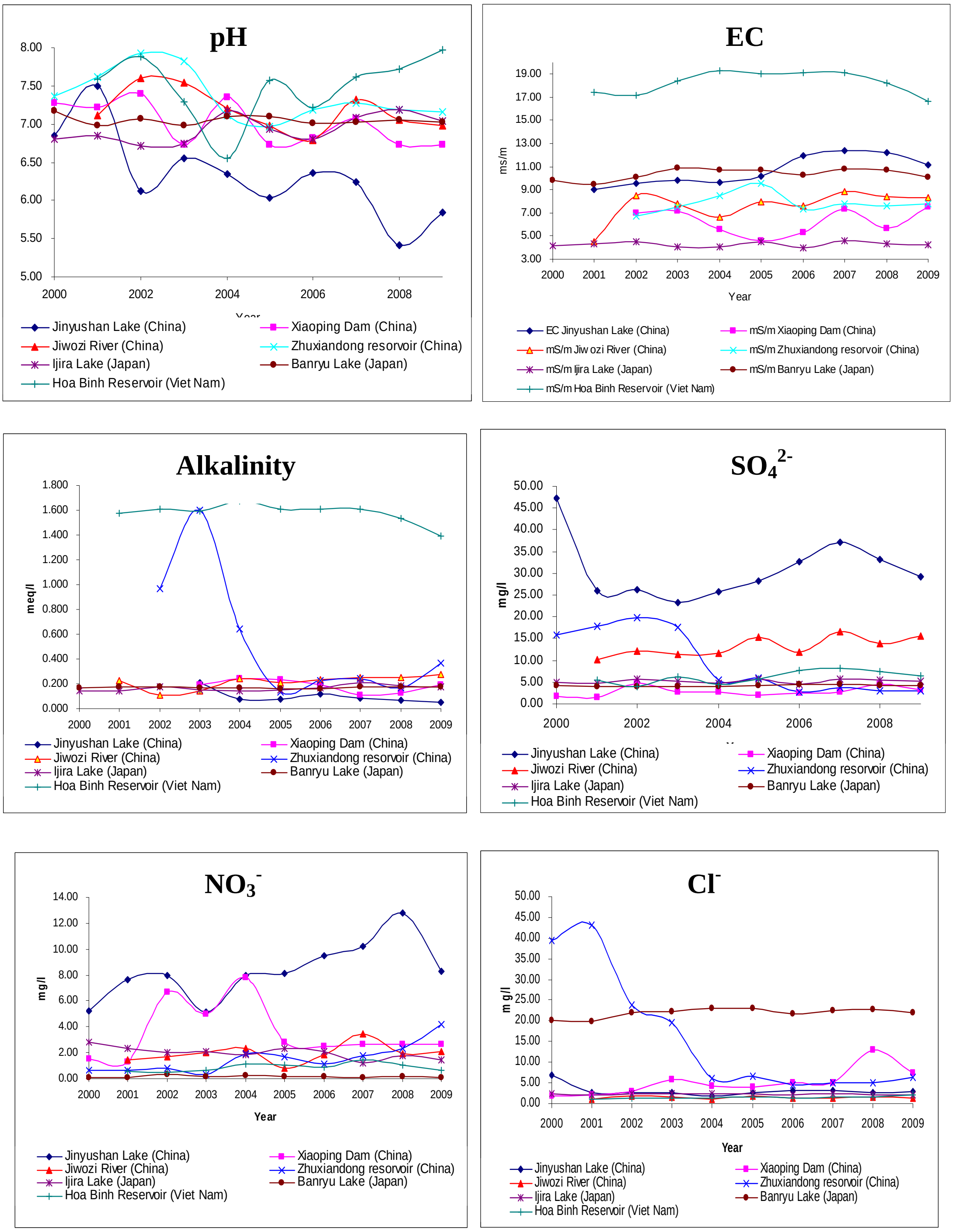

Figure 5.4.3 a). Changes in annual average values of pH, EC, Alkalinity, SO_4^{2-}, NO_3^- and Cl^-in inland water at monitoring sites in China, Japan and Vietnam.

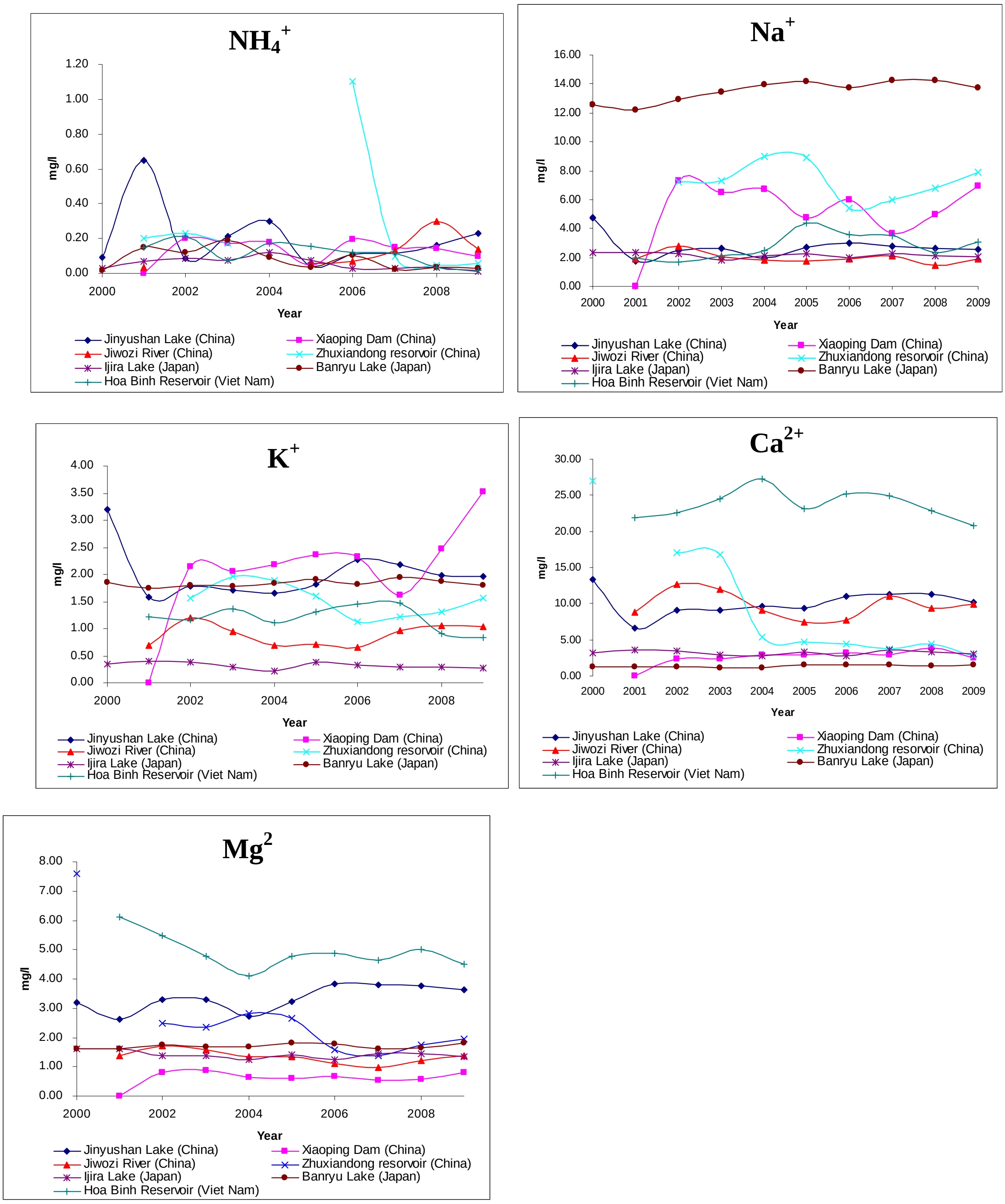

Figure 5.4.3 b). Changes in annual average values of NH_4^+, Na^+, K^+, Ca^{2+}andMg^{2+} in inland water at monitoring sites in China, Japan and Vietnam.

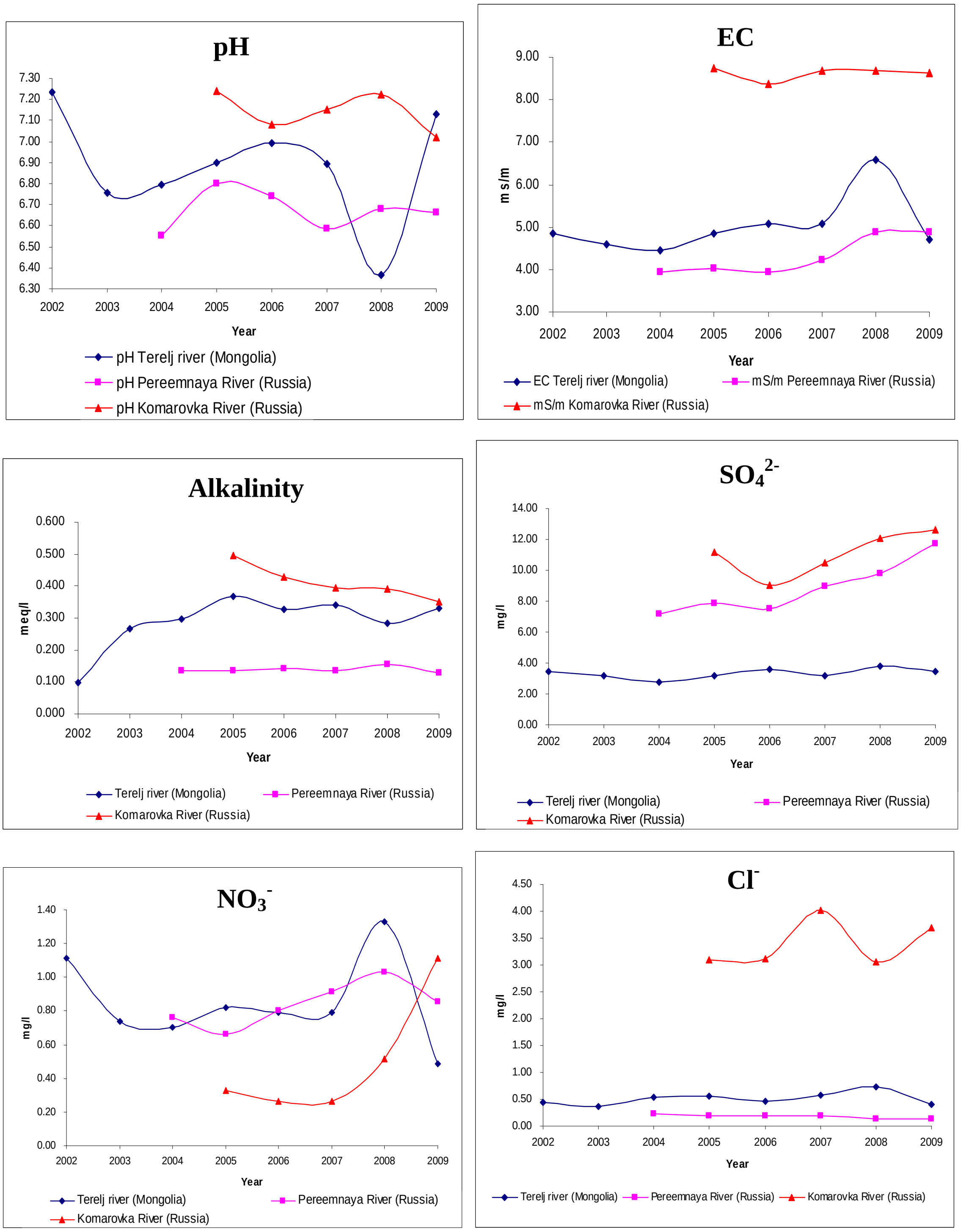

Figure 5.4.4 a). Changes in annual average values of pH, EC, Alkalinity, SO_4^{2-}, NO_3^- and Cl^- in inland water at monitoring sites in Mongolia and Russia.

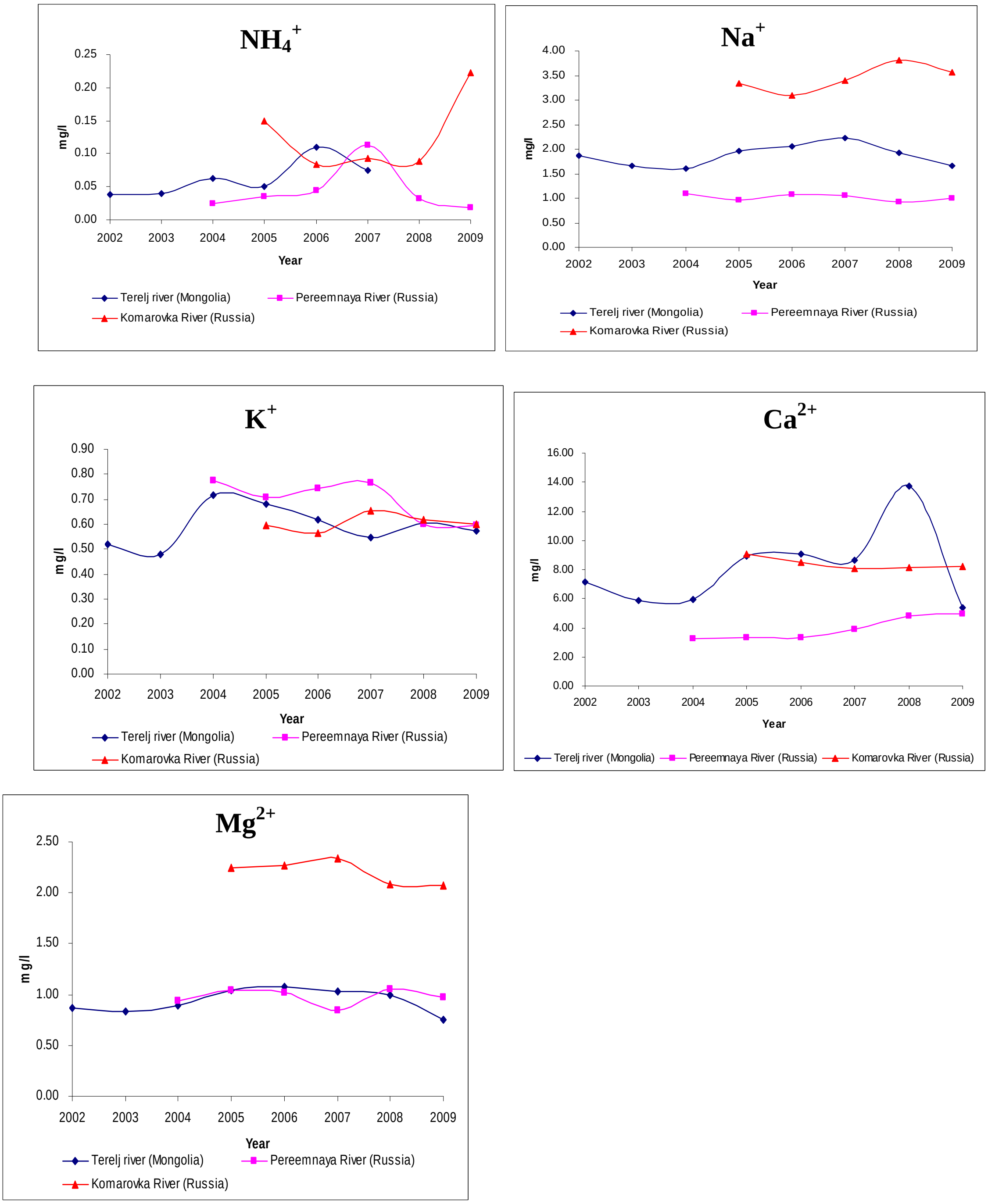

Figure 5.4.4 b). Changes in annual average values of NH_4^+, Na^+, K^+, Ca^{2+} and Mg^{2+} in inland water at monitoring sites in Mongolia and Russia.

5.5. Catchment-scale analysis in EANET countries

5.5.1. Case studies of the catchment-scale analysis in EANET countries

Integrated monitoring, including the atmosphere, vegetation, soil, and inland water, is required to evaluate effects of atmospheric deposition on forest ecosystems qualitatively and quantitatively. Catchment-scale analysis of biogeochemical processes may be one of the solutions for this requirement. Since the East Asian region is latitudinally wide from tropical to boreal zones, various types of ecosystems have been developed depending on the respective climatic zones. The Network Center for EANET (NC) has been promoting case studies of the catchment-scale analysis as research projects in cooperation with experts of the EANET countries, taking the climatic zones into account. The joint research projects are conducted in a tropical rainforest, tropical seasonal forest, and temperate coniferous forest, namely, Danum Valley site, Sabah State, Malaysia, Sakaerat site, Nakhon Ratchasima Province, Thailand, and Kajikawa site, Niigata Prefecture, Japan. In this section, major scientific outcomes from the projects were highlighted.

5.5.2. Study sites and field surveys

Characteristics of the study sites and outline of the field surveys were summarized in Table 5.5.1. Danum Valley and Sakaerat sites are similar in catchment area, soil type and geology, while vegetation is different depending on their climates. The catchment area of Kajikawa site was very small but the vegetation, soil type and geology are common in Japanese forest area.

Surveys on output were conducted mostly by the same methods to estimate ion flux by the stream. Water level was monitored continuously at 10-20 minutes intervals to estimate water discharge, while a weir was constructed in the case of the Danum Valley and Kajikawa sites. Stream water was collected basically biweekly or twice a month. The pH, electric conductivity (EC), alkalinity, anions, cations and SiO_2 as an indicator of weathering rate were measured for the stream water. Ion fluxes by the stream were estimated based on the water flux and the ion concentrations.

Table 5.5.1. Study sites and outline of the field surveys in the joint research projects on catchment analysis in Malaysia, Thailand and Japan.

	Danum Valley (since 2008)	Sakaerat (since 2005)	Kajikawa (since 2002)
Catchment area	Approx. 44 ha	Approx. 35 ha	3.84 ha
Vegetation	Tropical rainforest	Tropical seasonal forest (dry evergreen forest)	Temperate coniferous forest (Japanese cedar forest)
Soil type	Acrisols, Alisols	Orthic Acrisols	Dystric Cambisols
Geology	Mudstones, sandstone	Sandstone	Granodiorite
Surveys on output (ion flux by the stream)	● Water level (flux) ● Stream water chemistry	● Water level (flux) ● Stream water chemistry	● Water level (flux) ● Stream water chemistry
Surveys on Input (ion flux by atmospheric deposition)	● Rain gauge ● Resin sampling of Rainfall (RF)/ throughfall (TF)	● Rain gauge ● RF/TF/ stemflow (SF) ● Resin sampling of RF/TF	● Rain gauge ● RF/TF/SF
Other surveys	● Soil chemical properties ● Ion flux in soil ● Soil solution ● Soil moisture content	● Soil chemical properties ● Ion flux in soil ● Soil solution ● Soil moisture content ● Litter fall ● Particulate matters	● Soil chemical properties ● Soil solution ● Soil moisture content ● Particulate matters ● Leaf surface condition (wettablility, epicuticular wax, SEM observation, etc.)
Collaborators	Malaysian Meteorological Department (MMD), Asia Center for Air Pollution Research (ACAP), Universiti Putra Malaysia (UPM), and National Institute for Agro-Environmental Sciences (NIAES)	Royal Forest Department (RFD), Environment Research and Training Center (ERTC), ACAP, NIAES, King Mongkut's University of Technology Thonburi (KMUTT) and Meisei University	Niigata University, Niigata Prefectural Institute of Public Health and Environmental Sciences, ACAP
Funding	Global Environment Research Funds (GERF: C-082 and B-0801) of Ministry of the Environment of Japan (MOEJ)	GERF (C-052, C-082 and B-0801) of MOEJ, KAKENHI of Ministry of Education, Culture, Sports, Science and Technology (MEXT)	MOEJ, ACAP, GERF (C-052, C-082 and B-0801) of MOEJ

Note. EANET station is located in the same compound in case of Danum Valley site and Sakaerat site. Sample collection for the surveys on input and output was conducted basically biweekly or twice a month, while the resin sampling was done at several-month intervals.

Since electricity was not available in the forest area of these sites, bulk sampling was conducted for the surveys on input. In Sakaerat and Kajikawa sites, rainwater from rainfall outside forest canopy (RF), throughfall under forest canopy (TF), and stemflow (SF) were collected by bulk sampling at the same interval as the stream water collection. For the ions such as SO_4^{2-}, in which canopy uptake or leaching are negligible, deposition by TF+SF can be considered as total deposition (Lindberg & Lovett, 1992). The pH, EC, anions and cations were measured for the rainwater. Ion flux by atmospheric deposition was estimated based on precipitation amounts (from sample amounts or rain gauge) and the ion concentrations. However, in the case of Danum Valley site, the resin sampling method using ion-exchange resin was applied (Fenn & Poth, 2004), since the experience in Sakaerat site suggested that nitrogen compounds were quickly consumed by microbial activities during the sampling period in tropical climate. The ions trapped from rainwater during several months were extracted and analyzed in the case of the resin sampling.

Moreover, additional surveys on soil, soil solutions, ion flux in soil, etc. were conducted to discuss more detailed biogeochemical processes in the ecosystems.

5.5.3. Outcomes from Kajikawa site

The surveys in Kajikawa site started in 2002. The input-output budget for eight water years has been estimated from 2002 to 2010, as shown in Table 5.5.2. Two scientific papers based on the surveys were published in international journals (Sase *et al.*, 2008; Kamisako *et al.*, 2008). Major findings as presented from the said publications were introduced in the following sections.

1) Clear seasonality and the highest level of deposition

For most ions, deposition amounts by RF and TF+SF increased from late autumn to winter and then decreased in spring in Kajikawa site (Sase *et al.*, 2008; Kamisako *et al.*, 2008). The prevailing wind direction at the Niigata Meteorological Observatory was the West-Northwest (WNW) in the winter from December to March (Japan Meteorological Agency, 2003; 2004). Since the valley of Kajikawa site is open to the WNW, toward the sea and the continent, the site may experience a relatively direct effect of seasonal winds in winter. The transport of anthropogenic pollutants from the west through the sea is also a potential factor of the seasonal trends (Ohizumi *et al.*, 2001). As shown in Table 5.5.2, annual deposition amounts of S and N were significantly large, which were comparable or larger than those at the Lake Ijira catchment; total depositions (wet + dry) of SO_4^{2-} and NO_3^- were 0.57 kmol ha^{-1} yr^{-1} (18.3 kg S ha^{-1} yr^{-1}) and 1.37 kmol ha^{-1} yr^{-1} (19.2 kg N ha^{-1} yr^{-1}), respectively (Nakahara *et al.*, 2010). According to Hayashi & Yan (2010), the N deposition from RF at Kajikawa site was the highest level of wet N deposition in Japan.

Table 5.5.2. Input-output budget of sulphur (S) and nitrogen (N) for eight water years in Kajikawa site.

	Water year	S	N	Precipitation
		kg ha^{-1} y^{-1}		mm
Rainfall	2002-2003	20.2	15.1	2453
	2003-2004	23.9	20.1	2405
	2004-2005	20.5	15.1	2385
	2005-2006	23.7	20.3	2354
	2006-2007	22.2	20.4	2259
	2007-2008	21.6	27.9	2362
	2008-2009	19.3	15.3	2378
	2009-2010	17.2	14.2	2155
	Mean	21.1	18.5	2344
TF + SF	2002-2003	29.0	17.3	2040
	2003-2004	30.0	17.2	1771
	2004-2005	33.3	16.7	1865
	2005-2006	31.4	18.8	2077
	2006-2007	32.2	19.4	2044
	2007-2008	28.9	20.6	2071
	2008-2009	30.3	16.3	2255
	2009-2010	24.4	12.9	1825
	Mean	29.9	17.4	1994
Stream	2002-2003	26.4	10.0	1512
	2003-2004	20.6	7.1	1198
	2004-2005	28.1	10.2	1657
	2005-2006	23.6	8.5	1402
	2006-2007	24.0	9.1	1425
	2007-2008	23.1	10.8	1427
	2008-2009	18.1	7.2	1129
	2009-2010	18.5	7.6	1273
	Mean	22.8	8.8	1378

Note: Water year, June to the next May.

2) Canopy interactions of nitrogen compounds in Japanese cedar forest

As shown in Table 5.5.2, the S deposition from TF+SF was 40% higher than that from RF suggesting effects of dry deposition. However, annual N depositions from RF and TF+SF were not significantly different.

Moreover, deposition amounts of K^+, NO_3^- and NH_4^+ especially from TF+SF did not show clear seasonality (Sase *et al.*, 2008). Thus, it was suggested that depositions of those ions from TF+SF had large effects of canopy interactions. Contact angle (CA) of water droplets on leaves can be used as an indicator of leaf surface wettability. The K^+ concentration in TF at the Kajikawa site was negatively correlated with the CA of one-year-old leaves of Japanese cedar (*Cryptomeria japonica*), while the NH_4^+ concentration was positively correlated with the CA (Sase *et al.*, 2008). Moreover, the net fluxes of NH_4^+ and NO_3^- from TF were positively correlated with the CA as shown in Figure 5.5.1. It was suggested that the increase in wettability of the leaves may accelerate leaching of K^+ or uptake of NH_4^+ (and possibly NO_3^-). The effect of wettability on the foliar N uptake from wet deposition was confirmed by ^{15}N-labelled exposure test for European tree species, namely, *Fagus sylvatica*, *Quercus robur*, *Betula pendula* and *Pinus sylvestris* (Adriaenssens *et al.*, 2010). In their case, the uptake rate was also higher in NH_4^+ than in NO_3^-. Leaf surface properties, leaf wettability in particular, may be one of the regulatory factors for the leaching of K^+ and uptake and/or consumption of N compounds on the forest canopy. Small changes in leaf surface properties on the canopy may contribute to biogeochemical cycles in forest ecosystems. Moreover, it can be pointed out that precise estimate of dry deposition flux is very important to discuss a material budget in forest ecosystems, especially for N compounds.

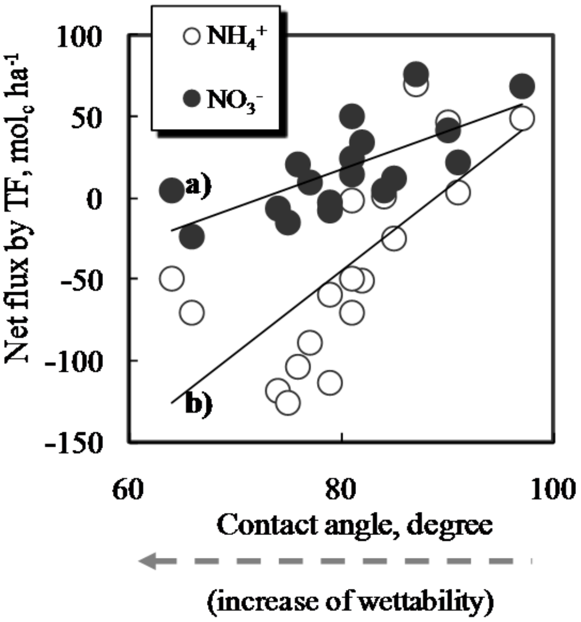

Figure 5.5.1. Relationship between leaf wettability and net flux by throughfall (TF).

Net flux, TF minus rainfall; Regression lines a) and b) represent significant correlations in NO_3^- and NH_4^+respectively (Speaman, R = 0.723, p=0.001 and R = 0.810, p=0.000). (Redrawn based on Sase *et al*., 2008).

3) Possible nitrogen saturation

As shown in Table 5.5.2, the N deposition at Kajikawa site was the highest level in Japan, which was larger than the threshold value in Europe at 10 kg N ha^{-1} yr^{-1}, for enhancing N discharge to stream water (Kamisako *et al*., 2008). In fact, mostly 50% of N from the atmospheric deposition may have flowed out from the ecosystem (see Table 5.5.2) and the NO_3^- concentration was relatively high. Nevertheless, the NO_3^- concentration in the stream water seemed to be still regulated by plant uptake, since the NO_3^- concentration was negatively correlated with air temperature and decreased in summer (Kamisako *et al*., 2008). In case of the Lake Ijira catchment, where N saturation was reported, the stream water NO_3^- concentration lost such seasonality (Nakahara *et al*., 2010), as introduced in Chapter 6. In this sense, the catchment of Kajikawa site has not actually been nitrogen-saturated yet. However, during high discharge periods in heavy rain events, the stream water pH at Kajikawa site clearly dropped with the increase of the NO_3^- concentration (Kamisako *et al*., 2008), which was similar to the phenomenon observed in the Lake Ijira catchment (Nakahara *et al*., 2010). This may be the first symptom proceeding to N saturation.

5.5.4. Outcomes from Danum Valley site and Sakaerat site

As shown in Figure 5.5.2, Danum Valley site does not have clear seasonality, the monthly mean temperature shows mostly flat and the monthly precipitation kept at high level, higher than 150 mm even at the lowest. Therefore, the site was in perhumid condition through the year. On the other hand, Sakaerat site shows clear seasonality, having a dry season from November to February and a perhumid wet season from April to October, while the precipitation decreased temporarily in July/August. The clear seasonality at Sakaerat site may affect biogeochemical processes in the ecosystem as discussed below.

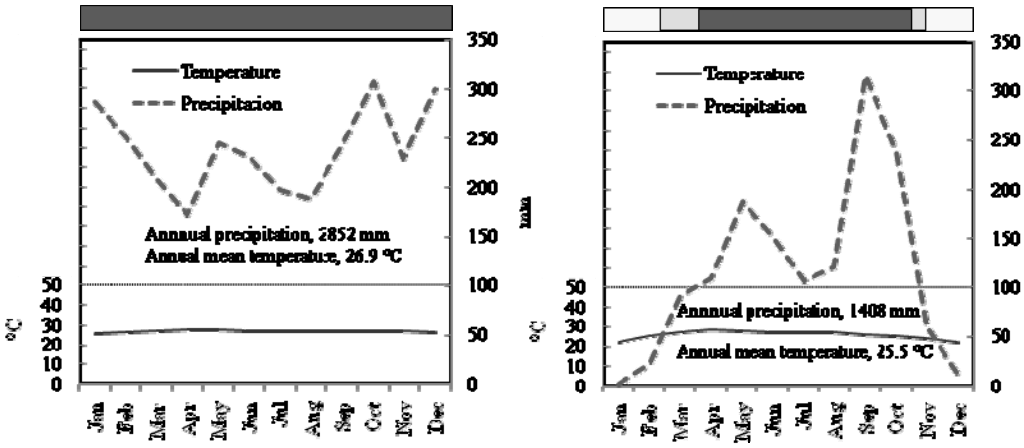

Figure 5.5.2. Climate plots of Danum Valley site (left) and Sakaerat site (right).
Drawn based on the monthly precipitation and monthly mean temperature from 1992 to 2006 in Danum Valley Field Center (DVFC) and from 2001 to 2008 in Sakaerat Silvicultural Research Station). Dark color, light color, and white color in the bars on the top indicate perhumid period of wet season, other period of wet season, and dry season, respectively.

1) Sensitivity of the stream water to acid deposition

Table 5.5.3 shows stream water chemistry of the Danum Valley and Sakaerat sites. All the parameters expect Cl^- were lower at Sakaerat site than the Danum Valley site as shown in Table 5.5.3. In particular, alkalinity at Sakaerat site was nearly 10% of that at the Danum Valley site. The stream water at Sakaerat site may have lower acid neutralizing capacity (ANC) than that at Danum Valley site. Moreover, the SiO_2 concentration, an indicator of weathering rate, was also half of that at Danum Valley site. High weathering rate at Danum Valley site may be related to the higher ANC of the stream water.

Table 5.5.3. Stream water chemistry of Danum Valley site and Sakaerat site.

Catchment	pH	EC	SO_4^{2-}	NO_3^-	Cl^-	Ca^{2+}	Mg^{2+}	alkalinity	SiO_2
		$mS\ m^{-1}$	$\mu mol_c\ L^{-1}$						$mg\ L^{-1}$
Danum Valley (n=55)	7.1	4.7	38.0	9.8	36.4	168	148	373	5.8
Sakaerat (n=96)	5.7	2.1	7.8	4.4	99.1	35.8	58.7	41.3	2.6

Note. Volume weighted mean.

2) Material flux

Vertical flux of major anions and cations were shown in Table 5.5.4. Deposition amounts of SO_4^{2-} and NO_3^- from TF were larger in Sakaerat site than in Danum Valley site, while the depositions of NH_4^+ were not different. Regional surveys on wet deposition by resin sampling also showed that wet depositions of S and N were larger in Nakhon Ratchasima Province around Sakaerat site than in Tawau Division around Danum Valley site; Nakhon Ratchasima Province, N: 0.68 $kmol_c\ ha^{-1}\ yr^{-1}$; S: 0.18 $kmol_c\ ha^{-1}\ yr^{-1}$, Tawau Division, N: 0.22 $kmol_c\ ha^{-1}\ yr^{-1}$; S: 0.13 $kmol_c\ ha^{-1}\ yr^{-1}$. It seemed that atmosphere was relatively polluted in the area around Sakaerat site compared with the area around Danum Valley.

Table 5.5.4. Vertical flux of materials in Danum Valley site and Sakaerat site.

		SO_4^{2-}	NO_3^-	NH_4^+	Na^+	K^+	Ca^{2+}	Mg^{2+}
		$kmol_c\ ha^{1}\ yr^{1}$						
Danum valley	Rainfall	0.12	0.03	0.09	-	-	0.03	0.06
	Throughfall	0.17	0.04	0.41	-	-	0.38	0.27
	Soil 0cm	0.09	6.28	0.45	1.69	14.1	15.4	7.8
	20cm	0.16	8.63	1.21	1.17	8.5	6.4	5.2
	70cm	0.37	6.56	1.12	0.96	6.8	9.6	5.4
	Stream	0.46	0.13	0.02	1.41	0.18	2.11	1.84
Sakaerat	Rainfall	0.20	0.14	0.27	-	-	0.11	0.04
	Throughfall	0.53	0.30	0.43	-	-	0.40	0.26
	Soil 0cm	4.26	1.54	3.4	0.9	11.2	7.2	6.3
	20cm	3.47	0.65	1.7	0.4	1.9	1.8	1.9
	60cm	2.43	1.70	1.4	0.6	1.8	1.9	4.1
	Stream	0.02	0.01	0.01	0.15	0.04	0.10	0.15

Note: Fluxes from rainfall, throughfall, and soil layers were measured by the resin sampling method. Since trapped ions by ion-exchange resin were extracted by KCl, Cl^- was not determined.

Vertical fluxes in soil layer and fluxes by stream water for most ions were generally larger in Danum Valley site than in Sakaerat site, while the fluxes of SO_4^{2-} was larger in Sakaerat site. The fluxes of NH_4^+ were slightly larger in Sakaerat but the total inorganic N fluxes (NO_3^- + NH_4^+) were clearly larger in Danum Valley. Moreover, the vertical fluxes in soil layer were significantly larger than those by throughfall. The internal material cycle in soil-plant system may be quite large compared

169

with the deposition amounts. The internal cycle may also be larger in Danum Valley site than in Sakaerat site. It was suggested that the catchment of Danum Valley site had larger acid buffering capacity than that of Sakaerat site. Annual flux of $SiO_2$from the stream at Danum Valley site was significantly larger than that at Sakaerat site; 86.9 kg ha^{-1} yr^{-1} and 7.8 kg ha^{-1} yr^{-1} respectively. This also indicates high buffering capacity of Danum Valley site. Actually, the stream water pH at Danum Valley site did not easily decrease even in high-discharge periods by heavy rains (detailed data was not shown here).

3) Response of stream water to atmospheric deposition at Sakaerat site

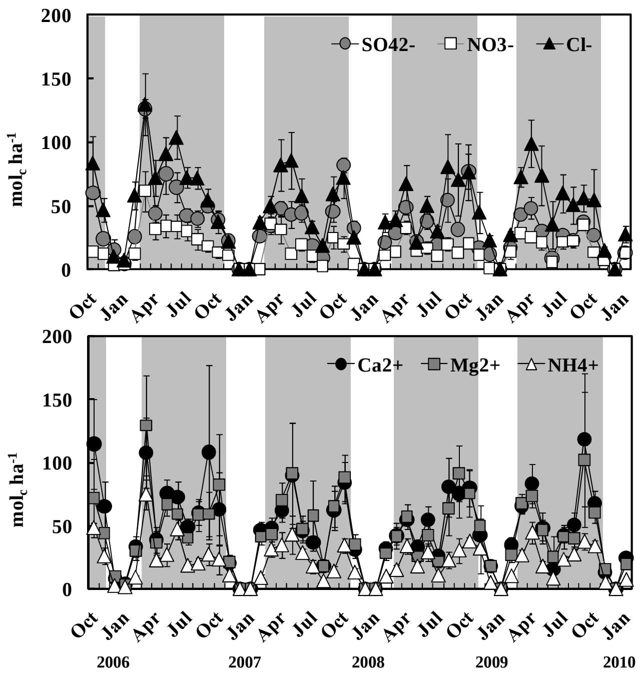

Figure 5.5.3. Changes in monthly depositions by throughfall and stemflow at Sakaerat site. *Shaded part indicates wet season. Error bar shows standard error of five replicates.*

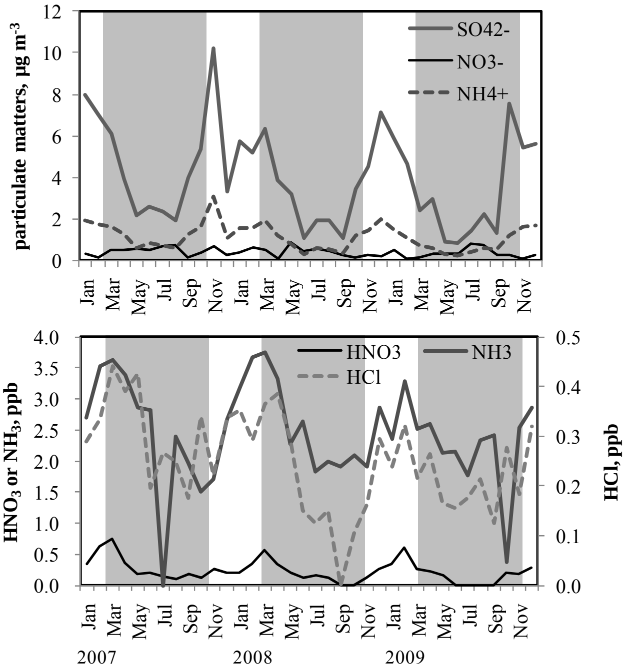

Figure 5.5.4. Changes in monthly mean air concentrations of gaseous and particulate matters at Nakhon Ratchasima EANET station. *Shaded part indicates wet season. (Drawn based on the EANET data by the filter pack method)*

Seasonal changes in depositions by TF+SF at Sakaerat site were shown in Figure 5.5.3. The deposition of all ions significantly increased in the beginning of wet season and the second peaks were observed in late wet season, reflecting the precipitation pattern (Figure 5.5.2). Seasonal changes in air concentrations at Nakhon Ratchasima EANET station, which is located in the same compound, showed the opposite pattern to those of the deposition as shown in Figure 5.5.4. Scavenging process of the atmosphere or accumulated dry deposition during dry season may contribute to the high deposition amounts in the beginning of the wet season.

Seasonal changes in stream water chemistry at Sakaerat site were shown in Figure 5.5.5. In the beginning of wet season, the pH and EC increased with alkalinity and base cations (alkalinization). Then, the pH dropped with the peak of SO_4^{2-} in middle or late wet season (acidification).

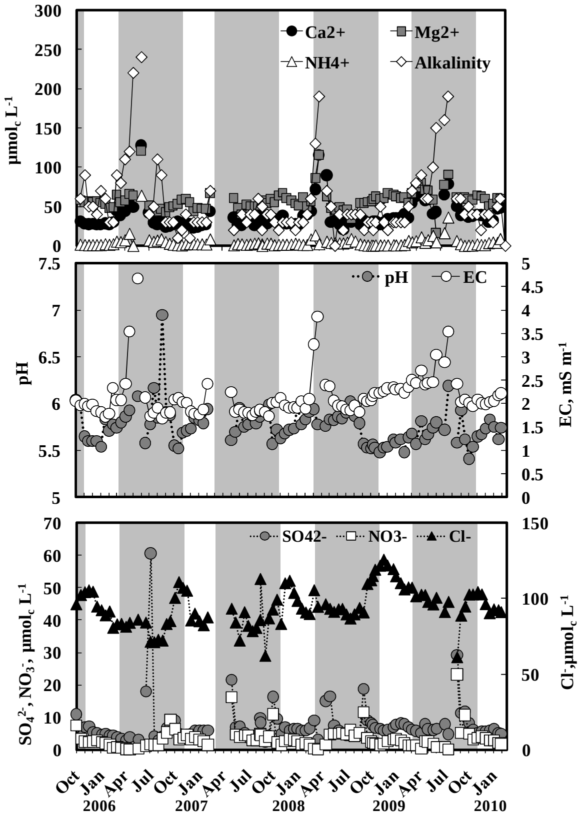

Figure 5.5.5. Changes in stream water chemistry at Sakaerat site. *Shaded part indicates wet season.*

The high depositions in the beginning of wet deposition may contribute to both alkalinization and acidification. However, the high depositions cannot explain enough the mechanisms.

As for the alkalinization, Yamashita *et al.* (2010) found that vertical fluxes of NO_3^- and NH_4^+ in shallow soil layers at Sakaerat site were significantly larger in early wet season than in other seasons, suggesting high mineralization rate of organic matters in early wet season. The first precipitations in the beginning of wet season probably enhanced microbial activities after severe dry season. The respiration of microorganisms may also contribute to increase in alkalinity (Van Breemen *et al.*, 1983). It was suggested that base cations and NH_4^+ derived from mineralization of organic matters and alkalinity from high microbial activities were major causes of the alkalinization of the stream water in the beginning of wet season. As discussed above, vertical fluxes by atmospheric deposition were limited compared with those in soil layers. Therefore, the mineralization may contribute to the alkalinization of the stream water more than the atmospheric deposition, although the precipitation pattern was the main factor for the mechanism.

As for the acidification, time lag of SO_4^{2-} leaching may be the key mechanism, even if leached SO_4^{2-} was derived from the high deposition in the beginning of wet season. Retention of SO_4^{2-} in soil can be described in the following formula (Van Breemen *et al.*, 1983):

$$\text{Exchangeable-}SO_4^{2-} + 2H_2O = \text{Exchangeable }(OH)_2 + SO_4^{2-} + 2H^+$$

With increase in SO_4^{2-} and H^+ in soil solution (declining of pH), more SO_4^{2-} can be retained in soil (chemical process: right to left). With decrease in SO_4^{2-} and H^+ (increasing of pH), SO_4^{2-} can be released from soil (left to right). The SO_4^{2-} concentration in soil solution must be relatively higher in the beginning of wet season because of the high deposition. Moreover, soil pH at Sakaerat site showed clear seasonality, declining from dry season to early wet season, and increasing from middle wet season to late wet season (Yamashita *et al.*, 2010). Therefore, such soil condition in the beginning of wet season may be able to retain SO_4^{2-} derived from the high deposition. The continuous precipitations during early wet season may dilute SO_4^{2-} concentration in soil solution gradually and soil pH also became increased in middle wet season. The soil may be able to release SO_4^{2-} in middle or late wet seasons with changes in its condition. In fact, we confirmed that SO_4^{2-} adsorption capacity of soils from Sakaerat site depended on SO_4^{2-} concentration and pH (detailed data is not shown here). The mechanism above can explain the destiny of SO_4^{2-} derived from the high deposition in the beginning of wet season.

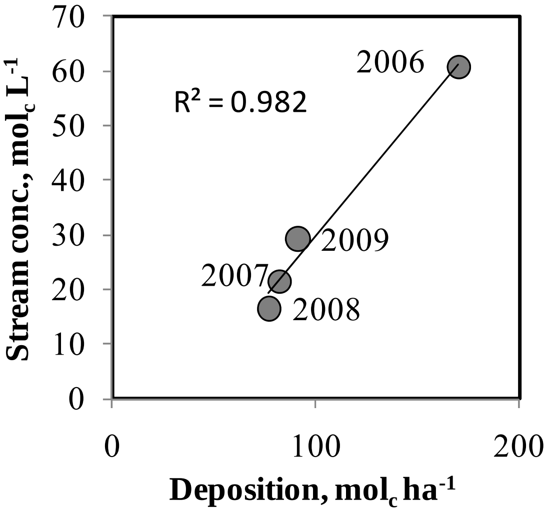

Figure 5.5.6. Relationship between the SO_4^{2-} deposition of the initial two months of wet season and the peak SO42- concentration in each year. *The number shows year.*

Figure 5.5.6 showed relationship between the SO_4^{2-} depositions by TF+SF during the initial two months (March and April) of wet season and the peak SO_4^{2-} concentration in each year from 2006 to 2008. The peak concentration responded clearly to the deposition amount. The relationship also supported the hypothesis above, in which SO_4^{2-} derived from the high deposition in the beginning of wet season must be leached to the stream as the peak. It was suggested that the acid-sensitive catchment of Sakaerat site reacted sensitively to the atmospheric deposition.

5.5.5. Perspective

Based on the experience through the research projects above, the Guideline for Catchment-scale Monitoring in East Asia was developed by the Task Force on Soil and Vegetation Monitoring of EANET. The Guideline was endorsed by Scientific Advisory Committee at its Tenth Session (SAC10) in October 2010. It is expected that catchment-scale monitoring will be conducted on a regular basis using the Guideline in the EANET countries. This is the great outcome of the project, which can contribute to the future EANET monitoring and evaluation of ecosystem impacts in the region.

5.6.Conclusions

Definite progress is being made in the monitoring of impacts of acid deposition in ecosystems in the East Asian region. In ecological monitoring sites where more than a single observation have been made already, temporal analyses in some cases have already revealed certain trends in the changes in the soil, forest and inland water monitoring parameters. The data trend generated over the years of monitoring in the ecological stations across the EANET participating countries suggests possible acidification or nitrogen saturation in several sites. There is, however, a need to isolate other existing environmental conditions that may have contributed to such an observation.

An increase in the nitrogen flux in the soil in several ecological monitoring sites has been observed. This report has advanced a number of reasons for the occurrence of such mostly related to forest growth and/or degradation, litter production and decomposition in stands and how these processes contribute to the nitrogen dynamics in the soil and inland waters. However, as pointed out earlier in this report, the amount and kind of data generated thus far in the ecological monitoring stations are not yet sufficient to attribute fully the nitrogen flux to atmospheric deposition.

There were also evidences of decline in certain forest monitoring stations in Japan, China, and Mongolia. In the first two countries such decline symptoms were not directly and solely attributed to air pollution effects. However, in a recent study of Sase *et al*., (2005), in Mongolia, the possible direct effects of pollutants in tree decline has been well documented. There were no evidences of decline in other East Asian countries. The decrease in biometric figures of trees in certain monitoring plots in the region were attributed to natural phenomena like typhoons, pest infestations, the occurence of infectious tree diseases, and even anthropogenic factors.

The pH of water in five inland aquatic systems significantly decreased from 2000 to 2009. The accumulated data for the last years suggested the possible acidification or nitrogen saturation of the inland ecosystems in several EANET sites. But just like in the cases of the forest soil and vegetation, some other factors that can bring about nitrogen saturation in inland waters need further amplification.

The availability of data covering more than a single measurement over a period of time has led to the establishment of trends in certain parameters that are being investigated in the EANET monitoring framework. With the kind of data that is already available at the end of 2009, attempts have been made to identify the possible relationships that exist among the different phenomena that describe the dynamics of nitrogen and sulphur loading in both the terrestrial and inland aquatic environments. As the monitoring progresses through the coming years, there is a growing need to carefully discuss the relationship among components of the ecosystems, including soil, vegetation, inland water, and atmosphere.

The promotion of catchment scale analysis is an approach to clarify such relationship. This analysis takes the form of an integrated monitoring including atmosphere, vegetation, soil, and inland water to evaluate the effects of atmospheric deposition on forest ecosystems qualitatively and quantitatively. The EANET has been implementing a number of case studies in the East Asian region covering three sites representing three different forest types. The experiences in such studies have led to the formulation of the Guideline for Catchment-scale Monitoring in East Asia by the Task Force on Soil and Vegetation Monitoring of EANET. The Guideline was endorsed by Scientific

Advisory Committee at its Tenth Session (SAC10) in October 2010. With this development the monitoring initiatives of the EANET takes on a new dimension to better explain the phenomenon of acid deposition and its concomitant effects in the East Asian Region.

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Annex

Appendix Table 5.1.Total average values of major parameters in inland water at the EANET monitoring sites.

Site name	pH	EC	Alkalinity	SO_4^{2-}	NO_3^-	Cl^-	NH_4^+	Na^+	K^+	Ca^{2+}	Mg^{2+}
		$mS\ m^{-1}$	$meq\ L^{-1}$	$mg\ L^{-1}$							
Jinyunshan Lake	6.42	10.6	0.10	29.1	8.64	2.67	0.21	2.51	1.88	9.73	3.35
Xiaoping Dam	7.08	6.27	0.18	2.92	3.89	5.54	0.15	5.92	2.36	2.86	0.69
Jiwozi River	7.25	7.59	0.22	13.2	1.96	1.40	0.14	1.94	0.87	9.69	1.33
Zhuxiandong Stream	7.17	8.08	0.30	3.90	2.18	5.59	0.33	7.35	1.46	4.20	2.04
Pattengang Lake	7.56	6.86	0.45	5.98	0.24	4.59	0.29	4.24	1.25	5.92	2.08
Gunung Lake	6.95	5.70	0.45	4.42	0.05	1.62	0.44	2.74	0.88	4.83	2.05
Ijira Lake	7.10	4.28	0.16	5.22	2.00	2.28	0.06	2.18	0.32	3.18	1.41
Banryu Lake	7.06	10.3	0.17	4.18	0.13	21.9	0.09	13.5	1.84	1.32	1.70
Semenih Dam	7.16	2.56	0.30	2.23	0.67	2.57	0.12	4.78	3.95	6.10	1.37
Tembaling River	7.44	17.6	1.82	8.17	0.71	1.58	0.02	5.03	1.43	19.7	9.77
Terelj River	7.04	5.03	0.29	3.34	0.87	0.51	0.07	1.87	0.60	8.17	0.94
Pandin Lake	7.98	18.0	1.74	0.63	2.28	3.35	0.63	8.09	5.23	13.4	7.72
Ambulalakao Lake	5.66	0.43	0.08	0.32	1.40	0.18	0.09	0.28	0.10	0.30	0.11
Pereemanaya River	6.71	4.32	0.14	8.82	0.84	0.18	0.04	1.02	0.70	3.92	0.98
Komarovka River	7.16	8.62	0.41	11.1	0.88	3.40	0.13	3.44	0.61	8.39	2.20
Vachiralongkorn Dam_1	7.67	11.4	1.05	2.83	0.11	1.36	0.03	1.28	1.13	16.5	3.39
Vachiralongkorn Dam_2	7.65	11.0	0.98	2.33	0.44	1.38	0.04	1.32	1.25	16.3	2.95
Hoa Binh Reservoir	7.60	18.1	1.58	6.16	0.82	1.51	0.13	2.71	1.18	23.5	5.09

Chapter 6. Other Related Studies for EANET Region*

6.1 Introduction

Due to continuing rapid economic growth after the year of 2000, Asia keeps attracting much more concern than before from the view of global, hemispherical and regional air pollution. In addition to the effort of EANET, there have been many research activities and organizational initiatives focusing regional air pollution in East Asia as well as in global and hemispherical perspectives. In this chapter, such research outputs and initiatives in the last few years are reviewed in order to provide useful information on present status of acid deposition and regional air pollution to supplement EANET activities. The covered topics are atmospheric observation, emission inventory, modeling and impact studies, which are not necessarily directly related to EANET, but would be useful for understanding acid deposition and air pollution in East Asia. This chapter does not intend to provide comprehensive review, but the observational studies focusing on regional representativeness, modeling studies utilizing EANET data for validation and analysis, and ecological studies conducted by collaboration with EANET countries are introduced with higher priority. In principle, research outputs published in peer-reviewed international journals are targeted here. As for emission inventories and organizational initiatives, several documents that are not necessarily published in regular journals are also included. In addition to acidic species, EANET data of surface ozone have been widely used in research papers in the last few years reflecting the growing concern on this species from health and ecosystem impacts and hemispherical air pollution, and therefore the topics on ozone are covered in this chapter. Although EANET data of PM_{10} mass concentrations have also been accumulated, they are not much used in research papers yet, and are not much covered here.

6.2 Observational studies for EANET region

6.2.1 Field campaigns for acid deposition/ regional air quality

One of the objectives of acid deposition monitoring in EANET is to grasp the effect of acidification and the long-term trends of causative agents and the other relevant air pollutants. The accumulated long-term monitoring data are useful to evaluation of the impact on ecosystem and human health. However, in order to elucidate impacts of specific atmospheric events and chemical and physical processes relevant to assessing the effects of air pollutants, the intensive observation studies will be helpful in addition to the long-term monitoring. In this section, field campaigns for acid deposition and regional air quality in East Asia are reviewed.

It should be noted that some external scientific data and conclusion in this chapter does not reflect EANET official view.

6.2.1.1 Intensive observation study in Central Eastern China

Central Eastern China (CEC), located at 30–40°N and 111–120°E, has received considerable attention as a region with high concentrations of trace gases and aerosol pollutants, which are causative agents of serious air pollution and important components contributing to global radiation budgets. An intensive field observation campaign at the summit of Mount Tai, named as the Mount Tai Experiment 2006 (MTX2006), was carried out in June 2006 to elucidate the emission sources, tropospheric chemistry, transformation and transport processes of O_3, aerosols, and their precursors over CEC. Mount Tai is free from local sources but is located at the center of a regionally polluted area of CEC. The scientific outcomes of MTX 2006 were published in the special issue of Atmospheric Chemistry and Physics (Irie *et al*., 2008, Li *et al*., 2008, Kanaya *et al*., 2008, Wang *et al*., 2009, Suthawaree *et al*., 2010, Inomata *et al*., 2010, Yamaji *et al*., 2010).

The impact of open crop residual burning on O_3, CO, Black Carbons (BC), and Organic Carbons (OC) concentrations over CEC during the MTX2006 was evaluated using the regional chemical transport model, the Models-3 Community Multiscale Air Quality Modeling System (CMAQ) (Yamaji *et al.*, 2010). Figure 6.2.1 shows hourly O_3, CO, NOx, and BC concentrations observed during MTX2006. The period of MTX2006 was roughly divided into two parts: 1) Polluted days with heavy open crop residual burning in the first half of June; 2) Cleaner days with negligible field burning in the latter half of June. Additionally, the first half of June was defined by two high pollution episodes during June 5–7 and 12–13, and a relatively cleaner episode during June 8–10 between these two high pollution episodes. In the first polluted episode, the CMAQ model identified high O_3, CO, BC, and OC concentrations at the summit of Mount Tai which were affected by open crop 20 residual burning in the South of CEC and northward transport. For this episode, the impacts from open crop residual burning were 12% for O_3, 35% for CO, 56% for BC, and 80% for OC over CEC. The daily emissions from open crop residual burning were an essential factor to evaluate the atmospheric concentrations of pollutants during the MTX2006. These emissions have a large impact not only on primary pollutants but also on secondly pollutions, such as O_3, in the first half of June over the Northeastern Asia.

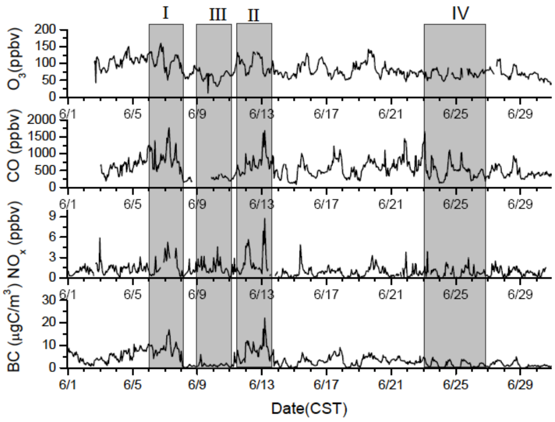


Figure 6.2.1. Hourly O_3, CO, NOx, and BC concentrations at Mt. Tai in June 2006.

A 3-D regional chemical transport model, the Nested Air Quality Prediction Model System (NAQPMS), reproduced the temporal and spatial variations of near-ground ozone and other pollutants during MTX2006 (Li *et al*., 2008), and it captured highly polluted and clean cases well. The simulated near-ground ozone level over CEC was 60–85 ppbv, which was higher than values in Japan and over the North Pacific (20–50 ppbv). Due to high anthropogenic and biomass burning emissions that occurred in the Southern part of the CEC, the contribution to ground ozone levels produced in this area played the most important role (32.4 ppbv, 37.9% of total ozone) in the monthly mean ozone concentration at Mt. Tai; values reached 59 ppbv (62%) on 6–7 June 2006. The monthly mean horizontal distribution of ozone indicated that photochemical reactions controlled the spatial distribution of O_3 over CEC. The regional-scale transport of pollutants also played an important role in the spatial and temporal distribution of ozone over CEC. Chemically produced ozone from the Southern part of the study region can be transported northeastwardly to the Northern rim of CEC; the mean contribution was 5–10 ppbv, and it reached 25 ppbv during high ozone events. Studies of the outflow of CEC ozone and its precursors, as well as their influences and contributions to the ozone level over adjacent regions/countries, revealed that the contribution of CEC ozone to mean ozone mixing ratios over the Korean Peninsula and Japan was 5–15 ppbv, of which about half was due to the direct transport of ozone from CEC and half was produced locally by ozone precursors transported from CEC.

6.2.1.2 Regional integrated experiments on air quality over Pearl River Delta

The aims of the Program of Regional Integrated Experiments on Air Quality over Pearl River Delta of China were to characterize in depth the pollution, and to improve the understanding of the chemical and radiative processes in the atmosphere of Pearl River Delta (PRD) in the South-Eastern China. This comprehensive program integrated ground-base in situ measurements, vertical observations (including aircraft) and model simulations. The intensive field campaign (PRIDE-PRD2004) was firstly conducted from 4 October to 5 November 2004 at two super-sites: an urban site in Guangzhou city (23.13°N, 113.26°E) and a non-urban site at Xinken (22.61°N, 113.59°E). They were coordinated with concurrent meteorological observations as well as measurements at stations of a regional monitoring network. Quality control and quality assurances, application of closure studies and observation-based modeling were the key elements of the research strategies for the PRIDE-PRD2004 campaign. Knowledge and understanding of the important issues, including 3-D distribution of air pollution, ozone formation process and its control strategies, sources of volatile organic compounds, chemical and physical characteristics and radiative properties of aerosols, and the important role of nitrous acid atmospheric chemistry, have been improved substantially by this study. The scientific outcomes of PRIDE-PRD2004 were published in the special issue of Atmospheric Environment (Zhang *et al*., 2008a, Wang *et al*., 2008a, Zhang *et al*., 2008b, Su *et al*., 2008, Wang *et al*., 2008bc, Liu *et al*., 2008bcd, Gnauk *et al*., 2008, Hu *et al*., 2008). Figure 6.2.2 shows time series of O_3, NOy, CO, $PM_{2.5}$ and SO_2 concentrations observed during MTX2004. A long continuous pollution episode was observed from October 6,

2004. Primary pollutants SO_2 and CO concentrations were quite high even though they met national air quality standard in general. At the same time, O_3 concentration was frequently higher than the China air quality standard of 93 ppb (200 µg.m^{-3}), and $PM_{2.5}$ mass concentration was much higher than 65 µg.m^{-3}. Formation of inversion layers in PRD and weak prevailing winds caused severe regional pollution of primary and secondary pollutants. Transport of pollutants from the outside of PRD was insignificant during the campaign.

After PRIDE-PRD2004, the further intensive observation study was carried out at a rural site approximately 60 km Northwest of the mega-city Guangzhou in the Southeast China during 1–30 July 2006. The scientific outcomes of PRIDE-PRD2006 were published in the special issue of Atmospheric Chemistry and Physics (Hua *et al.*, 2008, 2010, Verma *et al.*, 2010, Yue *et al.*, 2010, Lou *et al.*, 2010, Shao *et al.*, 2011).

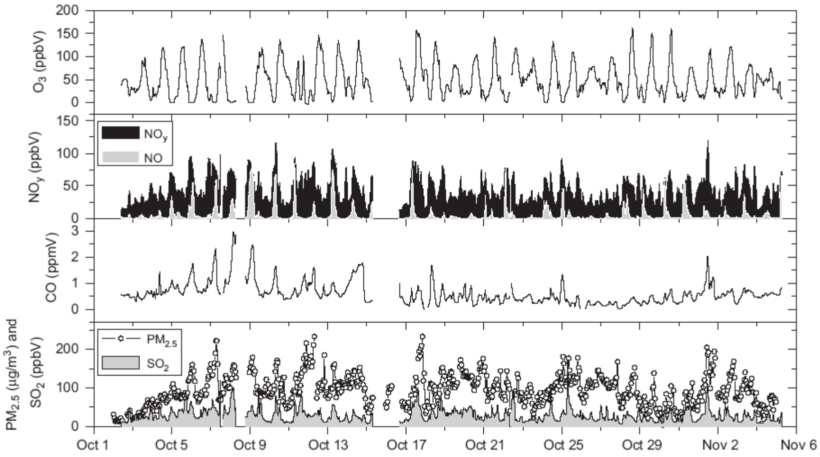


Figure 6.2.2. Time series of observed O_3, NOy, CO, SO_2, and $PM_{2.5}$ at Xinken, China.

6.2.1.3 Asian brown cloud project

United Nations Environment Programme (UNEP) report on Atmospheric Brown Cloud (ABC) and its multifarious impacts on health, agriculture and climate on both regional and global scales, has led to considerable concern. The brown haze has been observed during the period January to March over the South Asian region, and the tropical Indian Ocean, Arabian Sea and Bay of Bengal. It occurs only in the season following the Southwest monsoon and Northeast monsoon seasons. UNEP reported that the impact of the haze assessed with the help of an atmospheric general circulation model is a decrease in rainfall in the Northwestern Asia (including Saudi Arabia, Pakistan, Afghanistan).

East Asian Regional Experiment 2005 (EAREX 2005) was carried out in March–April 2005 as one of the first phase activities under the ABC project (Nakajima *et al.*, 2007). Some outstanding features of obtained aerosol characteristics and its direct radiative forcing in the East Asian region was compared with the results obtained in another ABC early phase regional experiment, ABC

Maldives Monsoon Experiment (AMMEX) conducted in the South Asian region. Time series of aerosol optical thickness (AOT), single scattering albedo (SSA), aerosol extinction cross section profile and CO concentration shows that air pollutants and mineral dust were transported every 5 to 7 days in the EAREX region to produce single scattering albedo (SSA) values at wavelength of 700 nm from 0.86 to 0.96 and large clear-sky shortwave forcing efficiency at 500 nm from 60 W m^{-2} to 90 W m^{-2}, though there are some unexplained inconsistencies depending on the evaluation method. The simulated whole-sky total forcing in the EAREX region is −1 to −2 W m^{-2} at TOA and −2 to −10 W m^{-2} at surface in March 2005 which is smaller in magnitude than in the AMMEX region, mainly because of large cloud fraction in this region (0.70 at Gosan versus 0.51 at Hanimadhoo in the total cloud fraction).

Some intensive field programs were performed to measure atmospheric aerosols at Fukue Island (32.752°N, 128.682°E) in Nagasaki prefecture, and at Cape Hedo (26.867°N, 128.249°E) in Okinawa prefecture, located in the West and Southwestern Japan. Aerosol chemical composition and gaseous species were measured at Cape Hedo, Okinawa, Japan (CHO), during the ABC/EAREX project period in March 2005 (Takami *et al.*, 2007), using high-time-resolution instruments including an Aerodyne quadrupole aerosol mass spectrometer (Q-AMS), a tapered element oscillating microbalance (TEOM), and gas monitors in order to investigate the transport and subsequent chemical transformation of aerosol in the East Asian region. Sulphate was the dominant species in fine aerosol mode and the average concentration of ammonium, sulphate and organics was 1.25, 6.37 and 2.16 μg.m^{-3}, respectively (Figure 6.2.3). The sulphate concentration observed at CHO in 2005 was about 1.5–2 times higher than that in 1994. For the majority of high-sulphate observations, the air mass was transported from the central East Chinese region (between Shanghai and the Shandong Peninsula). Sulphate transport was intermittent and strongly correlated with the passage of synoptic-scale high-/low-pressure systems. The high sulphate concentration required heterogeneous conversion of SO_2 to sulphate at a rate of 2.0% hr^{-1}.

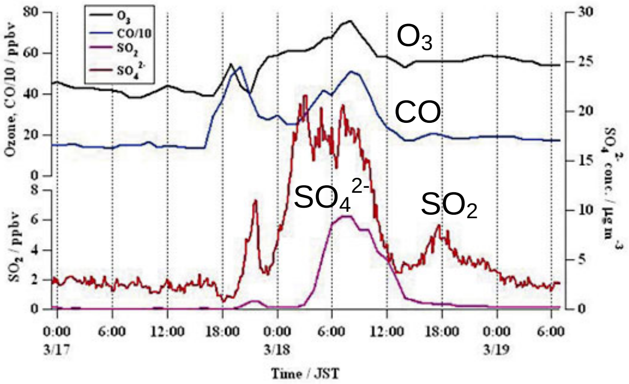


Figure 6.2.3. Time profiles of O_3, CO, SO_2 and sulphate concentrations at Cape Hedo in March 2005.

6.2.1.4 ACE Asia project

The Aerosol Characterization Experiments (ACE) are designed to increase our understanding of how atmospheric aerosol particles affect the Earth's climate system. ACE-Asia was the fourth in this series of experiments organized by the International Global Atmospheric Chemistry (IGAC) Program. ACE-Asia took place during the spring of 2001 off the coast of China, Japan and Korea. The ACE-Asia region includes many types of aerosol particles of widely varying composition and size derived from one of the largest aerosol source regions on Earth. These particles include those emitted by human activities and industrial sources, as well as wind-blown dust. Results from ACE-Asia have improved our understanding of how atmospheric aerosols influence the chemical and radiative properties of the Earth's atmosphere. Here are the main outcomes of ACE-Asia. (1) Dust has been observed by satellite, which is transported half way around the globe being mixed with pollution. (2) Dust transported from East Asia to the Pacific does not absorb as much light as the dark aerosol from South Asia or some previous measurements of dust from the Sahara Desert. There are dramatic regional differences in the chemical and optical properties of aerosols. (3). Combining ACE-Asia suborbital and satellite measurements, monthly averaged (April 2001) cloud-free aerosol radiative forcing at the surface exceeded -30 W m^{-2} in a plume covering the Yellow Sea, East China Sea, Sea of Japan and region downwind of Japan.

In the ACE-Asia Project, not only aerosol characteristics, wet deposition was investigated (Decesaria *et al*., 2005). Water-soluble organic compounds (WSOC) in size-segregated aerosol samples, cloud water and wet depositions collected at Jeju Island were characterized. WSOC in the size-segregated aerosol samples at the Gosan coastal site show the occurrence of oxidized species, enriched in the accumulation mode, and a functional group composition characteristic of continental industrialized areas, with a small impact from biomass burning sources. The chemical classes identified and quantified by liquid chromatography accounted for 88% of the water-soluble organic carbon on average (Figure 6.2.4). The relatively high proportion of polycarboxylic acids (PA, 33–40% of total characterized WSOC) observed throughout the campaign indicates the persistence of WSOC in air masses which spent up to 5 days in the marine boundary layer (MBL) of the Yellow Sea. The analysis of cloud water and wet depositions collected at a mountain site in Jeju shows substantial differences in the organic composition. PA acids contribute to cloud water WSOC in far smaller proportions (6–11%) than in the fine MBL aerosols. It was proposed that such differences resulted from the entrainment of aerosol particles from the lifted layers into the stratiform clouds. These hypothesized features of the vertical profiles in the inorganic and organic aerosol chemical composition are in agreement with the results of aircraft-based measurements performed during the same experiment.

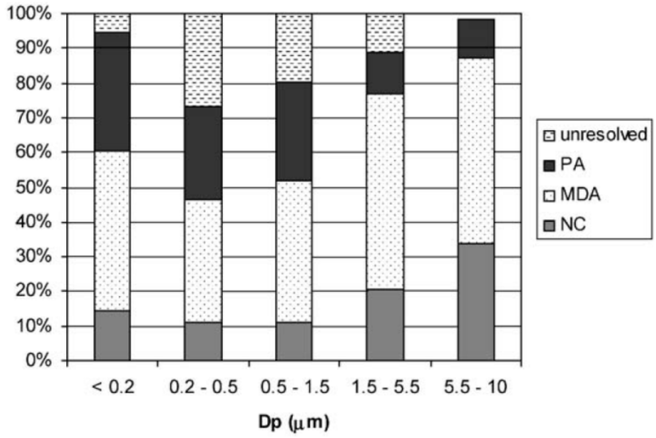


Figure 6.2.4. Mean distribution of Neutral compounds (NC), mono/di carboxylic acids (MDA) and (polycarboxylic acids) PA in size segregated aerosol samples collected at Gosan, Korea.

6.2.1.5 Intensive observation at Gosan, Korea

Gosan, located on Jeju Island, Korea in the East China Sea is a super-site for meteorological and atmospheric measurements and has been utilized for many studies of dust storms and pollution transport in recent decades. It has been a primary ground-based monitoring site during the ACE-Asia, ABC and the PACific Dust EXperiment (PAC-DEX) (Stith *et al*., 2009). The springtime at Jeju Island is associated with the highest aerosol loadings caused by dust events originating over the Asian mainland. Polluted air masses typically travel to Jeju from the northern part of China and contain a combination of dust and anthropogenic sources. The dust in the region is thought to originate primarily in the Gobi desert and has been called Kosa or Yellow Sand. In recent decades, dust storms have been becoming more frequent in East Asia. As a part of the PAC-DEX campaign, particulate matter was collected at Gosan, Korea, a remote location in the East China Sea, from late-March through May, 2007. (Stone *et al*., 2011) Two sizes of particles, fine ($PM_{2.5}$) and coarse ($PM_{10-2.5}$) modes, were analyzed for chemical composition (Figure 6.2.5). Samples were analyzed by mass, elemental and organic carbon, and inorganic ions. Organic molecular markers were also measured using solvent-extraction gas chromatography mass spectrometry in order to identify different pollution sources. This observation demonstrated that carbonaceous aerosols from biomass burning, coal combustion, and motor vehicles containing organic and elemental components that are typically found in fine particles were included in coarse particle mass during dust events. Primary and secondary carbonaceous particles were found to become internally mixed with dust, which is expected to have implications on the dust's chemical composition and surface properties.

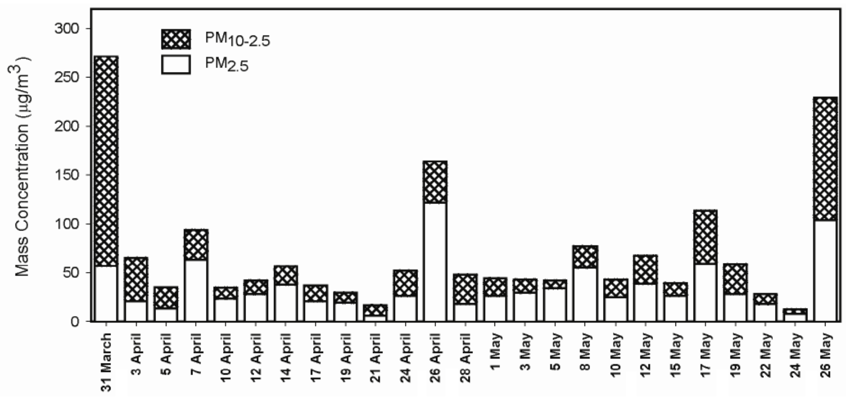


Figure 6.2.5. Ambient concentrations of fine ($PM_{2.5}$) and coarse ($PM_{10-2.5}$) particles measured at Gosan, Korea in 2007.

6.2.2 Acidification and nitrogen leaching in Japanese catchments

In Japan, acid deposition was not considered to be likely to lead to mark increases in acidity of surface waters because most catchments have a high acid-neutralizing capacity (ANC) (Ohte *et al.*, 2001). Several studies have revealed the high ANC of Japanese forest soils compared to those in Europe and the U.S. (Tokuchi and Ohte, 1998; Shibata *et al.*, 2001). However, significant decreases in both pH and alkalinity were recently found in Lake Ijira catchment in central Japan, where the wet depositions of H^+, non-sea-salt (nss) SO_4^{2-} and NO_3^- were at the highest levels in Japan (Yamada *et al.*, 2007; Nakahara *et al.*, 2010). Moreover, acidification of river water was reported in the rivers also in central Japan, especially in the areas covered by granite, rhyolite or similar silicate-rich rocks (Kurita and Ueda, 2006; Matsubara *et al.*, 2009). This paper summarizes the current situation of acidification and nitrogen leaching in Japanese catchments.

6.2.2.1 Acidification and nitrogen leaching in Lake Ijira catchment

The Ministry of the Environment of Japan (The Environment Agency of Japan, until 2000) conducted the Japanese Acid Deposition Survey (JADS) from 1983 to 2002 (Committee for Acid Deposition Measures, 2004). As a part of JADS, long-term monitoring was implemented in five representative lakes from Hokkaido (northernmost) to Kyushu (southernmost) in Japan. Based on the accumulated data since 1988, Yamada *et al.* (2007) investigated effects of acid deposition in these lakes. They found that the pH of Lake Ijira water had declined slightly since the mid-1990s, corresponding with the significant downward trends seen in the pH and alkalinity of the stream water flowing into the lake. Moreover, significant upward trends in the EC and NO_3^- concentrations were also found in both the lake and stream water. Such significant acidification trends were only observed in Lake Ijira. Upward trends in pH and alkalinity, upward trend in alkalinity and no trend were found in Lake Banryu, Lake Kamakita, and the other two lakes, respectively.

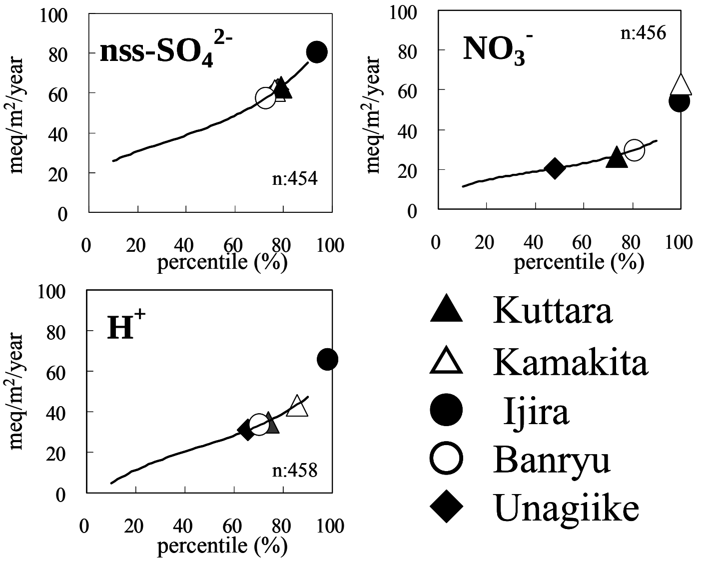


Figure 6.2.6. Ogive of the wet deposition in the five study sites and the other JADS monitoring sites in Japan *(Committee for Acid Deposition Measures, 2004; Yamada et al., 2007)*.

In fact, the wet depositions of H^+ and nss-SO_4^{2-} were the highest at Lake Ijira, although the NO_3^- was the second to that of Lake Kamakita, as shown in the ogive of the annual wet depositions in the monitoring sites (Figure 6.2.6). The Lake Ijira is located about 40 km North of the Chukyo industrial area near Nagoya, one of the biggest industrial areas in Japan. It was suggested that air pollution from this industrial area was transported to the mountainous areas that include the Lake Ijira catchment (Kitada *et al*., 2000). Although the depositions at some other lakes were also at high levels in the JADS sites, lakes other than Lake Ijira showed no significant trends revealing acidification of the water.

Table 6.2.1. General information on the geology, surface soil, vegetation and water chemistry in the catchments of the five lakes *(modified table from Yamada et al. 2007)*.

	Kuttara	Kamakita	Ijira	Banryu	Unagiike
Lake Type	Caldera	Dammed lake	Dammed lake	Dammed lake	Caldera
Bedrock	Volcanic rock, non-alkaline mafic rock; formed in the late Pleistocene	Sedimentary rock, Chert including schalstein and limestone; formed in the Paleozoic	Chert and mudstone; formed in the Mid Jurassic to Early Cretaceous	Sedimentary rock	Volcanic rock, non-alkaline mafic rock; formed in the Quaternary
Surface soil	Brown forest soil, volcanic ash soil	Brown forest soil	Brown forest soil	Brown forest soil	Andosol, red-yellow podzolic soil

Vegetation	Japanese oak, Painted maple	Japanese cedar, Japanese cypress	Japanese cypress, Japanese cedar, Japanese red pine	Japanese red pine, Japanese black pine	Japanese cypress, Japanese cedar
pH*	7.51	8.24	7.20	7.09	8.15
EC*, mS m^{-1}	6.40	13.65	4.38	9.91	11.90
Alkalinity*,µg L^{-1}	305	857	151	136	602

**Median values from 1995 to 1997 in surface water collected from the center of the lakes.*

Characteristics of the lakes may be related to the trends. As shown in Table 6.2.1 (based on Yamada *et al*., 2007), Lake Kuttara and Lake Unagiike are both caldera lakes, and their catchments are covered with volcanic rocks with high ANC. The geology of the Lake Kamakita catchment also includes bedrock with high ANC, such as limestone. Lake Banryu was strongly affected by sea salt (e.g. Cl^-, 21.2 mg L^{-1}; Yamada *et al*. 2007). On the other hand, the bedrock of the Lake Ijira catchment is mainly chert with low ANC. Brown forest soils with low ANC are dominant in the catchment. The alkalinity of both the stream and lake waters was low among the lakes studied. This indicates that the aquatic environment of the Lake Ijira catchment is more sensitive to acid deposition than those of the other catchments. It was suggested that the acidification in Lake Ijira and inflowing streams was related to the geology of the catchment and the high acid deposition loading.

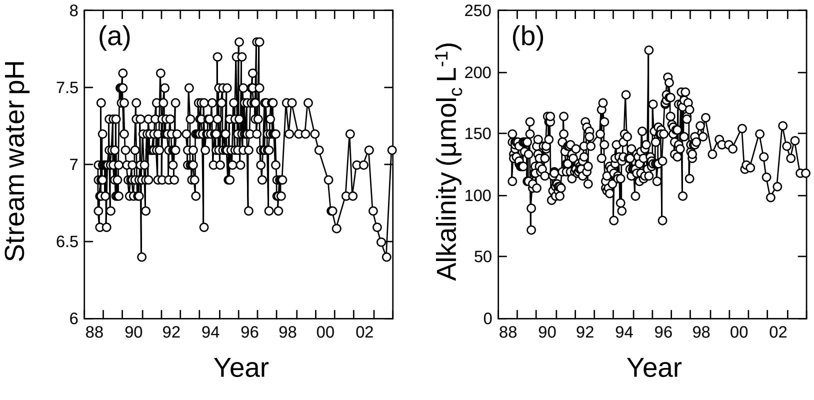


Figure 6.2.7. (a) pH and (b) alkalinity of the stream water inflowing Lake Ijira *(Nakahara et al., 2010)***.**

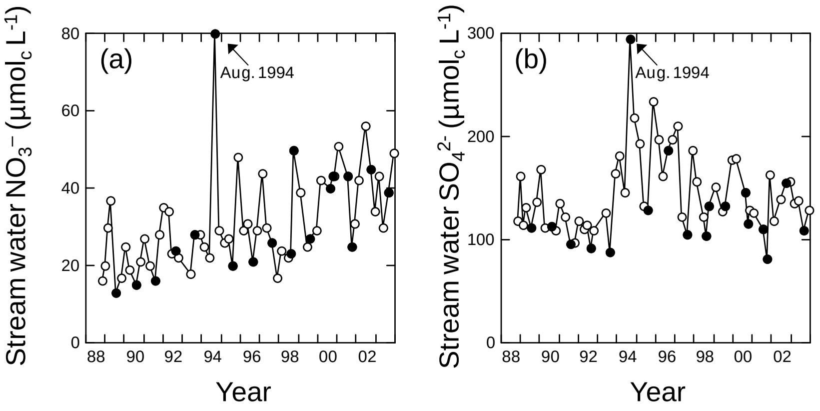


Figure 6.2.8. Concentrations of (a) NO_3^- and (b) SO_4^{2-} in the stream water. Closed circles represent data observed in summer (July, August, and September) and open circles represent data observed in the other seasons. *(Nakahara et al., 2010)****.*** *The peak concentration in August 1994 was observed when the water level was very low due to extremely low precipitation in July 1994.*

6.2.2.2 Possible mechanisms on acidification and nitrogen leaching in Lake Ijira catchment

The pH and alkalinity of the stream water inflowing the Lake Ijira did not show a linear trend. The stream water pH increased until 1996/1997, and then decreased thereafter (Figure 6.2.7a). The stream water alkalinity increased from 1995 to 1996/1997, and decreased from 1996/1997 onward (Figure 6.2.7b). Thus, actual acidification of the stream water started from mid-1990s.

The stream water NO_3^- concentration increased throughout the monitoring period as shown in Figure 6.2.8a). In contrast to the previous period, high NO_3^- concentrations were often observed during the summer from 1996/1997 onward (Figure 6.2.8a). The NO_3^- concentration could not be suppressed by vegetation uptake after 1996/1997. The enhanced NO_3^- discharge during the growing season may be the primary causes for the stream water acidification since 1996/1997 (Nakahara *et al*., 2010). According to the classification proposed by Stoddard (1994), the stage of nitrogen saturation at the Lake Ijira catchment shifted from stage 1 before 1996/1997 to stage 2 thereafter. Moreover, the stream water SO_4^{2-} concentration increased greatly in August 1994, remained relatively high until around 1996, and then decreased gradually (Figure 6.2.8b). It seemed that some changes in the biogeochemical process occurred in mid-1990s.

The H^+ and nitrogen deposition in the Lake Ijira catchment increased since 1994, and sulphur deposition decreased until 1994 (Nakahara *et al*., 2010). It was suggested that these trends were partly responsible for the trends in stream water pH, NO_3^- and SO_4^{2-}, respectively, during the corresponding periods. Nakahara *et al*. (2010) estimated the total depositions based on the wet deposition by the wet-only sampler and the dry deposition by the inferential method. Mean annual wet depositions in Lake Ijira catchment were 0.68 kmol ha^{-1} yr^{-1} for H$^+$, 0.42 kmol ha^{-1} yr^{-1} for SO_4^{2-}, and 1.00 kmol ha^{-1} yr^{-1} for dissolved inorganic nitrogen (DIN, NO_3^- plus NH_4^+). Dry depositions in 2003/2004 were 0.15 kmol ha^{-1} yr^{-1} for H$^+$, 0.15 kmol ha^{-1} yr^{-1} for SO_4^{2-}, and 0.37 kmol ha^{-1} yr^{-1} for DIN. Thus, total depositions (wet + dry) were 0.83, 0.57, and 1.37 kmol ha^{-1} yr^{-1}, respectively. Nitrogen deposition of 1.37 kmol ha^{-1} yr^{-1} in the catchment exceeded the widely observed range of thresholds of 0.5 to 0.7 kmol ha^{-1} yr^{-1} (7 to 10 kg N ha^{-1} yr^{-1}) that are reported to enhance NO_3^- discharge into surface waters in Europe and North America.

Surface mineral soils in the Lake Ijira catchment became strongly acidified until the mid-1990s (Nakahara *et al.*, 2010). Generally, soil pH decreased, exchangeable Al increased, and exchangeable Ca^{2+} remained low until 1994/1995 (Figure 6.2.9). However, exchangeable Ca^{2+} began to increase in 1994 and showed a large peak in 1996/1997 (Figure 6.2.9c). The average soil pH of 3.9 measured in 2003 was among the lowest observed in Japanese forests (Takahashi *et al.* 2001). The continuous and rapid decrease in soil pH until mid-1990s (Figure 6.2.9a) may have increased SO_4^{2-} adsorption on soil minerals and decreased SO_4^{2-} discharge into streams (Nodvin *et al.* 1988), as shown in Figure 6.2.8b). Moreover, the simultaneous increases in pH and exchangeable Ca^{2+} in 1996/1997 were similar to the soil chemical changes induced by lime application, which could stimulate net nitrification and NO_3^- leaching from soils (e.g. Nohrstedt 2002).

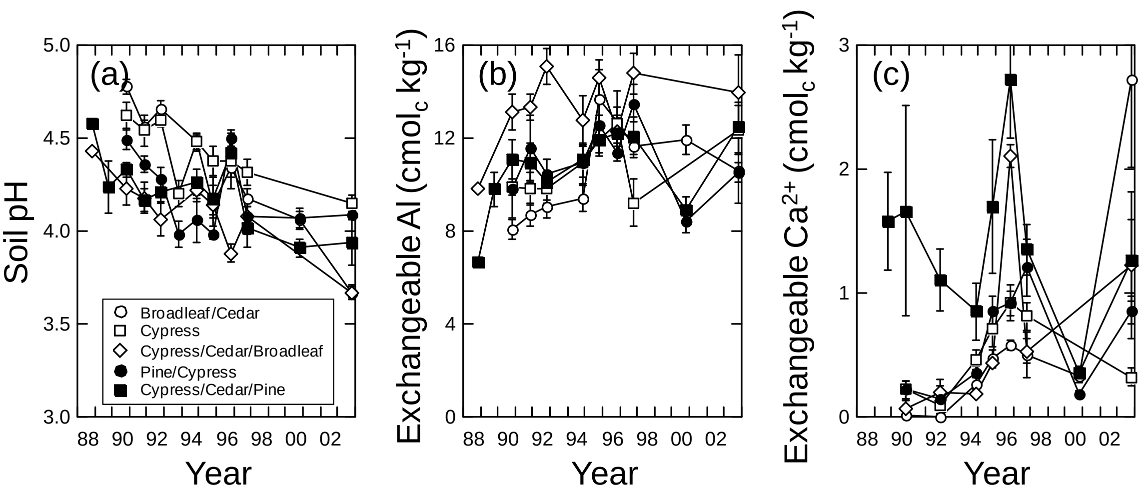


Figure 6.2.9. (a) Soil pH, (b) exchangeable Al, and (c) exchangeable Ca^{2+} of the surface mineral horizon (A horizon) in the Lake Ijira catchment. *(Nakahara et al., 2010). Data represent mean values of five samples from each plot. Error bars represent standard errors. The legend shows dominant species in each stand in decreasing order.*

There were two climatic anomalies in this area during the monitoring period (Geng *et al.* 2000); an extremely cool and wet summer in 1993 and extremely high temperatures and severe droughts in 1994. The monthly mean temperatures recorded in July 1993 (23.8°C) and July 1994 (29.4°C) were the lowest and highest since 1950, respectively. The monthly precipitation in July 1994, 28 mm, was less than one-tenth of the average monthly precipitation in July (347 mm) and the lowest monthly precipitation in July since 1950. The anomalously cold summer in 1993 and the summer drought in 1994 might have decreased nitrogen retention since the mid-1990s (Nakahara *et al.*, 2010).

The basal area index (BAI) of Japanese cedar and Japanese cypress in the Lake Ijira catchment increased during the 1960s and 1970s, reached a maximum around the early 1980s, and decreased from the mid-1980s onward (Figure 6.2.10). After the peak (in 18 yrs for Japanese cedar and 21 yrs for Japanese cypress), BAI decreased in a stair-step pattern, with two significant periods of decrease: the first decrease began in 1982/1984 and ceased around 1988, and the second decrease began in 1994/1993 and ceased around 1998. This pattern suggests that factors other than forest age had a strong effect on forest growth. The climate anomalies might have triggered the second decrease in BAI in 1993/1994 (Nakahara *et al.*, 2010).

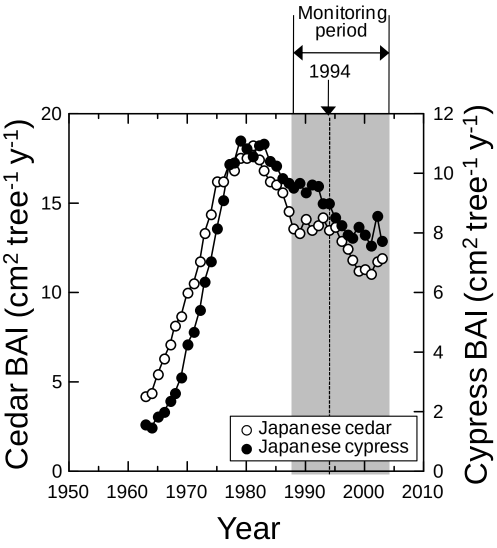


Figure 6.2.10. Mean basal area index (BAI) of Japanese cedar (n = 50) and Japanese cypress (n = 50). ***(Nakahara et al., 2010).*** *Average ages in 2004 were 41 yrs for Japanese cedar and 42 yrs for Japanese cypress. The arrow indicates the year of the most severe summer drought (1994).*

As described above, acidification and nitrogen leaching in the Lake Ijira catchment occurred in the several conditions:

i. The geology of the catchment is dominated by Chert, which may have low ANC.
ii. The stream water has low alkalinity and EC, reflecting the geology.
iii. Atmospheric depositions were among the highest levels in Japan.
iv. The nitrogen deposition exceeded the threshold of 0.7 kmol ha^{-1} yr^{-1} (10 kg N ha^{-1} yr^{-1}) for enhancing NO_3^- discharge into surface waters in Europe and North America.

The deposited nitrogen had been retained in the ecosystem of the catchment. The leaching of NO_3^- to the stream water was controlled by vegetation uptake prior to 1996/1997. It seemed that the ecosystem was still balanced against the high deposition loads, although soil had been acidified. However, the imbalance proceeded from the climatic anomalies in 1993 and 1994, probably with the following steps:

v. The catchment experienced anomalously cold summer and drought summer in 1993 and in 1994, respectively.
vi. The climate anomalies triggered the decrease of plant growth, which resulted in nitrogen retention by vegetation since the mid-1990s.
vii. The 1994 summer drought also enhanced needle litterfall in coniferous stands and reduced Ca^{2+} uptake by vegetation, which resulted in an increase of exchangeable Ca^{2+} and pH in surface mineral soil. The sudden increase in soil pH accelerated H^+-generating net nitrification and led to the accumulation of NO_3^- in surface soils.
viii. The H^+ and nitrogen deposition increased since 1994 additionally.
ix. In spite of much NO_3^- supply, its retention by vegetation was reduced since mid-1990s.
x. After 1996/1997, the stream water NO_3^- concentration increased with increasing stream discharge, and the seasonality of stream water NO_3^- concentration was lost.
xi. Stream pH and alkalinity began decreasing in 1996/1997.

It seemed that the climatic anomalies triggered the imbalance of biogeochemical processes in the acid-sensitive ecosystem under the high deposition loads.

6.2.2.3 River water acidification in central Japan

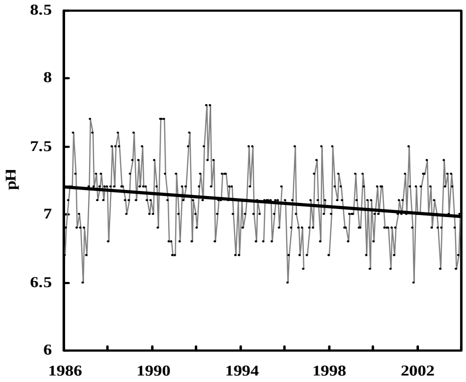


Figure 6.2.11. Long-term variation of monthly data in river water pH at k17 (Tokonami River, a blanch of Agano River) in Niigata Prefecture. *(Matsubara et al., 2009).*

In the neighboring areas of the Lake Ijira catchment in Gifu Prefecture, river water acidification has also been reported recently. Based on the historical data longer than 30 years, Kurita and Ueda (2006) reported declining trends of pH in upstream rivers and lakes in mountainous area in Nagano Prefecture, which was located the Northeast of Gifu Prefecture. As shown in Figure 6.2.11, Matsubara *et al*. (2009) also reported declining trends of pH for the recent 15 years in rivers in Gifu Prefecture and Niigata Prefecture, which was located the Northeast of Nagano Prefecture. Thus, river water acidification was reported in three neighboring prefectures in central Japan. The papers above unanimously reported that the acidification trends were found in the rivers from the areas, which were dominated by low-ANC geology, such as granite, rhyolite and chert. In fact, the river waters had low pH, EC and alkalinity, which reflected the geology (Matsubara *et al*., 2010).

Moreover, atmospheric deposition may be large in central Japan. As described above, Gifu Prefecture is expected to be affected by emissions from Chukyo Industrial Area. Niigata Prefecture may be affected by long-range transport from the Asian Continent, especially by seasonal winter winds (Kamisako *et al*., 2008; Matsubara *et al*., 2009). The wet depositions of nitrogen and nss-sulphur in 2003 at Nagaoka station in the center of Niigata Prefecture were among the highest levels in Japan, 19 kg N ha^{-1} yr^{-1} and 18 kg S ha^{-1} yr^{-1}, while those at the Lake Ijira station were 17 kg N ha^{-1} yr^{-1} and 18 kg S ha^{-1} yr^{-1}, respectively (Matsubara *et al*., 2010). The high mountainous area in Nagano Prefecture may have both effects of long-range transport from Asian Continent and transport from Tokyo and/or Chukyo Industrial areas in winter and summer, respectively (Kurita and Ueda, 2006).

Other factors, such as hot spring drainage and land-use changes, cannot adequately explain all of the acidification phenomena (Matsubara *et al*., 2010). River water in the areas dominated by low-ANC geology may become acidified if acid loading from the atmosphere is quite large.

The facts above suggest that high acid deposition loads may cause acidification of surface waters, in particular, in acid-sensitive catchments. Therefore, the area, whose geology has low ANC and in which acid deposition is high, should carefully be monitored as “hotspots” for acidification. Probably, the hotspots for nitrogen leaching should also be considered based on vegetation features and nitrogen deposition amount, although it is still uncertain whether the threshold value, 10 kg ha^{-1} yr^{-1}, is directly applicable to Japanese catchments or not. In addition to continuous monitoring in the acidified catchments, identification of the hotspots should be promoted taking into account distribution of geology, soil, vegetation, and atmospheric deposition.

6.3 Emission Inventories

6.3.1 Global scale inventories

This section describes the global emission inventories for anthropogenic emissions based on Granier *et al*. (2011). The list of the global emission inventories, covering comprehensive pollutants including NO_x and SO_2 are summarized in Table 6.3.1.

Table 6.3.1 List of global emission inventories for anthropogenic emission during 1980-2010.

Inventory	Year	Spatial resolution	Author, web site
ACCMIP	1980–2010	0.5 deg	Lamarque *et al*., 2010 http://ether.ipsl.jussieu.fr– ECCAD
ACCity	1980–2010	0.5 deg	http://ether.ipsl.jussieu.fr – ECCAD
RCPs	2000–2010	0.5 deg	http://ether.ipsl.jussieu.fr – ECCAD
EDGAR v3	1990, 1995, 2000	1 deg	Olivier *et al*., 2005; http://edgar.jrc.ec.europa.eu
EDGAR v4.1	1970-2005	1 , 0.5, 0.1 deg	http://edgar.jrc.ec.europa.eu
HYDE	1980, 1990	1 deg	Van Aardenne *et al*., 2001 http://www.pbl.nl/en/themasites/hyde/index.html
RETRO	1980–2000	0.5 deg	http://retro.enes.org/data_emissions.shtml
GAINS	1990–2010	–	http://gains.iiasa.ac.at/gains

6.3.1.1 Global emission inventories

This section indicates the fundamental characteristics of main global inventories covering comprehensive pollutants.

ACCMIP emissions and representative concentration pathways (RCPs)

The main purpose of this work was to provide consistent gridded emissions of short-lived gases and aerosols for use in chemistry model simulations needed by climate models for CMIP5 (Coupled Model Intercomparison Project Phase 5). ACCMIP (Atmospheric Chemistry and Climate Model Intercomparison Project) emissions was a dataset of monthly, sectoral, gridded anthropogenic and biomass burning emissions covering the historical period (1850–2000) at a horizontal resolution of 0.5° in latitude and longitude (Lamarque *et al*, 2010). The reference year is 2000 and historical emissions were estimated on the basis of the emission inventories discussed below. The projections of future emissions, RCP (Representative Concentration Pathways), are based on the outcomes of four independent modeling teams. Each applied a set of algorithms to ensure consistency with the 2000 emission inventory. The RCPs are named according to their 2100 radiative forcing level, i.e. RCP 8.5 corresponds to a radiative forcing of 8.5 Wm^{-2}.

EDGARv32, EDGARv4.1 and HYDE inventories

Originally EDGAR (Emission Database for Global Atmospheric Research) (1992–2004) was developed by the Netherlands Environmental Assessment Agency (PBL) and the Netherlands Organization for Applied Scientific Research (TNO). Since 2005, the EDGAR project is a collaboration between PBL and the European Commission’s Joint Research Center (JRC). EDGARv32 provides emission of direct greenhouse gases for the period of 1970–1995. Emissions of ozone precursor and acidifying gases for the 1990–1995 are also available by country and on a 1x1 degree grid (Olivier and Berdowski, 2001). EDGARv32-FT2000 is the global anthropogenic emission inventory for the year of 2000. The HYDE (Hundred Year Database for Integrated

Environmental Assessments) inventory (van Aardenne *et al*., 2001) has been developed by PBL and Utrecht University. HYDE provides emissions of greenhouse gases, ozone precursor gases, and acidifying gases for the period 1890 to 1990, with a 10-year interval, and a 1x1 degree resolution. The EDGARv4.1 is the latest dataset of EDGAR inventory. EDGARv4.1 provides emissions in 3 different spatial resolutions (1, 0.5, and 0.1 degrees) for the period of 1970 to 2005.

RETRO inventory

Within the RETRO project (REanalysis of the TROpospheric chemical composition) (Schultz *et al*., 2007), global gridded data sets were generated, which provide anthropogenic and vegetation fire emissions of several trace gases. The RETRO dataset covers the period from 1960 to 2000 at a monthly time resolution. Anthropogenic emissions in the RETRO inventory are derived from the regional inventories and cover combustion sources only.

GAINS-RAINS inventory

Emissions for Europe (Kupiainen and Klimont, 2007), Asia (Klimont *et al*., 2009) as well as at the global scale (Isaksen *et al*., 2009) are developed at IIASA (International Institute for Applied Systems Analysis), using the GAINS (Greenhouse Gas and Air Pollution Interactions and Synergies) model (http://gains.iiasa.ac.at/gains) which was extended to include greenhouse gases from the previously developed RAINS (Regional Air pollution INformation and Simulation) model. GAINS enables an examination of the co-benefits of simultaneous reduction of air pollution and greenhouse gas emissions. This model systematically compiles country- and sector-specific information for all countries and regions of the world.

6.3.1.2 Comparison of emission inventories and fundamental feature of global trends

We compare the emissions by different global inventories and discuss the global trends for NO_x and SO_2 during 1980-2010. Figure 6.3.1 displays the global anthropogenic emissions of NO_x and SO_2 at the global scale. For NO_x, all inventories show a slight increase in the emissions during 1980-2010. In 1980, the difference between the three totals available is 13%. In 1990 and 2000, five inventories are available, and the maximum difference between the totals is 18% and 17%, respectively. In 2010, the emissions from the RCPs are all within an 8% range, while GAINS estimates are about 20% lower.

For SO_2, most inventories indicate a significant decrease in the emissions during 1980-2010. In 1980, the difference between the lowest and highest estimate is 18%, and reaches 42% in 2000, when the EDGAR v3.2 inventory provides larger values than other inventories. After 2000, the emissions from the AEROCOM show a significant increase, contrary to the other inventories, which do not show significant changes. SO_2 emissions have significantly decreased in most part of the world after 2000, except in Asia. In the AEROCOM inventory, SO_2 emissions are also increasing in China, but they are either constant or increasing as well in other parts of the world, which explains the increase in the global emissions totals.

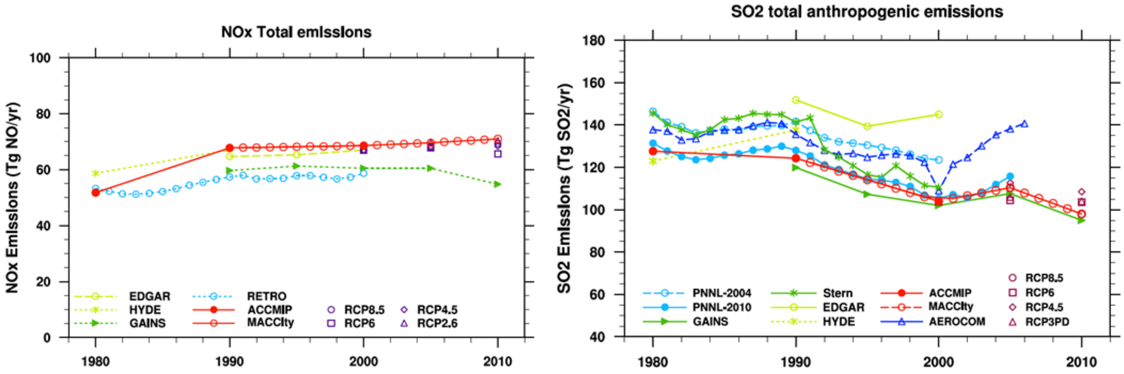


Figure 6.3.1. Comparison of global emissions of NO_x and SO_2 during 1980‐2010.

(Granier et al., 2011).

6.3.1.3 Comparison of emissions trends in global and Asian scale

Figure 6.3.2 demonstrates the temporal variations of global emissions and the ratio of Asian emissions to global emissions for NO_x and SO_2 during 1980‐2005 based on the EDGAR v4.1 inventory. The global NO_x emissions show a slight increase which is driven by the increase of Asian emissions. The ratio of Asian to global NO_x emissions has doubled from 20% in 1980 to 40% in 2005. For SO_2, the global emissions show a decrease, while the Asian emissions increase. As a result, the ratio of Asian to global SO_2 emissions has significantly increased from 18% in 1980 to 50% in 2005. Thus, the contribution of the Asian emissions in the global scale is growing.

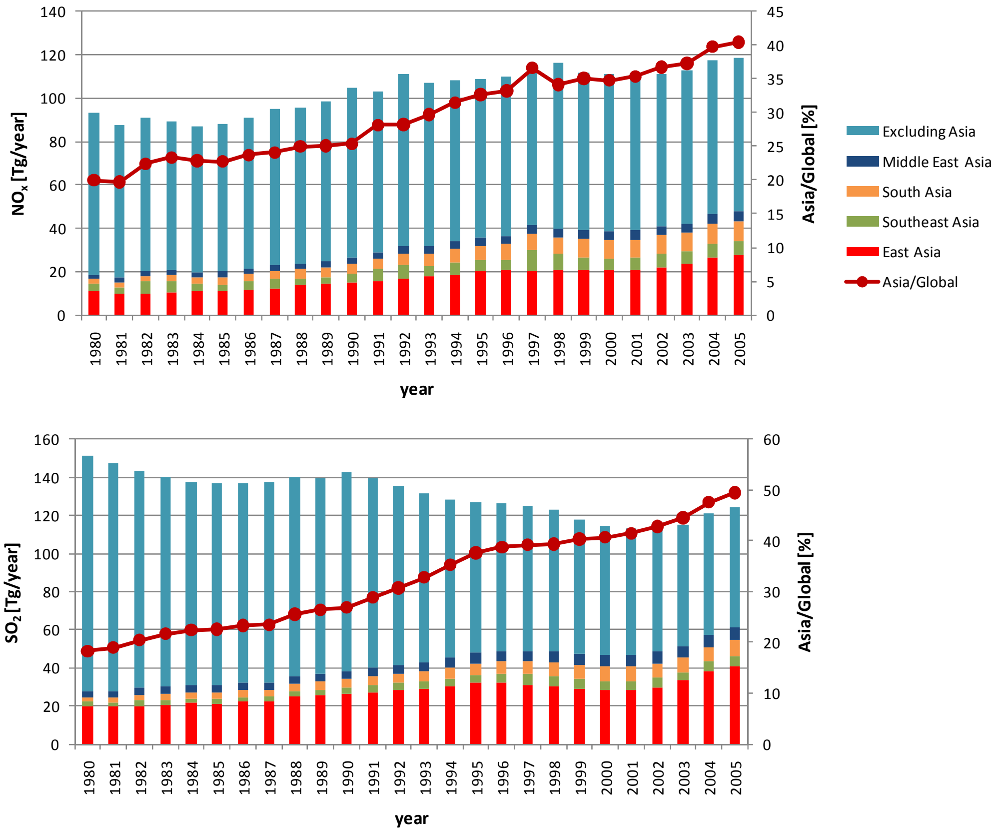


Figure 6.3.2. Temporal variations of global emissions and the ratio of Asian emissions to global emissions for NO_x and SO_2 during 1980-**2005.** *(Source: EDGAR v4.1)***.**

6.3.2 Regional scale inventories in Asia

Anthropogenic emissions in Asia are larger than those in Europe and North America today and will continue to increase in the future (Akimoto, 2003). In fact, recent tropospheric satellite observations have demonstrated that NO_x emissions in China have accelerated impressively since 2000 (Richter *et al.*, 2005; Irie *et al.*, 2009). On the other hand, SO_2 emissions in China have decreased since 2007 due to the penetration of fuel-gas desulfurization (FGD) equipment in the power plants (Lu and Streets, 2011). In the last few years a number of authors reassessed and developed their emission inventories for Asia. The emission inventories for air pollutants in Asia and China are reviewed in this section. Figures 6.3.3 and 6.3.4 compare the Asian and China's emissions from 1980 to the year in the latter half of 2000s according to different emission inventories for SO_2 and NO_x. We compared the historical and current emissions in various inventories: REAS (Regional Emission inventory in ASia) (Ohara *et al.*, 2007), GAINS (Klimont *et al.*, 2009), INTEX-B (Intercontinental Chemical Transport Experiment-Phase B) (Zhang *et al.*, 2009) and others. Table 6.3.2 shows the summary of REAS, GAINS, and INTEX-B.

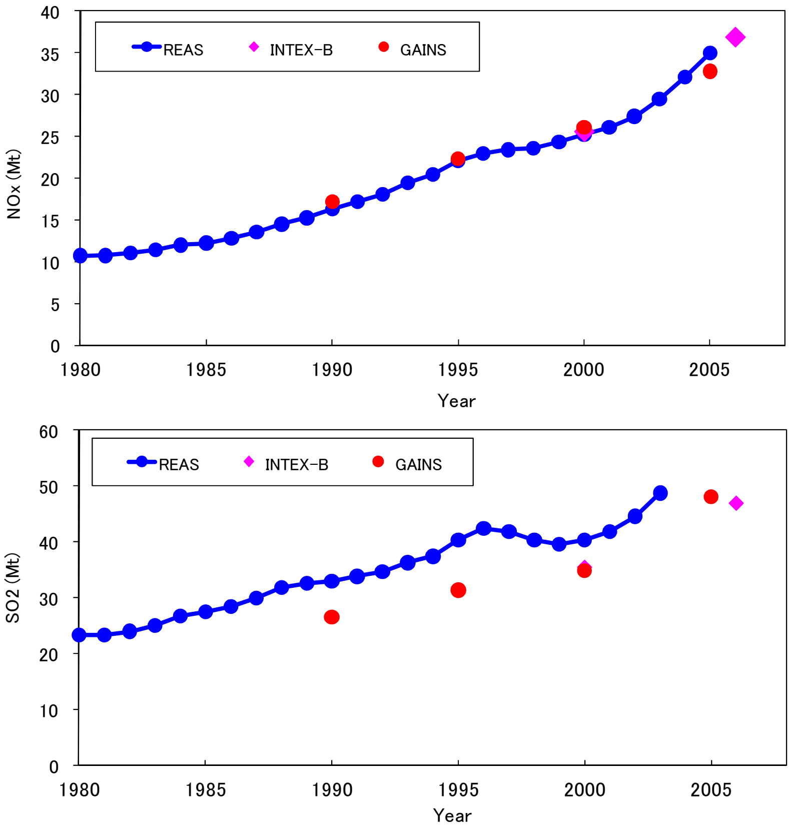


Figure 6.3.3. Time series of NO_x and SO_2 emissions in Asia.

Table 6.3.2. Summary of comprehensive emission inventories for anthropogenic sources in Asia.

	Substances	Year	Spatial resolution
REAS (Ohara *et al*, 2007)	SO_2, NO_x, CO, VOC, NH_3, BC, OC, CO_2, CH_4, N_2O	1980–2003	0.5 deg
GAINS (Klimont *et al.*, 2009)	SO_2, NO_x, VOC, NH_3, PM, CO_2, CH_4, N_2O, F-gas, (BC, OC)	1990–2005	Country, administrative unit
INTEX-B (Zhang *et al.*, 2009)	SO_2, NO_x, CO, VOC, BC, OC, PM_{10}, $PM_{2.5}$	2006	0.5 deg

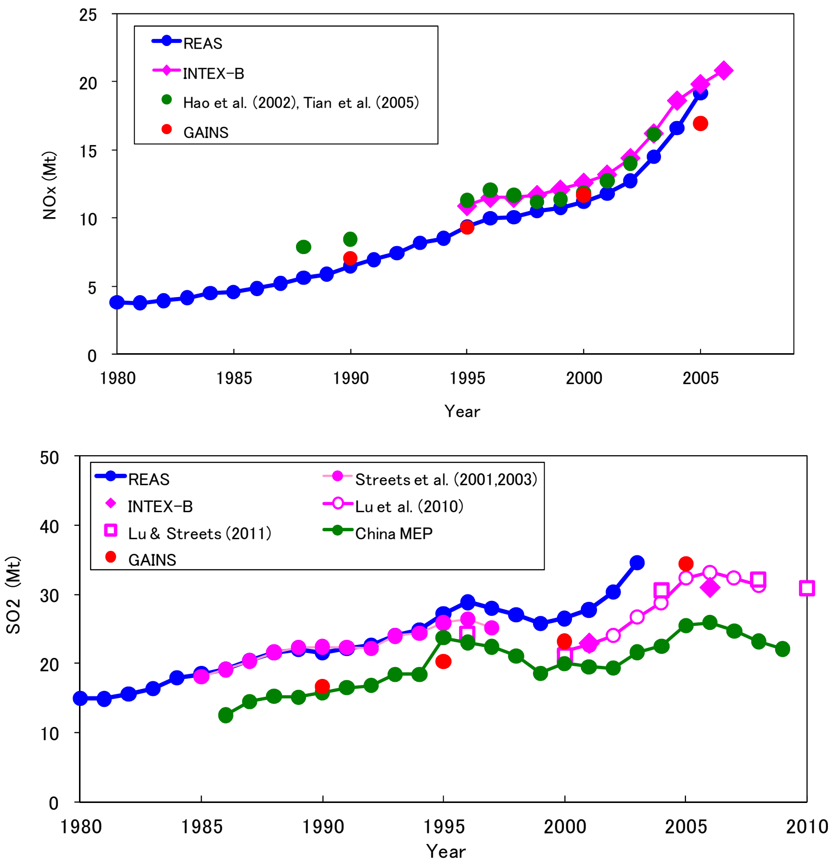


Figure 6.3.4. Time series of NO$_x$ and SO$_2$ emissions in China.

6.3.2.1 NO$_x$

For the Asian total NO$_x$ emissions for 2000 and 2005, the values of REAS, GAINS, and INTEX-B (for 2001 and 2006, respectively) are within a 10 percent variance (25.1–26.1 Mt for 2000, 32.9 – 36.8 Mt for 2005). For Chinese NO$_x$ emissions for 2000, the values of REAS (11.2 Mt), GAINS (11.7 Mt), and Tian *et al.* (2005; 11.9 Mt) were almost the same, whereas the INTEX-B value for 2001 (13.4 Mt) was higher than the values of other inventories. In more recent year, the REAS value (19.2 Mt for 2005) was almost the same as the INTEX-B one (20.8 Mt for 2006), whereas the GAINS value (16.9 Mt for 2005) was almost 10% lower than the REAS value. Total emissions of NO$_x$ in Asia showed a monotonic increase between 1980–2006 with no dips in contrast to the pattern of SO$_2$ emissions. According to the REAS inventory, the emissions increased by 3.5 times from 1980–2005, with values of 10.7 Mt in 1980 and 35.0 Mt in 2005.

Chinese NO$_x$ emissions increased dramatically by 5.0 times from 1980 to 2005, with the higher growth after 2000 (by 1.7 times over only 5 years (Ohara *et al.*, 2007)). These trends in China's NO$_x$ emissions in REAS were consistent with those in other inventories, including GAINS, INTEX-B, and Chinese group's results (Hao *et al.*, 2002; Tian *et al.*, 2005). Zhang *et al.* (2007) present compared a 10-a regional trend of NO$_x$ emissions in China from 1995 to 2004 with the NO$_2$ column trends observed from GOME and SCIAMACHY. The China's NO$_x$ emissions are increasing by 70% during 1995–2004. NO_x emissions and satellite-based NO_2 columns show agreement in temporal evolution. However, the growth rate from the emission inventory is lower than that from the satellite observations, especially in wintertime. The reasons for the discrepancy cannot yet be identified.

6.3.2.2 SO_2

Historical estimates of SO_2 (1990–2006) in Asia show a similar pattern in all studies, although the absolute values vary greatly. According to the REAS inventory, total emissions of SO_2 in Asia increased from 1980 to 1996, but subsequently decreased till 1999, reflecting a decrease in fuel consumption due to the Asian economic crisis; after 2000 SO_2 emissions increased at a phenomenal rate. Those emissions increased by 2.2 times in the period 1980–2003, and, notably, by 3.2 times in India and 2.5 times in China (Ohara *et al.*, 2007). For SO_2 emissions in 2000 in Asia, the GAINS (34.9 Mt) was almost the same as the INTEX-B (35.5 Mt) but lower than REAS (41.2 Mt). In more recent years, GAINS (48.1 Mt) for 2005 is similar to INTEX-B (47.0 Mt) for 2006.

For SO_2 emissions in 2000 (in 2001 for INTEX-B) for China, the GAINS (23.2 Mt) was almost the same as the INTEX-B (22.9 Mt) but higher than MEP (Ministry of Environmental Protection, China) (2010) one (20.0 Mt) and lower than REAS (27.6 Mt). Similarly, for 2005 (in 2006 for INTEX-B), the relative order in emissions is similar to that for 2000 and the maximum difference is almost 30%. (GAINS (34.6 Mt), INTEX-B (31.0 Mt), SEPA (State Environmental Administration, China) (25.9 Mt)). The main reason is the differences in the fuel consumption values used in each inventory (Ohara *et al.*, 2007) and the differences in the impacts of emission control（see Zhang *et al.*, 2009; Klimont *et al.*, 2009. The overall variations of Chinese SO_2 emissions for the period of 1990 to 2006 are similar between REAS, GAINS, SEPA, and Streets *et al.* (2001, 2003) with the INTEX-B. Before 2000, the REAS variation corresponded to that of Street *et al.* (2001, 2003). The variation in MEP (2010) was similar to that in REAS during 1986–1994, but about 7 Mt yr^{-1} lower. In contrast, there were large differences between SEPA and REAS in the decreasing trend of SO_2 emissions during 1995 and 2000, reflecting the differences in coal consumption in each inventory (Akimoto *et al.*, 2006). After 2006, the SO_2 emissions in China began to decrease due to the penetration of FGD equipment in the power plants（Lu *et al.*, 2010; Lu and Streets, 2011; MEP, 2010).

6.3.3 National emission inventory in EANET member countries

Currently, the degradation of air quality due to the rapidly economic development in East Asia is a severe problem especially in the city and industrial zone. The air pollutant emission inventory is one of the efficient tools for managing the air quality. Emission Inventory data can provide the basis information of where air pollutants are emitted and where do they come from. Emission inventories are considered as an indispensable tool for a wide range of air pollution measurements and its control. This can help on quantitative emissions estimates to promote a better understanding of the actual emissions for policy makers and the general public awareness racing. Through this process, the major emission sources can be identified, priorities for emission reduction defined and any data gaps requiring further work are revealed.

An air pollutant emissions inventory details the amounts and types of air pollutants released into the air by source category for a specific geographic area over a specific time period. Pollutants can be emitted from fuel combustion, garbage combustion, transportation, and industrial activities. USEPA suggest there are 5 (five) source type, that is:

Point source

Sources identified on an individual facility basis or as a single source are called point sources. Refineries and industrial plants are examples of point sources. The emission characteristics of individual facilities vary widely and each facility is examined individually.

Area source

Stationary sources not identified individually are called area sources. This term is sometimes extended to cover groups of numerous small point sources, such as dry cleaners or gas stations. Area sources also include the diverse, unpermitted small sources which individually do not emit significant amounts of pollutants but which together make an appreciable contribution to the emission inventory.

Motor vehicle source

On-road motor vehicles consist of passenger cars, trucks, buses and motorcycles.

Non-road mobile source

These sources include boats and ships, trains, aircraft, garden, farm and construction equipment.

Natural source

In addition to man-made air pollution, there are significant quantities of pollutants from natural source. Natural Sources include biological and geological sources, wildfires, windblown dust, and biogenic emissions from plants and trees.

In East Asia, governments and researchers have developed several emission inventories including country, regional and global ones. Regional and global emission inventories by researchers covering East Asia has been reviewed in the previous sections (6.3.1 and 6.3.2) so that we will introduce here national emission inventories reported mostly by the government of by each EANET countries. There are distinct difference in nature of inventories developed by researches and governments. The inventories developed by researchers are mostly for scientific use coupled with three dimensional chemical transport models to analyze regional, hemispherical, and global air pollution focusing long-range transport of air pollutants. On the contrary, inventories developed by governments are mainly for policy analysis mostly for big urban or industrial area suffering serious air pollution. They are also sometimes used being coupled with urban scale chemical transport models.

There are advantage and disadvantage of inventories developed by researchers and government. The advantage of researcher-based inventories is their transparency identifying sources of used parameters, and in many cases published in open literature. The advantage of government-based inventories is that they sometimes are based on more detailed country specific information such as emission factors and activity data which are not easily available to researchers. They have another big advantage that they are for "official" use. It highly desirable that the researcher-based and government-based inventories are harmonized since now a days, it has been realized that long-range transboundary air pollution has to be taken into account in many cases even for evaluating urban and industrial air pollution. However, a harmonized system to compile inventories that are transparent and comparable enough to be agreed among East Asian countries has not yet been established. In order to deepen an understanding of the state of regional air pollution in East Asia and develop capabilities of the countries on atmospheric environmental management, it will be necessary to establish such a system in which inventories can be improved continuously in cooperation with administrative agencies and inventory experts in East Asia. Experience elsewhere has demonstrated the importance of cooperation for enhancing capacity in the preparation of inventories within all countries of a region.

Available information on emission inventory in EANET member countries are listed in Table 6.3.3.

Table 6.3.3. National emission inventories in EANET member countries.

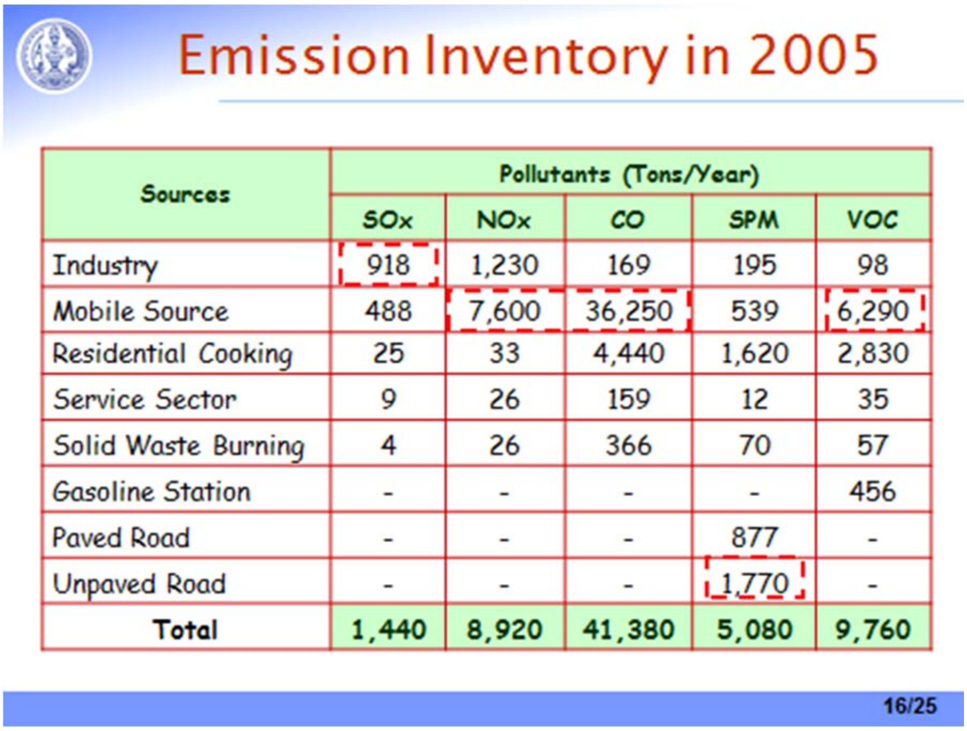
No	Country	Status	Parameters	Reference(s)
1	Cambodia	Case study	CO_2, CH_4, N_2O	http://unfccc.int/resource/docs/natc/khmnc1.pdf Greenhouse Gases Inventory and Future Development in Cambodia: CHEA Chan Thou
2	China	comprehensive	CO_2, SO_2, NOx, VOC, NH_3,	1. Outline of Air Pollutants Emission Inventories in China: Xue Zhigang 2. Construction of Chinese energy and emissions inventory: Glen Peters, Christopher Weber, Jingru Liu
3	Indonesia	Case study	CO_2, CH_4, N_2O, PFC	http://unfccc.int/files/national_reports/non-annex_i_natcom/submitted_natcom/application/pdf/indonesia_snc.pdf NAMAs and MRV MANNER, Ministry of Environment, Republic of Indonesia
4	Japan	Comprehensive	CO_2, CH_4, N_2O, HFCs, PFCs, SF_6, SO_2, NOx, NMVOC, CO;	http://unfccc.int/files/national_reports/annex_i_ghg_inventories/national_inventories_submissions/application/zip/jpn-2011-crf-26apr.zip (national inventory for GHGs and their precursors);

			VOC; SOx, NOx and soot	http://www.env.go.jp/air/osen/voc/inventory.html (VOCs inventory. Only available in Japanese); http://www.env.go.jp/air/osen/kotei/index.html (Comprehensive survey for air pollutants emission. Updated every three years. Only available in Japanese)
5	Lao PDR	Case study	Green house gas: CO_2 CH_4 CO N_2O NOx	http://unfccc.int/resource/docs/natc/laonc1.pdf Lao_People_s_Democratic_Country_Report.pdf
6	Malaysia	Case study	CO_2, CH_4, N_2O	http://unfccc.int/resource/docs/natc/malnc1.pdf The inventory of greenhouse gases (GHG) in Malaysia , By Azman Zainal Abidin
7	Mongolia	Case study	GHG Inventory	Dr. DORJPUREV JARGAL, EEC Mongolia
8	Myanmar	Case study	GHG Inventory	http://www-gio.nies.go.jp/wgia/wg8/pdf/3-wg2-7_khin_lay_swe.pdf
9	Philippines	Case study	PM, SOx, NOx, CO, VOC, TOGs,	http://www.adb.org/Documents/Reports/Urban-Air-Quality-Management/philippines.pdf National Air Quality Status Report [2005-2007] , www.emb.gov.ph
10	Republic of Korea	comprehensive	NOx, VOC, CO, NH3, SO_2, PM_{10}, TSP.	http://unfccc.int/files/national_reports/ Korea Emissions Inventory Processing Using the US EPA's SMOKE System Soontae Kim, Nankyoung Moon and Daewon W. Byun
11	Russia	comprehensive	SOx, NOx, VOCs, CO, NH_3, HMs, POPs, PM, etc.) CO_2; Methane (CH4); N_2O; (HFCs); Perfluoro carbons (PFCs); and Sulphur hexafluoride (SF6)	Review of the Russian experience in the implementation of national inventories under international agreements. Dr. V. Ginzburg, Dr. S. Gromov
12	Thailand	comprehensive	SOx, NOx, VOCs,	http://unfccc.int/resource/docs/natc/thainc1.pdf Experience in the development of the emission inventory in Thailand. Dr.Jariya Sukhapan
13	Vietnam	Case study in Hanoi, Ho Chi	NOx, SO_2, CO,	NILU, SVCAP,

		Minh city	BNZ, PM_{10},	Urban air quality modeling and management in Vietnam context, Dr. Ngo Tho Hung, NERI, DK www2.dmu.dk/pub/PhD_Hung.pdf.

An example of emission inventory of air pollutants in Phnom Penh City, Cambodia is shown in Table 6.3.4.

Table 6.3.4. Emission inventory of air pollutants in Phnom Penh City, Cambodia.



Emission Inventory in 2005

Sources	Pollutants (Tons/Year)				
	SOx	NOx	CO	SPM	VOC
Industry	918	1,230	169	195	98
Mobile Source	488	7,600	36,250	539	6,290
Residential Cooking	25	33	4,440	1,620	2,830
Service Sector	9	26	159	12	35
Solid Waste Burning	4	26	366	70	57
Gasoline Station	-	-	-	-	456
Paved Road	-	-	-	877	-
Unpaved Road	-	-	-	1,770	-
Total	**1,440**	**8,920**	**41,380**	**5,080**	**9,760**

16/25

Cited from Status of Air Quality Management in Phnom Penh City, Cambodia.

6.4 Chemical transport modeling studies

6.4.1 MICS-Asia II

The Model Inter-comparison Study for Asia (MICS-Asia) is an activity to help develop a better common understanding of the performance and uncertainties of chemical transport models (CTMs) in East Asia. Phase II was carried out during the period from 2003 to 2007 associated with ADORC (now ACAP) and IIASA as a part of EANET additional activities. The output of MICS-Asia II was published as several papers in Atmospheric Environment Vol., 42, Issue 15, 2008. The essential part of the publication is introduced here.

MICS-Asia II studies was expanded to include nitrogen compounds, O_3 and aerosols in addition to sulphur compounds which were studied in precedent MICS-Asia I, and examined four months in different seasons; March, July, and December 2001, and March 2002 (Carmichael *et al.*, 2008). Newly obtained observational data of EANET in those years were fully utilized as region-wide data base for comparison. Nine regional modeling groups participated the studies, and simulated O_3 and its precursors, sulphur dioxide, secondary aerosols and acid deposition. The common emissions (Streets *et al.*, 2003) and boundary conditions derived from the global CTM (Chemistry Transport Model), MOZART (Model for OZone And Related chemical Tracers) v. 2.4 has been used for all the simulations. The spatial resolution of the models ranges from 36 to 45 km or 0.5°.

Figure 6.4.1 shows the domains used by nine individual models together with the numbered 43 EANET observation sites used in the study. Here, some of the major features that have been identified through the intercomparison studies are presented. The comparison of prediction and observed monthly mean values of SO_2 are shown in Figure 6.4.2 (Carmichael *et al.*, 2008). It is claimed that estimates of sulphur emissions in East Asia are believed to be most accurate than any other species treated in MICS-II due to long-term studies of air pollution and acid deposition in the region, and thus the comparison of predictions for sulphur compounds provides a bench-mark for the intercomparison results. Note that the concentration scales in the figure are logarithmic. In general, spatial variability e.g. higher levels over China and moderate levels over Japan, which reflect the emission intensities in these regions. Systematic over-prediction at the Weishuiyuan site of China (site 4) and the Ijira site of Japan (site 22) has been thought to probably reflect rather

EANET Sites in 2002

ID	Site Name	Characteristics	Lat	Lon	Country
1	Guanyinqiao	Urban	29.6	106.5	China
2	Jinyunshan	Rural	29.8	106.4	
3	Shizhan	Urban	34.2	109.0	
4	Weishuiyuan	Rural	34.4	108.9	
5	Jiwozi	Remote	33.8	108.8	
6	Hongwen	Urban	24.5	118.1	
7	Xiaoping	Remote	24.9	118.0	
8	Xiang-Zhou	Urban	22.3	113.6	
9	Zhuxian-Cavern	Urban	22.2	113.5	
10	Jakarta	Urban	6.2	106.8	Indonesia
11	Serpong	Rural	6.3	106.6	
12	Kototabang	Remote	0.2	100.3	
13	Bandung	Urban	6.9	107.6	
14	Rishiri	Remote	45.1	141.2	Japan
15	Tappi	Remote	41.3	141.4	
16	Ogasawara	Remote	27.8	142.2	
17	Sado-seki	Remote	38.2	138.4	
18	Happo	Remote	36.7	137.8	
19	Oki	Remote	36.3	133.2	
20	Yusuhara	Remote	33.4	132.9	
21	Hedo	Remote	26.9	128.3	
22	Ijira	Rural	35.7	136.7	
23	Banryu	Urban	34.7	131.8	
24	Petaling-Jaya	Urban	3.1	101.7	Malaysia
25	Tanah-Rata	Remote	4.5	101.4	
26	Ulaanbaatar	Urban	47.9	106.8	Mongolia
27	Terelj	Remote	48.0	107.5	
28	Metro-Manila	Urban	14.6	121.1	Philippines
29	Los-Banos	Rural	14.2	121.3	
30	Kanghwa	Rural	37.7	126.3	Republic of Korea
31	Cheju	Remote	33.3	126.2	
32	Imsil	Rural	35.6	127.2	
33	Mondy	Remote	51.7	101.0	Russia
34	Listvyanka	Rural	51.9	104.9	
35	Irkutsk	Urban	52.2	104.3	
36	Primorskaya	Rural	43.7	132.1	
37	Bangkok	Urban	13.8	100.5	Thailand
38	Samutprakarn	Urban	13.7	100.6	
39	Patumthani	Rural	14.0	100.8	
40	Vachiralongkorn-Dam	Remote	14.8	98.6	
41	Mae-Hia	Rural	18.8	98.9	
42	Hanoi	Urban	21.0	105.9	Vietnam
43	Hoa-Binh	Rural	20.8	105.3	

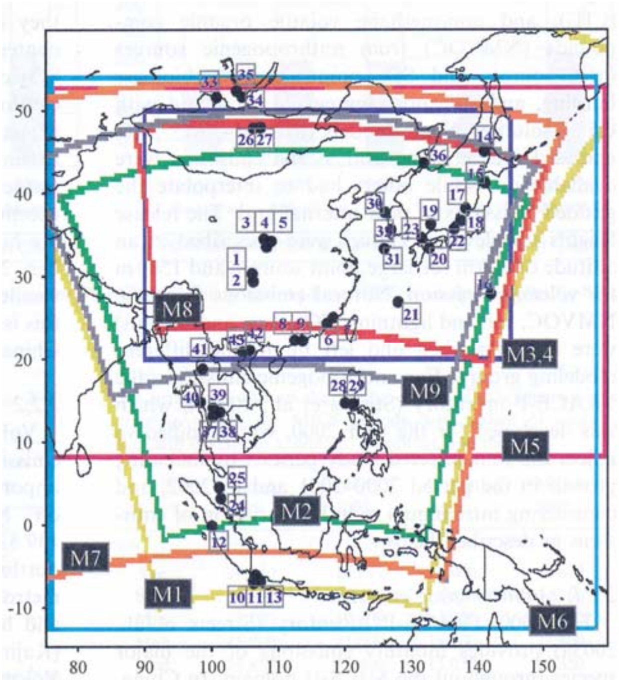


Figure 6.4.1. The MICS-Asia II study model domain and EANET observation sites used in the study. *(Carmichael et al., 2008).*

coarse resolution of the models and resultant inability to distinguish the gradient for rural sites that may be located in or nearby grid cells containing large sulphur sources. Systematically the sites in Russia (sites 33–35) are underpredicted due to the fact that Russian emissions were not included in the used emission inventory. The ensemble mean correlation coefficient over all available sites for the combination of four periods is 0.51. Figure 6.4.3 shows the ensemble mean monthly averaged near-surface sulphate distribution for March 2001. As shown, mean concentrations of sulphate are high near Chongqing, Shanghai and Seoul. It should be noted that the high sulphate concentration in the South of main island of Japan is due to volcanic plume of Mt. Miyake which erupted in 2000 and was still active in 2001 (Carmichael *et al.*, 2008).

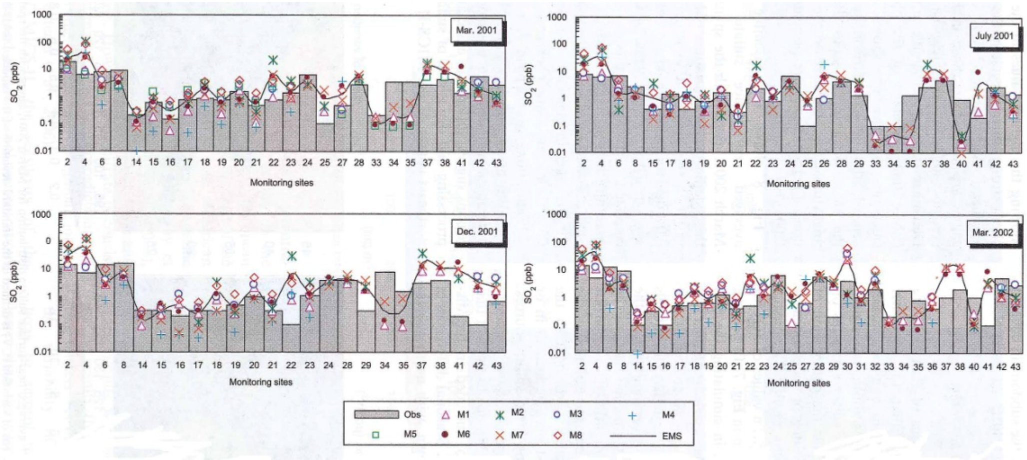


Figure 6.4.2. Predicted and observed monthly mean near surface SO_2 concentrations. Solid line is the ensemble mean prediction. *(Carmichael et al., 2008)*

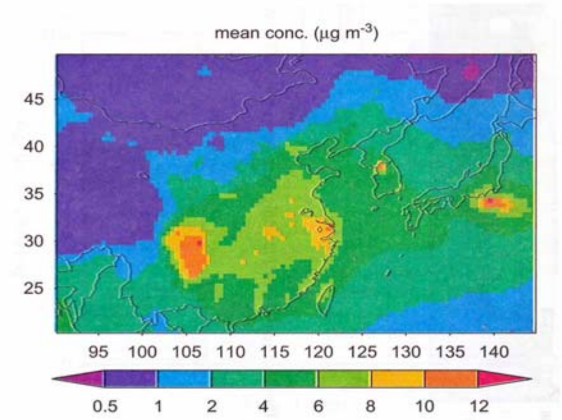


Figure 6.4.3. The ensemble mean monthly averaged near-surface distribution of SO_2 for March 2001. *(Carmichael et al., 2008).*

Depositions are also predicted in MICS-II studies. The comparison of the monthly mean predicted and measured sulphate (SO_4^{2-}) wet deposition for March and July 2001 is shown in Figure 6.4.4 (Carmichael *et al.*, 2008). As shown, although the ensemble mean values at these sites are consistent with observations with a correlation coefficient of 0.73, and 0.47 for March and July, there can be large differences among the models, much larger differences than for SO_2 shown in Figure 6.4.2. The ensemble mean monthly mean total sulphur deposition amounts as sulphate in March and July 2001 are shown in Figure 6.4.5. Wet deposition of sulphate and dry deposition of SO_2 are the two dominant modes of sulphur deposition. The total deposition of sulphur mainly occurs in the Southern China, the Western Pacific and Japan in spring. In summer, high deposition regions move northward, where a deposition center is located in the Northern China and another center in the Southwestern part of China.

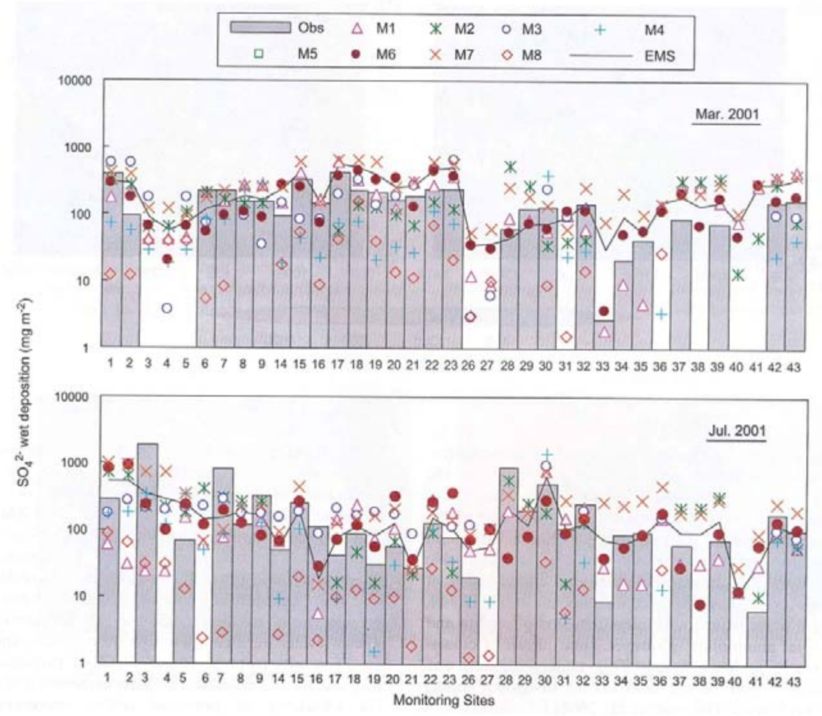


Figure 6.4.4. The predicted and observed monthly mean sulphate wet deposition amounts (as sulphate) at various sites. The solid line shows the ensemble mean prediction. *(Carmichael et al., 2008).*

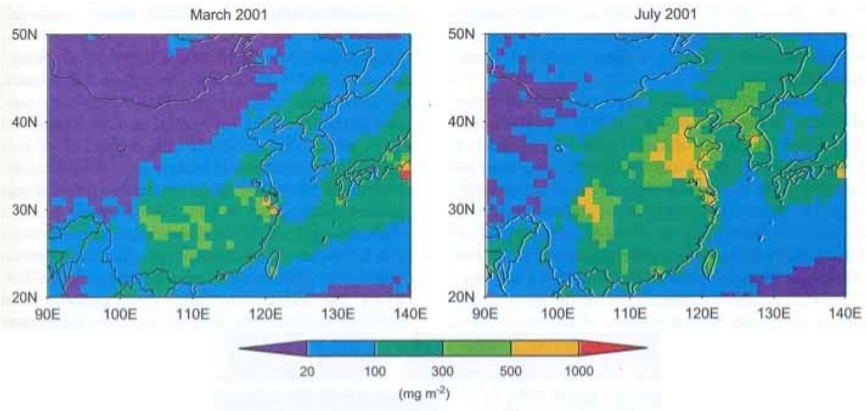


Figure 6.4.5. The ensemble mean monthly mean total sulphur deposition amounts (mg.m^{-3} as sulphate) in March and July 2001. *(Carmichael et al., 2008).*

The total nitrate (NO_3^-) and ammonium (NH_4^+) deposition amounts were also simulated and compared among models and with observation (Wang *et al.*, 2008a). The model prediction of wet deposition of NO_3^- scatters more widely than in the case of sulphate. The results indicate that the ensemble average reproduces observed deposition values better than any single model, but the correlation coefficient between ensemble mean and observation is rather low, 0.41, and 0.36 in March and July 2001, respectively. In contrast, ensemble mean of wet deposition of NH_4^+ shows good agreement with observation with a correlation coefficient of 0.59 for March and 0.61 for July.

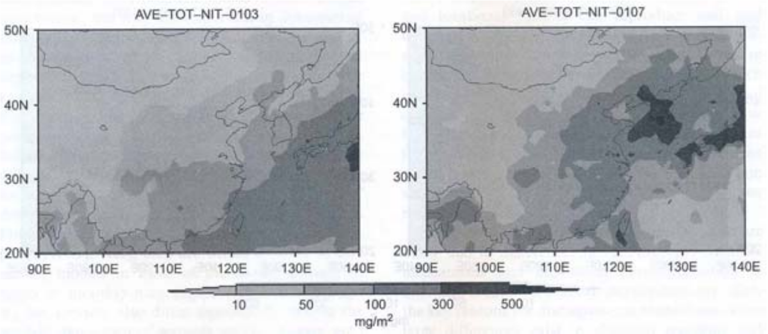


Figure 6.4.6. Ensemble mean model simulation of regional distribution of total nitrate deposition for March (left) and July (right) 2001. *(Wang et al. 2008ac).*

Total deposition of nitrate includes dry deposition of HNO_3 and nitrate aerosol, wet deposition of NO_3^-, but the deposition is dominated by dry deposition of HNO_3 and wet deposition of NO_3^-. Total deposition of ammonium includes dry deposition of NH_3 and wet deposition of NH_4^+. Figures 6.4.6 and 6.4.7 show the ensemble mean model simulation of regional distribution of total nitrate and ammonium deposition, respectively, for March and July 2001 (Wang *et al.* 2008c). For nitrate in spring, high wet deposition is located in Japan, the Southeast of China, part of the Northeast of China, and Korea. In summer, high wet deposition center of NO_3^- move northward following the moving motion of the rain belt in China, and also toward a part of the Southeast Asia since it is wet season in this region. It has been pointed out that the results of eight models have large discrepancies in simulating dry deposition of HNO_3 both in distributions and in amounts. It is reported that the distribution of simulated ammonium dry deposition of NH_3 and wet deposition of NH_4^+ are similar among models in all the seasons.

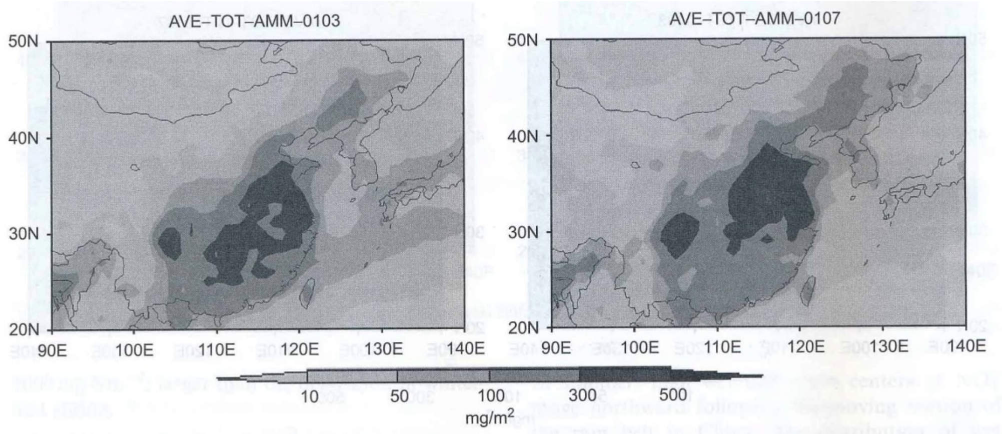


Figure 6.4.7. Ensemble mean model simulation of regional distribution of total ammonium deposition for March (left) and July (right) 2001. *(Wang et al. 2008c).*

The simulation of O_3 was also included in MICS-II. Regional models are being actively used to study O_3 in Asia as introduced below later, and MICS-II offered a valuable opportunity to compare these analysis tools. It is important to note that there are significantly fewer measurement sites for O_3 in EANET than for other species. The comparison of predicted and observed monthly mean O_3 is presented in Figure 6.4.8 (Carmichael *et al.*, 2008). In general, the ensemble of predictions captures the observed variations within the region with the correlation coefficient of 0.62 (Han *et al.*, 2008) for all the studied four months average.

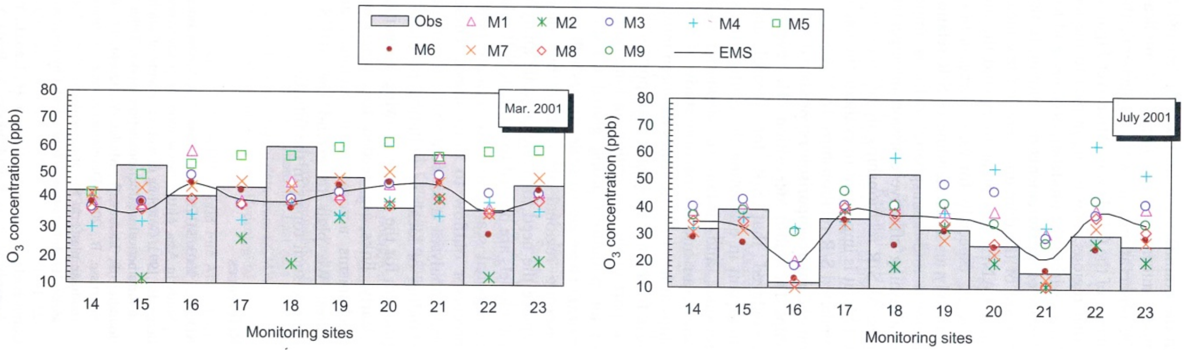


Figure 6.4.8. The predicted and observed monthly mean near surface ozone concentrations over Japan in March (left) and July (right) 2001. *The solid line is ensemble mean predictions. (Carmichael et al., 2008).*

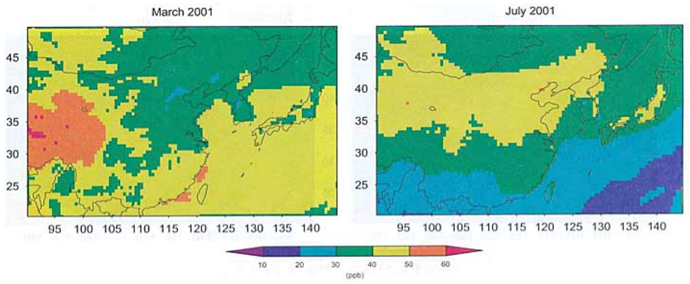


Figure 6.4.9. The ensemble mean surface monthly mean ozone concentrations. *(Carmichael et al., 2008).*

although the predicted variations tend to be lower than that observed. Figure 6.4.9 shows the spatial distribution of ensemble mean surface O_3 (Han *et al*., 2008). In March the mean distribution show a pronounced peak over Tibetan Plateau possibly reflecting stratospheric component of O_3, although no validation has been made due to the lack of observational data. In July, a region of O_3 in 40–50 ppb stretches across Northern China and Japan.

6.4. 2 Sulphur, nitrogen and acids in East Asia

6.4.2.1 Spatial distribution

In the ACCENT IPCC-AR4 modeling experiment, 26 global CTMs (chemical transport models) joined to calculate spatial distributions of sulphur and nitrogen deposition on regional and global scales for the reference year 2000 (Dentener *et al*., 2006). Early EANET data were used for the model evaluation with observation in the Northeast and Southeast Asia. Figure 6.4.10 shows an intercomparison of NOy (=NO + NO_2 + HNO_3 + NO_3^-) total deposition in East Asia (20–40°N; 110–130°E) for 24 models together with mean and median. Left bars denote over land and right bars denote over ocean. The total deposition over land shows larger variability (σ = 20%) than over sea (σ = 11%), and the total deposition taken from single model may differ by up to 30% from mean model. Figure 6.4.11 depicts calculated ensemble mean global distribution of annual dry deposition for NO_3^- (HNO_3 and aerosol nitrate), NHx (NH_3 and NH_4^+) and SO_4^{2-} wet deposition overlapped with observational data by circles. The relative importance of nitrogen and sulphur deposition in EANET region can be seen on global perspective. It is claimed that nearly all the information on wet deposition shows a good agreement with observations in Europe and North America, but models systematically underestimate NO_3^- and SO_4^{2-} deposition in East Asia.

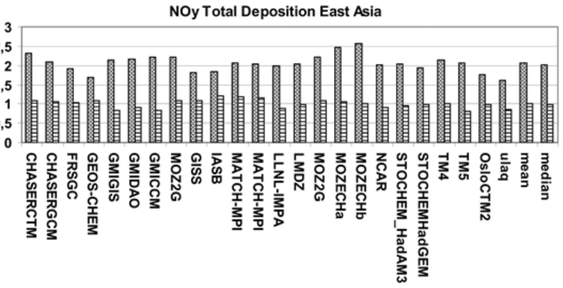


Figure 6.4.10. Intercomparison of annual NOy deposition budget (Tg (N) yr^{-1}) over East Asia (20 – 40°N; 110 – 130°E). Left bars denote land and right bars denote ocean. *(Dentener et al.. 2006).*

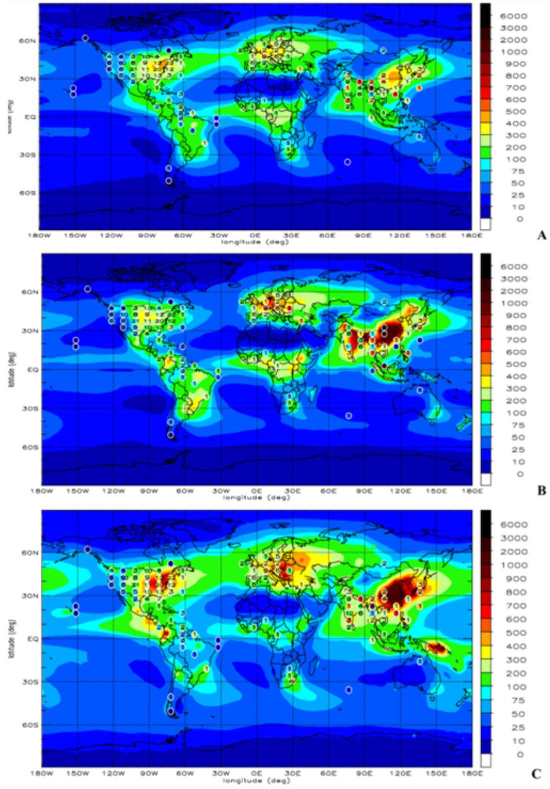


Figure 6.4.11. Global distribution of annual (A) NO_3^- dry deposition, (B) NHx wet deposition and (C) SO_4^{2-} wet deposition in 2000 with measurements grouped in 5° latitude and 10° longitude. *The numbers within the circles indicate the number of stations in this latitude/longitude band. (Dentener et al., 2006).*

Spatial distributions of wet deposition of nss-SO_4^{2-}, NO_3^-, and NH_4^+ in most of EANET region in 2001 have been presented by Lin *et al*. (2008a) using an MM5 (Mesoscale Meteorological model)/CMAQ (The Community Multi-scale Air Quality) regional model. Figure 6.4.12 depicts the calculated annual mean distribution of these loadings together with measurements as observed by the EANET network, and a scatter plot between the model and observation. The scatter plot demonstrates that the majority of Chinese and Japanese sites fall within the reference line for a factor of 2. On annual average the spatial variations and magnitudes of sulphate and ammonium deposition have been well reproduced by the model. Regarding the wet deposition of nitrate, however, there is a significant underestimation at all EANET sites, which is the similar tendency with the global model experiment described above. Several possible causes for the underestimation of the model have been discussed in the paper. In addition to the possible underestimation of NOx emissions in some parts of East Asia, some uncertainties in the model treatment including thermodynamic equilibrium between gaseous HNO_3 and aerosol nitrate, aqueous-phase chemistry and some heterogeneous reactions are suggested. In general model evaluation for nitrogen chemistry finds more difficulty compared with sulphur chemistry and deposition at this stage.

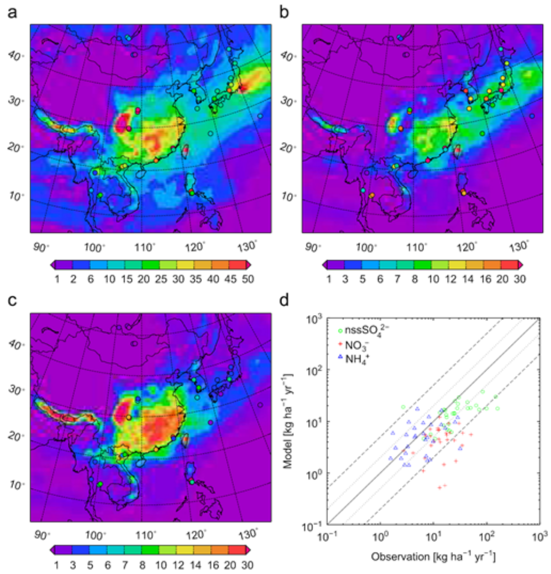


Figure 6.4.12. Annual wet deposition loads of (a) nss-SO$_4^{2-}$, (b) NO$_3^-$, and (c) NH$_4^+$, and a scatter plot between model calculation and EANET observation in 2001. *(Lin et al., 2008a).*

General characteristics of high wet deposition distribution of sulphate and nitrate in China from Sichuan basin toward middle and lower reaches of Yangtze River is well represented by the model agreeing with the spatial patterns of wet deposition synthesized by observational data from National Acid Deposition Monitoring Network (NADMN) in China (Lue and Tian, 2007). Note that very high wet deposition of nss-SO$_4^{2-}$ in the South of Tokyo metropolitan area of Japan is due to volcanic emission of Mt. Miyake that was erupted in 2000 and continued in 2001. The continental outflow of Asian pollutants is clearly depicted.

Han *et al.* (2006) simulated acidifying wet deposition in East Asia during December 2001 by a MM5/RACM model also fully utilizing EANET data for comparison. Precipitation amount and concentrations of nss-SO$_4^{2-}$, NO$_3^-$, NH$_4^+$, and nss-Ca^{2+} were well reproduced in general within a factor of 2 during the period with some exception of over-prediction and under-prediction. As for nss-Ca^{2+}, neglect of anthropogenic sources in the model was thought to give under-prediction at urban sites.

Lin *et al*. (2008a) noted that the model predicts concentrations in the Southeast Asia less accurately, that is related to larger uncertainty in the emission estimates and simulated precipitation fields. Modeling studies on sulphur and nitrogen focusing on the Southeast Asia are so far very scares. Siniarovina and Engardt (2005) applied MATCH (The Multi-scale Atmospheric Transport and Chemistry) model driven by meteorological data from ECMWF (The European Center for Medium range Weather Forecasting) to a model domain covering EANET region of the Southeast Asia including Southern Hemispheric part of Indonesia over the full year of 2000. It is of utmost importance to drive the model with correct precipitation field particularly in the Southeast Asia since previous studies identified that wet deposition as the most important removal mechanism for sulphur in this region (Ayers, *et al*, 2000, Engardt and Leong, 2001). They found that there is considerable scatter around 1:1 line in the scatter plot of precipitation amount but there is no large systematic difference, and the wet deposition scheme is quite sensitive to precipitation.

Figures 6.4.13 (a) – (c) show model domain with sulphur emissions in 0.5°× 0.5° resolution, modeled monthly mean near surface SO_2 mixing ratio during December and total (wet + dry) deposition of sulphur for the year 2000, respectively (Siniarovina and Engardt, 2005).

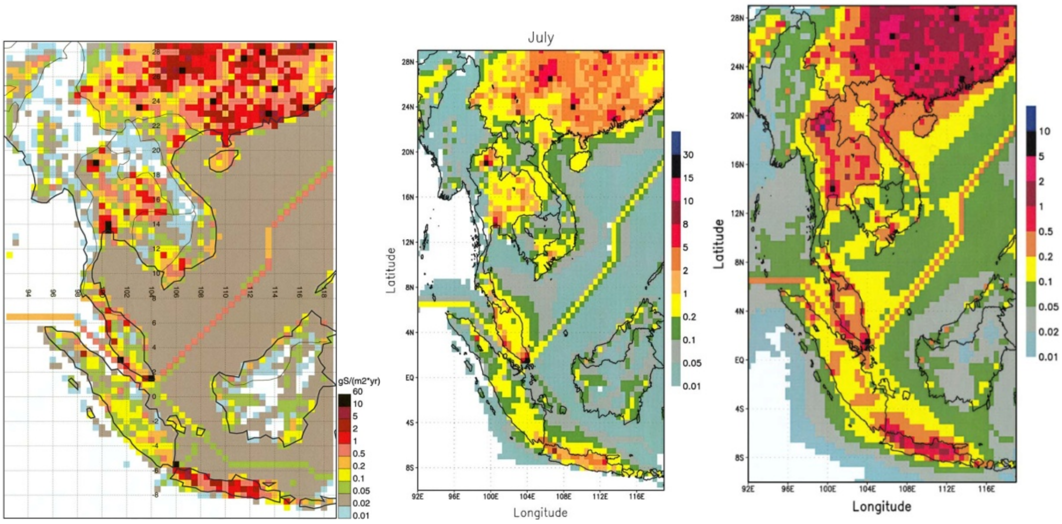


Figure 6.4.13 (a) Model domain and SO_2 emissions in 2000 from

(b) Modelled monthly mean SO_2 mixing ratio during July 2000. *Units: ppbv.*

(c) Annual mean total (wet and dry) deposition of sulphur in 2000. *Units: gS* m^{-2} yr^{-1}.

In July, emissions of SO_2 from Bangkok area (Figure. 6.4.13 (a)) are spread over most of central Thailand and similarly the emissions from Northern Thailand affect Laos as seen in Figure 6.4.13 (b). It should be noted, however, the sulphur emissions from Northern Thailand stems mostly from the power plant in Mae Moh, which has recently significantly reduced its emissions (Iverfeldt *et al.*, 2003). In Peninsula Malaysia, the modeled concentration of SO_2 in July spreads over the Peninsula due to Southwest monsoon, and also the emissions from Singapore tend to affect Southern Malaysia. As shown in Figure 6.4.13 (c), high total depositions of sulphur (> 1gS m^{-2} yr^{-1}) are found in Southern China, Northern Vietnam, parts of Java, and around major cities in Thailand, Malaysia and Singapore. Comparison of the modeled atmospheric concentration of SO_2 and SO_4^{2-} concentration in precipitation with the observational data of EANET and others showed in general much poorer agreement than in the Northeast Asia. In addition to the model uncertainty in the reproduction of precipitation, the uncertainty of some of observed values of SO_2 either by a filter pack or passive sampler in this region has also noted (Siniarovina and Engardt, 2005).

6.4.2.2 Trends

There has been published several papers showing the modeling analysis of atmospheric concentration and deposition trends of sulphur and nitrogen comparing with EANET monitoring data in the Northeast Asia after 2001, and discussing the correlation with emission trends in this region. Lu *et al*. (2010) showed that the trends of SO_2 concentration and acid rain pH and frequency in China as well as the increasing trends of background SO_2 and sulphate concentration in the Northeast Asia is consistent with the estimated emission in China. From 2000 to 2006 total SO_2 emission in China is estimated to have increased by 53% at an annual growth rate of 7.3%. Figure 6.4.14a presents a comparison of inter-annual trends of normalized SO_2 emissions and the simulated total sulphur concentrations (SO_2 + SO_4^{2-}) averaged over China. In general, the simulated total sulphur concentrations mirror the changing SO_2 emissions in China. As seen in the figure, the total sulphur increased from 2001 to 2006, leveled off in 2007, and dropped down dramatically in 2008. As shown in Figure 6.4.14 (b), it is noted that from 2000 to 2006 the SO_2 emission in North China increased faster than that in South China but the Northern emission decreased significantly in 2008 possibly preparing for the Olympic Games, which is reflected in the dropdown over China in Figure 6.4.14 (a).

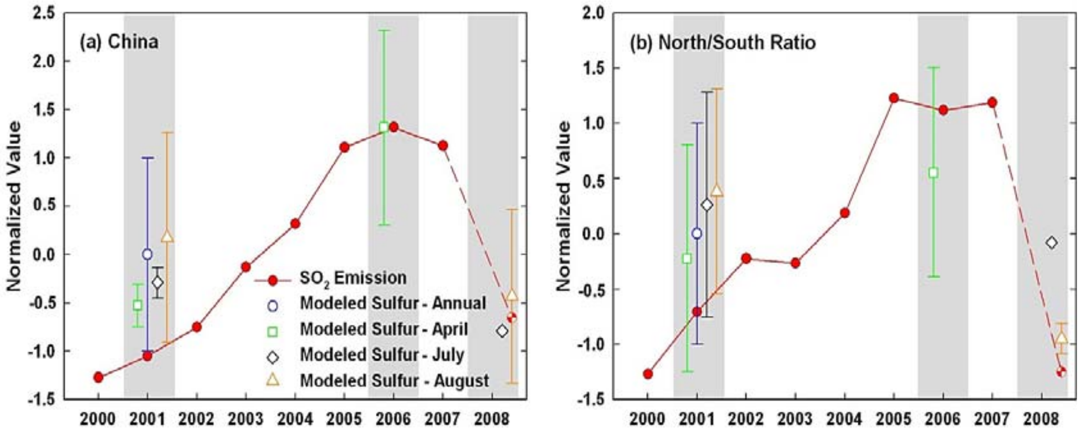


Figure 6.4.14. (a) Inter-annual trends of normalized SO_2 emissions and simulated surface sulphur (SO_2+SO_4^{2-}) concentrations over China and (b) ratios between north and South China. ***(Lu et al. 2010).***

Based on EANET monitoring data, Lu *et al*. (2010) also showed that average concentrations of SO_2 and SO_4^{2-} (in PM) at all sites in Korea and Japan increased by 15.0% and 24.1% from 2003 to 2007 in line with the SO_2 emission change in China by 21.0%. It was pointed out that their annual mean concentrations at each site are strongly affected by inter-annual variability in meteorology. They demonstrated that the normalized inter-annual variability of SO_2 concentrations in Ganghwa site in South Korea and Yusuhara site in Japan could be well simulated by the STEM model.

As for nitrogen, Morino *et al.* (2011) analyzed results of CMAQ model and observational data (nationwide monitoring database and EANET database) over Japan during 1989–2008. Figure 6.4.15 depicts temporal variations in annual averaged wet deposition rates of NO_3^- and NH_4^+ and precipitation rates in the Northeastern Japan (NEJ), central Japan (CJ), and the Southwestern Japan (SWJ). The model successfully reproduced the general pattern of spatial and temporal variations of observed NO_3^- wet deposition rates although NH_4^+ wet deposition rates were generally overestimated whereas precipitation rates were underestimated particularly NEJ and CJ, which may be reflected in the underestimate of NO_3^- wet deposition rates in these regions. Wet deposition rates of NO_3^- across Japan increased during 1989–2008 with the rates of 2–5%/yr depending on the region. Sensitivity simulations indicated that the increase of NO_3^- wet deposition rates was mostly (61–94%) explained by the increased emissions of NOx in China. Emissions from China accounted for 29–35% of NO_3^- wet deposition across Japan during 1989–1993, and for 43–61% during 2004–2008. Inter-annual variation of NO_3^- wet deposition was further evaluated using a meteorological index. The analysis revealed that simulated inter-annual anomalies of NO_3^- wet deposition rates and precipitation rates correlated well with the observed results.

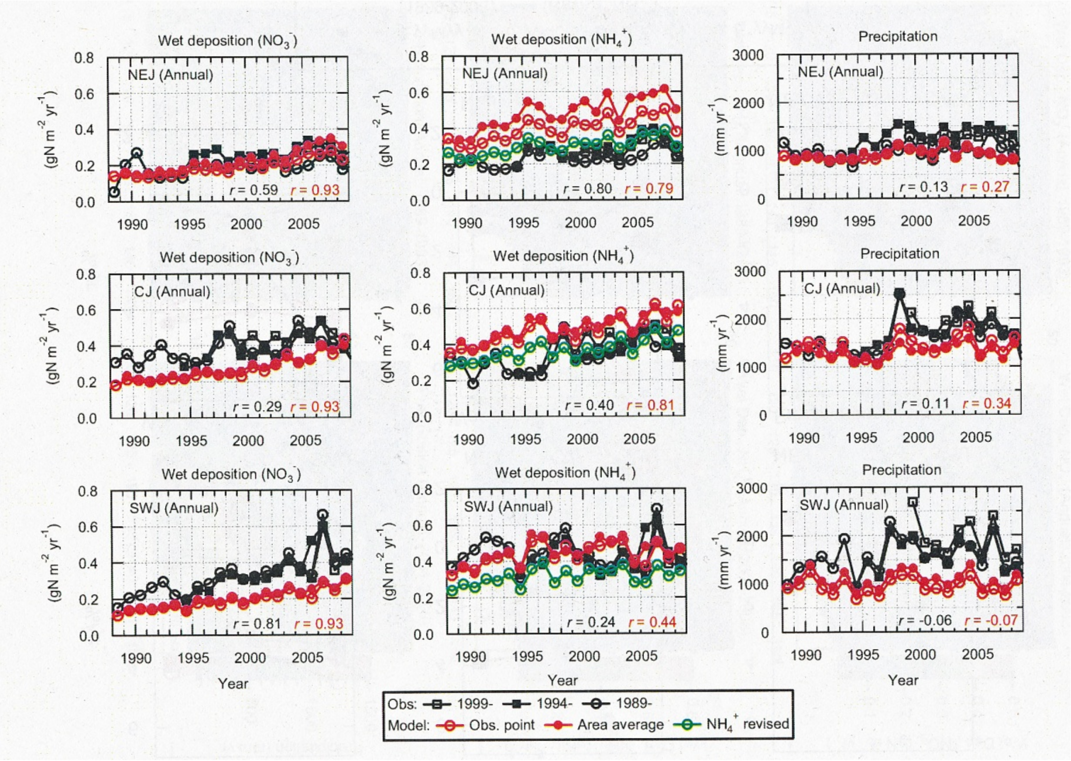


Figure 6.4.15. Temporal variations in annual averaged observed wet deposition rates of NO_3^- and NH_4^+ and precipitation rates in the Northeastern Japan (NEJ), central Japan (CJ) and the Southwestern Japan (SWJ). *(Morino et al. 2011).*

Concerning the interaction between air pollution and climate change, impact of the East Asian summer monsoon on long-term variations in the acidity precipitation in Central China was studied by Ge *et al.* (2011). Based on domestic Acid Rain Monitoring Network data, they could conclude that sulphate and nitrate concentrations in rainwater are increase by 68, and 77% during 1992–2003, respectively. They showed that about 60–80% of the changes in SO_4^{2-} concentration and 70–80% of the changes in NO_3^- concentration, and at least 60% changes in the pH of precipitation in Central China are caused by changes in emissions of SO_2 and NOx in China. In addition to emission change, changes in meteorological factors such as rainfall related the East Asian summer monsoon plays less important but significant role. In high monsoon index years caused higher concentrations of SO_4^{2-} and NO_3^- concentrations in rainwater in Central China.

6.4.2.3 Source-receptor relationships

Model intercomparison study on hemispherical scale intercontinental transport of both gaseous and aerosol species has been organized by the Task Force on Hemispheric Transport of Air Pollution (TF HTAP) (2007) (See 6.6.2). The multi-model mean approach was adopted in the project, since previous studies (e.g., Schulz *et al*., 2006; Stevenson *et al*., 2006; Dentener *et al*., 2006) have shown that mean model results compare better with observations than any individual model. Sanderson *et al*. (2008) reported the intercontinental transport and deposition of oxidized nitrogen obtained in this study. Figure 6.4.16 shows the ensemble mean (15-model) export fractions of NOy ($=NO + NO_2 + NO_3 + HNO_3 + HO_2NO_2 + 2{\times}N_2O_5 +$ organic nitrates and nitrate aerosols) from each region in each season. Between 12 and 24% of the NOx emitted is exported from each region annually. The strongest impact of each source region on a foreign region is: Europe (EU) on East Asia (EA) (2.4%), North America (NA) on Europe (2.1%), South Asia (SA) on East Asia (6.4%), and East Asia on North America (1.3%). Europe exports the most NOy, and East Asia the least. East Asia receives the most NOy from the other regions. Between 8 and 15% of NOx emitted in each region is transported over distances larger than 1000 km, with 3–10% ultimately deposited over the foreign regions. These proportions should be compared with the amounts of NOy exported. For example, SA exports 20% of the NOx emitted, of which 32% is deposited in EA. The scatter in the model results is most likely to be due to differences in the boundary layer and convection schemes used to mix and transport the NOy species, and the associated wet removal processes.

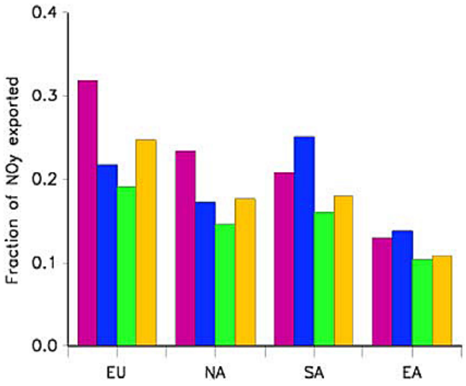


Figure 6.4.16. Ensemble mean export fractions of NOy from each HTAP region. *Winter (pink), Spring (blue), Summer (green), and autumn (yellow). (Sanderson et al., 2008).*

Liu *et al.* (2008a) evaluated source-receptor relationships between East Asian (EA) SO_2 emissions and Northern Hemisphere aerosol sulphate concentrations in the year of 1991 using a global CTM, MOZART-2. Figure 6.4.17 horizontal distribution of EA SO_4^{2-} concentrations ($\mu g.m^{-3}$) at the surface in four seasons. As seen the annual average simulated surface concentrations of EA SO_4^{2-} range from more than 10 $\mu g.m^{-3}$ over EA to less than 0.2 $\mu g.m^{-3}$ over the Western US, and the surface concentrations of SO_4^{2-} from EA sources over the Western US are the highest in March, April and May (MAM) (up to 0.15 $\mu g.m^{-3}$), and the lowest in December, January and February (DJF) (less than 0.06 $\mu g.m^{-3}$).

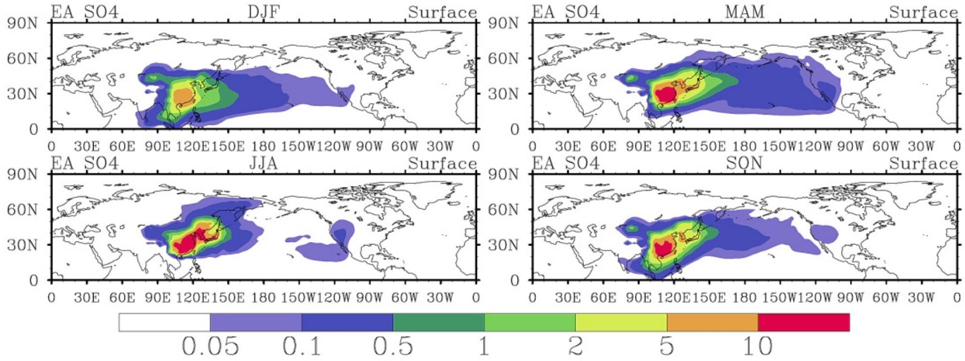


Figure 6.4.17. Horizontal distribution of East Asian aerosol sulphate concentration (unit: $\mu g.m^{-3}$ at the surface in December, January and February (DJF), March, April and May (MAM), June, July and August (JJA), and September, October and November (SON). *(Liu et al. 2008a).*

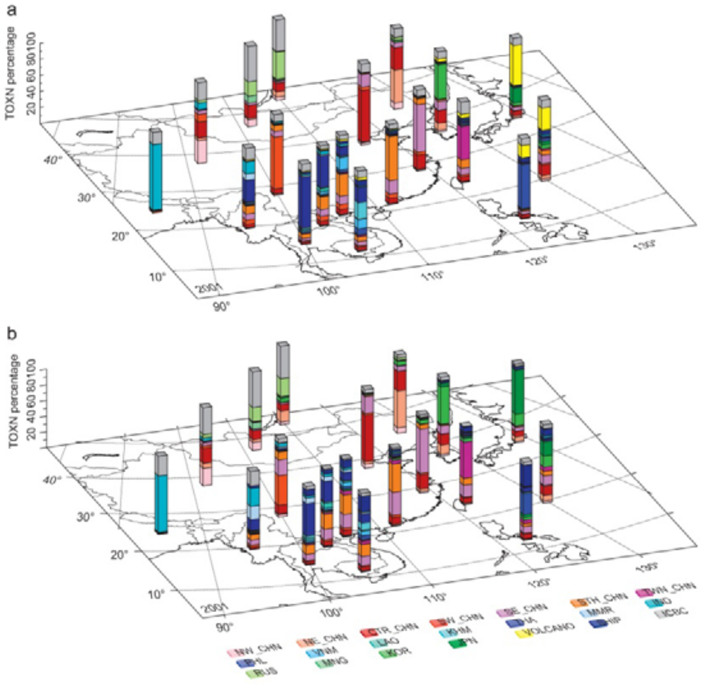


Figure 6.4.18. Region-to-region source-receptor relationship for (a) total sulphur and (b) total reactive nitrogen deposition. Vertical bars show the origin of substances deposited in each "receptor" region. *Values given show percentage of total deposition. (Lin et al., 2008b).*

Lin *et al.* (2008b) applied the state-of-the-art CMAQ model (version 4.5.1) with MM5 for meteorological field to quantify long-range transport of sulphur and reactive nitrogen in East Asia covering most of EANET region and apart of India with 81-km grid spacing. The model was validated against EANET observational data of nss-SO_4^{2-}, NO_3^-, and NH_4^+ in 2001 as mentioned in 6.4.2.1 (Lin *et al.*, 2008a). Figures 6.4.18 (a) and 6.4.18 (b) shows the source-receptor (S/R) relationships of sulphur and nitrogen depositions, respectively. Long range transport from the industrial areas of China contributes a significant percentage (>20%) of anthropogenic sulphur and reactive nitrogen depositions throughout East Asia. At the same time the Northwestern China receives approximately 35% of its sulphur load and 45% of nitrogen load from foreign emissions mainly from the Indian subcontinent. For the receptor areas in the Southeast Asia, except Thailand and parts of India, the dominating sources (>50%) are foreign emissions from Thailand, South China and Southeast China. In addition, the contributions of NOx emissions from international shipping are considerable for nitrogen deposition over Southeast Asia, especially in the Philippines. Within China, local emissions are the dominant source of sulphur deposition in the Southwest and South China, whereas for reactive nitrogen loading over these two areas, the Southeast China sources contribute up to 20–30%. In 2001 contribution of eruption of Miyakejima volcano is overwhelming (>80%) for sulphur deposition in Japan. Excluding the volcanic source, the contributions of sulphur to Japan are estimated to be Japan 37%, China 31%, Korean Peninsula 7%, and other foreign sources18%. For Vietnam, the contributions of sulphur are: Vietnam 11%, China 51%, Thailand 10%, other foreign sources 5%, and international ships 3%. The estimated contributions of nitrogen to Japan are: Japan 56%, China 21%, and Korean Peninsula 15%, and those to Korean Peninsula are: Korean Peninsula 47%, China 39%, and Japan 5%.

6.4.3 Ozone in East Asia

6.4.3.1 Spatial distribution and seasonal variation

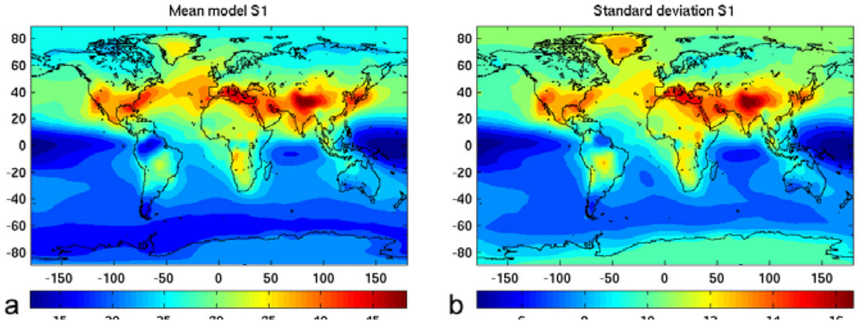


Figure 6.4.19. (a) Model ensemble mean annual-mean surface ozone concentration and standard deviation among models in ppbv. *(Ellingsen, et al., 2006).*

Ensemble mean global distributions of present day surface concentrations of ozone has been calculated by 18 global CTMs from the U.S., Europe, and Japan within "ACCENT Photocomp" experiment, which contributed to IPCC AR4 (Ellingsen *et al.*, 2008). Figure 6.4.19 (a) and (b) shows annual-mean surface ozone concentrations for the model ensemble mean and the standard deviation for the year 2000, respectively. They showed that the computed monthly average concentrations agree relatively well with observational data within 10 ppb in North America, Europe and Northern China. Figure 6.4.19 (a) shows that annual mean values over large industrialized areas in North America, Europe and Asia vary between 40–50 ppbv with 1σ standard deviation of ca. 20–30%. The Northern hemisphere average is found to be 33.7±3.8 ppbv.

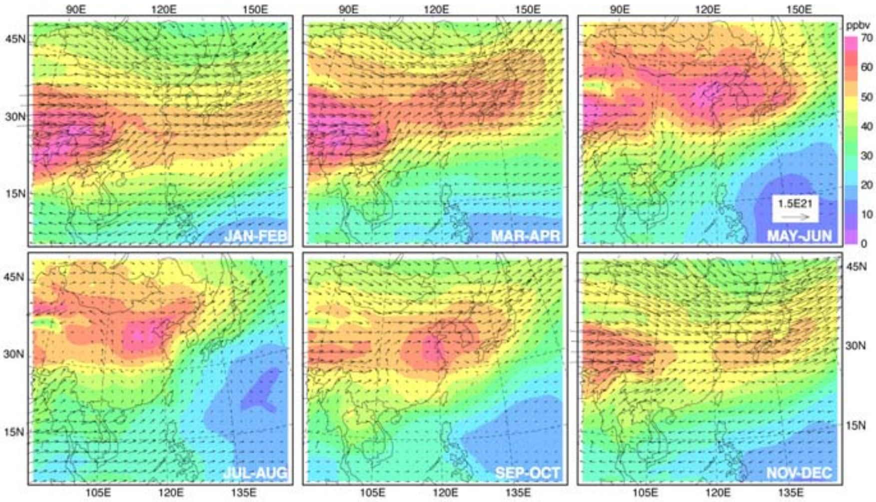


Figure 6.4.20. Comparison between observed (red lines) and simulated (black lines) daily ozone concentration at Japanese sites of EANET during 2002. *Each correlation coefficient between the observed and simulated concentration for every 2 months and full year (yr) is shown within the graph (Yamaji et al, 2007).*

Regional spatial distribution and seasonal variation of surface ozone in East Asia have been simulated by several regional CTM studies. Yamaji *et al*. (2006) successfully reproduced daily variation of EANET surface O_3 data in Japan for a whole year of 2002 by RAMS/CMAQ (v.4.4) as shown in Figure 6.4.20. The obtained correlation is 0.7–0.9 except the Northern sites of Rishiri, Tappi, and Sadoseki, where the coefficient is 0.4–0.6. Using this regional model, fundamental feature of seasonal variation of spatial distribution of 2-month averaged surface O_3 concentration and ozone flux in East Asia is presented in Figure 6.4.21.

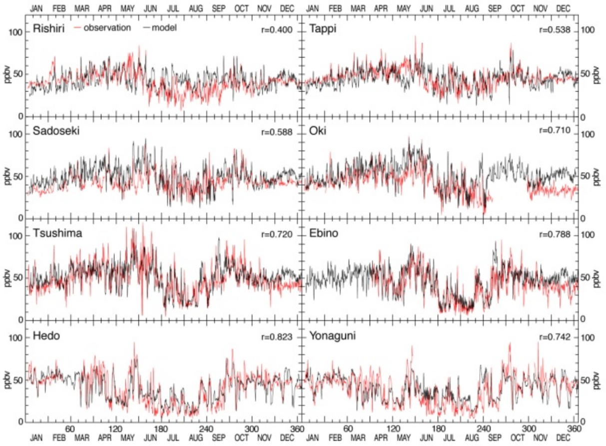


Figure 6.4.21. Spatial distributions of surface ozone concentrations (ppbv) and ozone flux (arrows) in the boundary layer in East Asia. *(Yamaji et al., 2006).*

In the wintertime from November to February, there are large O_3 fluxes due to strong westerlies at around 40°N from Mongolia and Northern China to Japan. The O_3 concentrations in the boundary layer over this area are kept at around 30–50 ppbv which are nearly the same as the Northwestern boundaries because the photochemical O_3 production is suppressed by weak solar radiation in this area. Ozone concentrations are relatively high (50–55 ppbv) in the latitude belt at 20–35°N over South and East China and Pacific Ocean South of Japan as seen in Figure 6.4.21. Northern part of Indochina Peninsula, O_3 concentrations are considerably high during January–April due to continental inflow and biomass burning emission during dry season (Pochanart *et al.*, 2003). Very high O_3 concentration near the Western boundary is due to the high altitude and has not been validated. In spring high O_3 continental outflow cover Korean Peninsula and Japan (March–June), and the concentration of O_3 from the Eastern China to Japan is 55–60 ppbv giving so called spring maximum at around 35–40°N agreeing with observation (Tanimoto *et al.* 2005).

In July–August, the Southern part of Korean Peninsula, Japan, and a coastal part of Southern China are under the effect of marine air mass, and thus O_3 concentrations decrease to 25–40 ppbv. Also, continental Southeast Asia is in the wet season under the influence of marine air and O_3 concentration is in general very low. On the other hand, except the coastal area the Eastern China is still covered with air mass of high O_3 concentrations of 60–70 ppbv due to active photochemical production, where the “cleaner” marine air masses do not reach. In autumn from September to October, the distribution of O_3 is similar to spring, when Japan and Korean Peninsula are influenced by continental air masses to give secondary O_3 peak.

Further model analysis of seasonal cycle of surface O_3 over East Asia has also been reported by Tanimoto *et al.* (2005), Li *et al.* (2007) and Lin *et al.* (2009). Comparison of seasonal cycle of surface O_3 concentrations in monthly averaged values between observation at EANET sites and simulation with other regional CTM (NAQPMS) with 81 km resolution are depicted in Figure

6.4.22.

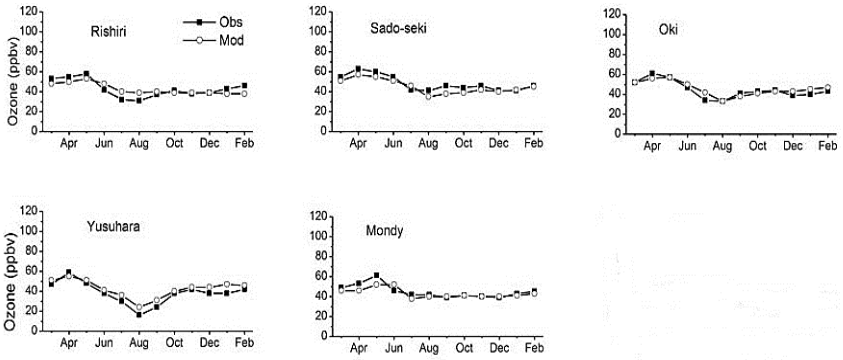


Figure 6.4.22. Observed (solid squares) and simulated (open circles) mean monthly ozone concentrations (ppbv) at selected EANET sites. *(adapted from Li et al., 2007).*

6.4.3.2 Long-term trend and inter-annual variation

Decadal trends of springtime surface ozone mainly based on EAENT observation at Japanese sites have been analyzed by Tanimoto *et al*. (2009). Regional CTM, RAMS/CMAQ with Asian emission database of REAS was used to explore how changed in Asian anthropogenic emissions have contributed to the observed trend. Figure 6.4.23 shows the comparison of trends of springtime

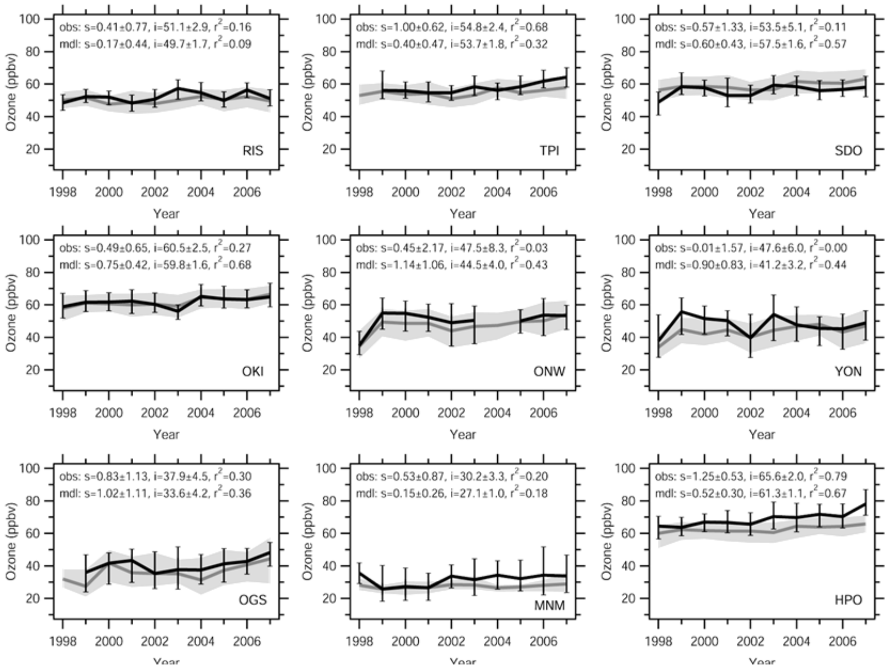


Figure 6.4.23. Trends of observed (black) and model simulated (grey) springtime surface ozone from 1998 to 2007 at EANET and WMO/GAW sites over Japan. *RIS, Rishiri; TPI, Tappi; SDO, Sadoseki; OKI, Oki; ONW, Okinawa/Hedo); YON, Yonagunijima (WMO/GAW); OGS, Ogasawara; MNM, Minamitorishima (WMO/GAW); HPO, Happo. (Tanimoto et al., 2009).*

surface ozone from between 1998–2007 between the observation and model simulation. As shown in Figure 6.4.24, except the mountainous site at Happo, six sea-level EANET sites all shows increasing trends between 0.41 to 1.00 ppbv yr^{-1} (average 0.63 ppbv yr^{-1}, 1.2% yr^{-1}) in observation, and 0.17 to 1.14 ppbv yr^{-1} (average 0.70 ppbv yr^{-1}, 1.4% yr^{-1}). Although the observed increasing trends are in the same range of model simulated increasing trends due to the increase of anthropogenic emissions in East Asia, large inter-annual variability obscure the long-term trend not being statistically significant. In contrast, the increasing trend at the mountainous EANET site, Happo (1,850 m altitude), shows statistically significant increase of 1.0 ppb yr^{-1} as the median trend (Tanimoto, 2009) for 1998–2006. However, it is noted that about half of the observed O_3 increase cannot be explained by the model simulation using the assigned increase of anthropogenic emissions from East Asia (Tanimoto *et al.*, 2009). Influence of meteorological variability on interannual variation is an important issue for extracting trends of atmospheric concentrations of any atmospheric species since year-to-year variations are usually much larger than the annual rates of increase of the long-term trend. Kurokawa *et al.*, (2009) analyzed the interannual variability of springtime boundary layer ozone (oxidant) over the Western and central Japan during 1981–2005 using the CMAQ model with REAS emission inventory, and successfully reproduced the observed increasing trend by the model. Figure 6.4.24 shows Time series of anomalies of springtime ozone (oxidant) (blue dots) and simulated BL O_3 using each-year's meteorology and emission ($E_{yy}M_{yy}$, black line) and each year's meteorology but fixed emission in 2000 ($E_{00}M_{yy}$, red line) over the Western and central Japan. Here anomalies are defined as deviations from the values averaged over 1985–2005. Observational data are taken from continuous measurements of general air quality monitoring sites in Japan. As shown in Figure 6.4.24, year-to-year variations are well reproduced by $E_{yy}M_{yy}$ simulation, and the observed increasing trends of anomalies during the period, 0.37 ppbv/yr is also well reproduced by the simulation, 0.42 ppbv/yr. In contrast, simulation with fixed emission in 2000 reproduces the interannual variability but not the increasing trend showing clearly the increasing trend is due to the increasing of emission in East Asia. The main cause of the anomaly was found to be related to surface pressure anomaly over the Pacific Ocean the East of Japan. When the anomaly has a large negative value, polluted air mass from continental Asia tend to be transported directly to Japan be westerly winds.

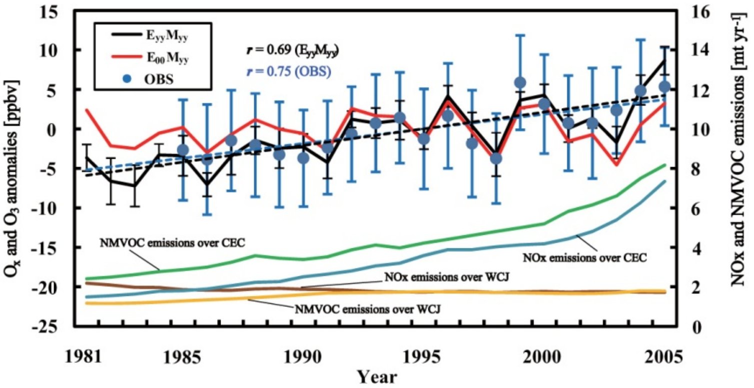


Figure 6.4.24. Time series of anomalies of springtime ozone (oxidant) (blue dots) and simulated BL O3 using each-year's meteorology and emission ($E_{yy}M_{yy}$, black line) and each year's meteorology but fixed emission in 2000 ($E_{00}M_{yy}$, red line) over West and central Japan. *The blue and black dashed lines are liner regression lines for observation and simulation by $E_{yy}M_{yy}$. (Kurokawa et al., 2009).*

6.4.3.3 Source-receptor relationships

The source-receptor relationship for surface O_3 in East Asia during the early 2000s (2000–2005) has been analyzed using a tagged tracer method with a global chemical transport model, CHASER [CHemical AGCM (atmospheric general circulation model) for Studies of atmospheric Environment and Radiative forcing] (Nagashima *et al*., 2010). Using a global CTM rather than regional model can provide global perspective of source assignment of ozone including stratospheric origin and inter-continental transport giving a closure of total contribution summing up to 100%. The horizontal resolution was 1.9° by 1.9° and emission database of REAS for Asian region and EDGAR for the rest of the world has been used. The source region was separated into 45 regions including 22 regions each in the planetary boundary layer (PBL) and the free troposphere (FT), and the stratosphere (STR). Figure 6.4.25 shows the six-year averaged seasonal mean relative contribution (%) from source regions to receptor area in East Asia for spring and summer. In spring, as shown in Figure 6.4.26, domestic contribution of O_3 account for about 15, 40, 60, 50% in Northeast (CH-NE), North Central Plain (CH-NCP), Yanze River Basin (CH-YZB) and the Southeastern China (CH-SE), about 20% in Korean Peninsula (KOR) and central Japan (JPN-E+W). In the CH-NE and CH-SE, large contribution of nearly 20% from East Siberia (ESB) and the Indochina Peninsula (IDC+) were identified. The contributions of China to Korea and Japan were estimated 20, and 12 %. The contribution of intercontinental transport (RMT) increased with latitude: 21% in CH-NE and 13–14% in CH-NCP, KOR and JPN-E+W. The contributions from the stratosphere also increase with latitude: about 25% in CH-NE, 20% in CH-NCP, KOR and JPN, and less than 15% in CH-YRB and CH-SE. It should be noted that the contributions from free troposphere (FT), which account 10–15% in all areas, cannot be assigned to each source region of ozone precursor emissions according to this methodology. In summer although the contribution of intra-continental trans-boundary transport was still significant, the domestic contribution increased greatly and inter-continental transport and stratosphere decreased greatly. It was also shown in the study that as for the hourly mean concentrations of surface O_3, domestically created O_3 accounted more than 50% in the very high concentration range of >90 ppbv even in spring, and the relative impact of foreign sources were identified as substantial in the mid-high concentration range of 60–90 ppb.

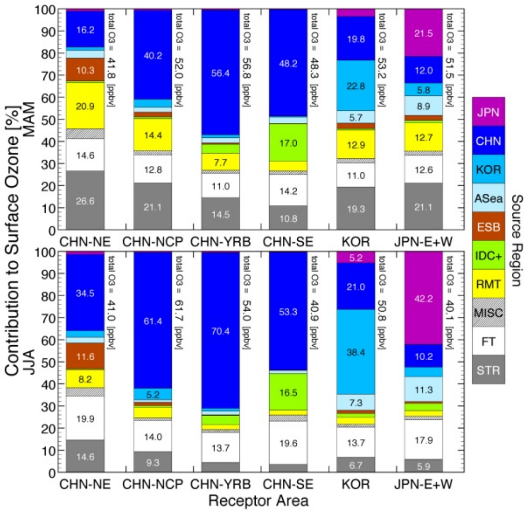


Figure 6.4.25. Six-year (200-2005) averaged seasonal mean relative contribution (%) from source regions to receptor area in East Asia for spring (top) and summer (bottom). *For the source regions: ASea (Adjacent Sea) is the sum of NPC, JPS, and ECS; IDC+ is the sum of IDC and IND, and RMT is the sum of AMN, EUR, CAS, MES, and NAT. For the receptor areas: JPN-E+W is the sum of JPN-E and JPN-W, and KOR is the sum of KOR-N and KOR-S. (Nagashima et al., 2010)*

6.5 Ecosystem impact assessment studies

6.5.1 Biogeochemical models

Ecosystem is a complex and dynamic system where nutrients are continually cycling through biotic and abiotic processes. Though the magnitude and feature of the nutrient cycling are varying depending on the meteorological, edaphic and vegetative conditions, the rate of internal cycle is generally much larger than the rate of atmospheric deposition. The effect of atmospheric deposition to the ecosystem is therefore long-term phenomena and long-term observation would be necessary to detect the changes in material flows or changes in water and soil properties in an ecosystem. In addition to the long-term monitoring of various parameters in catchment areas, biogeochemical models are expected to be useful to understand the mechanisms of the changes in ecosystem structure and functions by acidic deposition and to predict their future status under the scenarios on the atmospheric deposition induced by the changes in social and economical situation.

Considering from a view point of their origin and the processes emphasized on, the biogeochemical models are classified into three groups: originated from soil chemical process model, soil biological process model and plant circulation model.

The early stage models for estimating the effects of atmospheric deposition mainly focused on the acidification of soil, groundwater, lakes and stream water, based on the hydrological process and chemical processes in soil such as cation exchange, anion adsorption, aluminum hydrolysis, solute equilibrium processes and so on. Some models were improved or newly designed to include biogeochemical cycles due to biological processes occurring in the forest canopy, litter layer and root zone. The various models were reviewed for their objectives, processes included, time scale and spatial resolution etc. (Ball and Trudgill, 1995). ILWAS (Integrated Lake Water Acidification Study) (Goldstein *et al.*, 1985), NUCSAM (Nutrient Cycling and Soil Acidification Model) and RESAM (Regional soil acidification model) (de Vries *et al.*, 1998) are the most complex models including the most diverse processes among these kind of models (Table 6.5.1).

Table 6.5.1. The Major processes used in NUCSAM and RESAM

Hydrological processes	Multi-layer soil compartment with vertical flow, Root water uptake, Evapotranspiration
Soil chemical processes	CO_2 dissociation, Organic acid dissociation/protonation, Carbonate weathering, Silicate weathering, Al Hydroxide weathering, Cation exchange, Sulphate adsorption, Phosphate adsorption*, Complication reaction*
Soil biological processes	Organic matter mineralization / immobilization, Nitrification, Denitrification
Plant related processes	Root uptake, Foliar uptake, Reallocation, Foliar exudation, Litterfall, Root decay

**: NUCSAM only.*

Soil biological process model also has a long history, which considers carbon and nitrogen cycle in soil due to decomposition and mineralization processes of plant litter and soil organic matter. Many models assume several compartments with different decomposition rate for litter and soil organic matter, from rapid decomposable to recalcitrant (Molina and Smith, 1998) and estimate the carbon and nitrogen accumulation in soil and mineralization as an indicator of soil productivity. Some models were modified to evaluate the biogeochemical cycle in forest ecosystems by taking C and N cycle in the above ground vegetation into consideration. For example, CENTURY model originally developed to estimate the organic matter formation and mineralization in soil now have the processes of biogeochemical cycle of carbon, nitrogen, phosphorus and sulphur in forest ecosystems, as well as in the cropland and grassland ecosystems (Kirschbaum and Paul, 2002; Peng *et al.*, 1998; http://www.nrel.colostate.edu/projects/century/).

There are many plant circulation models such as FOREST-BGC (Running and Coughlan, 1988; Running and Cower, 1991), PnET-CN (Lumped Parameter model of Carbon (C), Water, and Nitrogen (N) interactions in forest ecosystems) (Aber *et al*., 1997) which simulate the carbon and nitrogen cycle by taking photosynthesis, respiration, allocation, litterfall production processes etc., based on the meteorological data, and are coupled with the soil biological processes. These models generally consider only carbon and nitrogen cycle and have been used to evaluate the effects of climate change on these cycles. PnET model was modified to an integrated biogeochemical model (PnET-BGC), which included the soil chemical processes similar to those shown in Table 6.5.1 (Gbondo-Tugbawa *et al*., 1991), and it has been used to evaluate the effect of atmospheric deposition to soil and water chemistry too (Chen and Driscoll, 2005).

These models, however, especially those with plant circulation processes were developed for the temperate or boreal forest in Europe and North America and have been rarely applied to Asian ecosystems. It is necessary to investigate if these models are applicable to Asian ecosystems where soil, vegetation and meteorological condition are different from those supposed by the model. For many of these models, investigation on the applicability requires large input and validation data and data for specifying suitable values to the parameters. Following are the examples of model application to Japan and of acquisition of model parameters. In order to get the realistic and adequate results by the biogeochemical models, it is inevitable to cooperate with the integrated measurements of material flows from atmospheric deposition to stream water export.

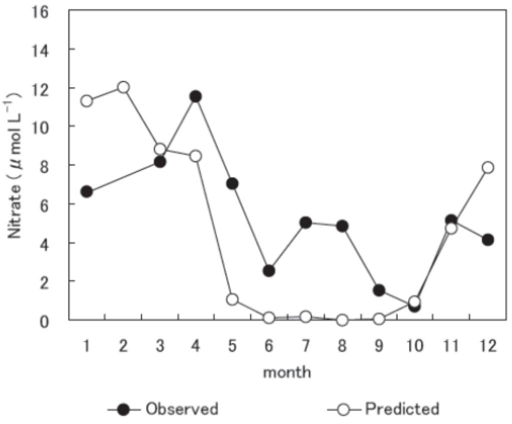


Figure 6.5.1. Seasonal pattern of predicted and observed monthly-mean stream NO_3^- concentration using PnET-CN model in Uryu Experimental Forest in the Northern Hokkaido, Japan. *(Dorokawa Experimental Watershed, 2003-2004, unpublished data) (Shibata et al.,2006).*

Shibata *et al*. (2006) applied the PnET-CN model to the forest watershed in the Northern Hokkaido. Although they used the default values for parameters, estimated seasonality of nitrate concentration in stream water was corresponded well with the observation (Figure 6.5.1). On the other hand, the application of the same model to the forests suffered from long-term disturbance failed the quantitative estimate of changes in biomass and leaf nitrogen concentration etc. It was considered that reconfiguration of the parameters based on the conditions in the targeted ecosystems would be necessary (Tokuchi *et al*., 2006).

For the estimation of soil and water chemistry, especially their acidification by these models, evaluation of mineral weathering rates are essential. Although this process is included in most of the models, it is difficult to assign the suitable rate constant to each objective ecosystem and weathering rates are often determined by calibrating the model. Field weathering rates were estimated based on the Sr isotope analysis for two forests: Kannondai (red pine stand on volcanic soil) and Yasato (deciduous stands on granitic soil) in Japan (Shindo *et al.*, 2001). As Sr concentrations in forest ecosystems were highly correlated with Ca and Mg concentrations, Sr isotope ratios can indicate the behavior of these two elements. Based on the yearly base cation (BC) budget shown in Figure 6.5.2, Ca and Mg weathering rates were calculated by the isotope mixing equation (1) (Miller *et al.*, 1993) assuming that soil solution is mixture of infiltrated solution from the upper layer and release from soil minerals and isotope ratio of exchangeable Sr is identical to that of the soil solution.

$$BC_W = BC_{IN}\ \{(R_W - R_{IN}) / (R_W - R_{SOLN})\} \qquad (1)$$

$$R = ({}^{87}Sr/{}^{86}Sr) / \{1 + ({}^{87}Sr/{}^{86}Sr)\}.$$

The Sr isotope ratio decreased from rain to throughfall to litter layer percolation to soil solution (Figure 6.5.3). The value for exchangeable cations of topsoil from Kannondai was very close to that of the soil solution at 20 cm depth. Because the ratio of the soil solution was smaller than that of the litter layer percolation, the isotope ratios of the soil minerals are expected to be smaller than that of the soil solution. In the clay and silt fractions of the soil, however, the ratios were quite high compared with the water samples from both areas. The sand fraction had the smaller isotope ratio suggesting that sand fraction could be the main source of BC weathering. The results of the weathering rates for the first and the second soil layers are shown with other fluxes derived from the field measurements in Table 6.5.2. The weathering rates of these two cations were 51% to 64% of the total ion release from the soils.

Table 6.5.2. Base cation fluxes based on budget analysis and Sr isotope ratios (kmol$_c$·ha^{-1}yr^{-1}).
(modified table from Shindo et al., 2001). Abbreviations were shown in Figure 6.5.2.

	BC_{IN}	BC_{SOIL}	BC_W		BC_{EX}	BC_{UP}	BC_{RF}	BC_{GU}	BC_{OUT}
			1st	2nd					
Kannondai Ca	2.48	2.27	0.88	0.28	1.11	3.3	2.2	1.1	1.45
Mg	1.22	0.89	0.43	0.14	0.32	1.31	0.96	0.35	0.8
Yasato Ca	3.17	1.38	0.42	0.40	0.56	3.38	2.98	0.4	1.17
Mg	1.73	0.9	0.23	0.28	0.39	1.91	1.6	0.31	0.72

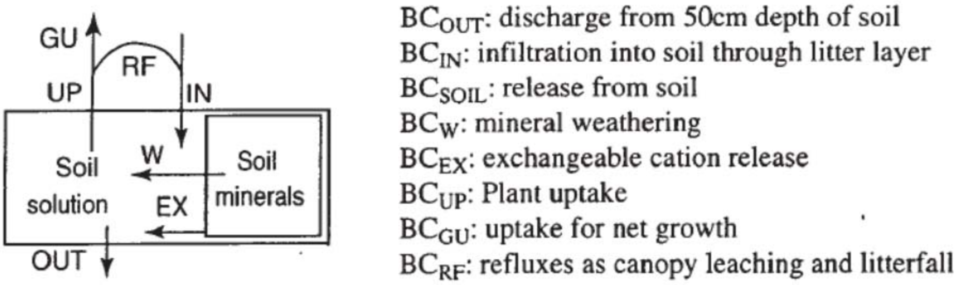


Figure 6.5.2. Base cation budget in soil layer. *(Shindo et al. 2001).*

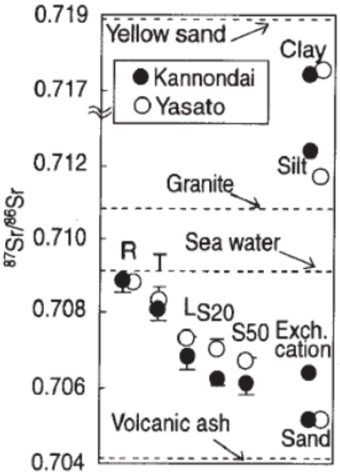


Figure 6.5.3. Means and standard deviation of Sr isotope ratios of rain (R), throughfall (T), litter layer leachate (L), soil solution at 20 and 50 cm depth (S20, S50) and soil fractions.

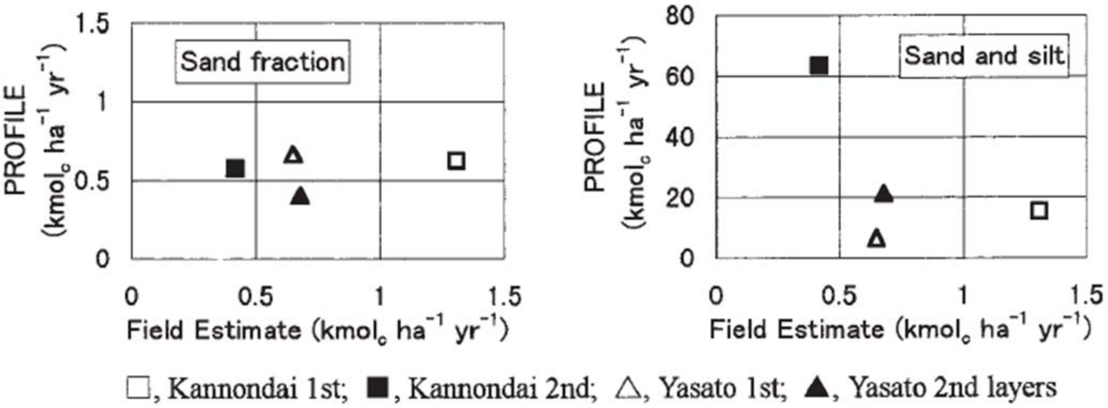


Figure 6.5.4. Comparison of weathering rate calculated by modified PROFILE and estimated by Sr isotope analysis*(Fumoto et al., 2001)***.** *BET surface area of sand fraction or sand and silt fraction was used in PROFILE calculation.*

Profile model (Warfvinge and Sverdrup, 1992) is a steady-state model of soil chemical processes dealing with the weathering rate of each particular mineral by a kinetic model assuming the reaction with H^+, H_2O, CO_2 and organic acid. The original model does not consider the dissolution of volcanic glass, the major primary mineral in volcanic ash soils common in East Asia including Japan. For the application of the model to soils in Japan, the rate coefficients of Si and BS release were estimated from perlite glass dissolution experiments in literature and average composition of volcanic glasses. The resulted rate coefficients were then incorporated into PROFILE model. BC

NB. BET = BET Theory, "BET" consists of the first initials of the authors' family names.*

(Ca and Mg) weathering rates estimated by the modified PROFILE model for Kannondai and Yasato soils were compared with the field estimates (Figure 6.5.4). When BET surface area of sand (2 mm–20 μm) fraction was used as the mineral surface area, calculated weathering rates were the same order of magnitude with field estimates, while it is 1–2 order higher when sand to silt (2 mm–2 μm) fraction was used. This result suggested again that sand fraction could be a reasonable estimate of weatherable mineral surface area of volcanic soils.

6.5.2 Risk of acidification and eutrophication

Monitoring of air pollution and acid deposition were conducted to determine the condition of the terrestrial environment and ecosystem on which human beings depend for healthy lives. The declines of forests have been reported in several countries but the causes remain difficult to be identified, although air pollution and acid rain have often been suspected. In 1980s, soil acidification was thought to be the key factor behind forest decline because aluminum toxicity in acidic soil harms fine roots and tree growth (Ulrich *et al.*, 1980). However, the decline involves several factors; for example, insect and fungal diseases, meteorological damages such as frost and drought, air pollution and acidic deposition, ozone exposure, and so on. Now we recognize that multiple causes and complex mechanisms are involved in ecosystem damage. Unfortunately, information remains limited on the mechanisms of the decline and death of forests. Even though the causes of the decline of vegetation may remain unknown, it would be useful to estimate areas at risk of ecosystem decline.

In addition to environmental acidification, the impact of climate change due to global warming, increasing temperature and change in rainfall pattern, would exacerbate ecosystem decline in future.

6.5.2.1 Decline of forests

Direct effects of air pollutions on forests have long been reported in areas with heavily polluted regions e.g. due to mining and heavy industry. Currently, developing countries with high economic growth rates face pollution problems previously experienced in Europe and North America. Several scientific reports have been compiled in Asia, namely China, and Republic of Korea, and Japan.

In the industrial complex of Yeosu City in Republic of Korea, the decline of the red pine was observed over a decade and the long term trend of tree ring chemistry was studied to identify the effects of acid depositions (Kwak *et al.*, 2009). Yang *et al.* (2009) also reported a significant relationship between pine tree decline and soil nutrient availability under the influences of acid precipitation; as detected by data collected from several industrial cities over Republic of Korea and concluded that Mg deficiency and excess Al were possible causes of the decline of pine trees. In the urban environment of the Seoul metropolis, the ecological degradation of the soil and vegetation structure was also reported (Lee *et al.*, 2008). Acidified fog chemistry was monitored at Daekwanreung, an the Eastern coastal city in Republic of Korea (Kim *et al.*, 2006), which is also suspected to be a cause of tree decline in Japan (Igawa *et al.*, 1998).

We also find many reports on forest declines in China. Despite several efforts to reduce pollutant emissions in China, high rates of economic growth are projected and pollutant emissions are set to increase in future (Ohara *et al.*, 2007). For example, in Chongqing city, with the worst polluted air in Southern China, Masson pine forests have sustained considerable damage (Wang *et al.*, 2007). Similarly, Masson pine trees in Guangdong province, Southern China, where a high frequency of acid deposition was monitored (Huang *et al.*, 2009), were damaged after the open-door policy triggered an economic boom (Kuang *et al.*, 2008). As for soil acidity, increases in the Al^{3+} / $(Ca^{2+}+Mg^{2+})$ molar ratio due to acid deposition were observed in Chinese forest soils (Guo *et al.*, 2006).

In Russia, the risk of lake acidification (Semenov *et al.*, 2006), lichen growth as an indicator of forest decline (Otnyukova, 2007), and mapping of the forest area impacted by air pollution around Baikal Lake (Mikhailova *et al.*, 2008) were reported from the Siberian region. In Mongolia, tree decline (Sase *et al.*, 2005) and the influence of air pollution on lichen were reported in forest stands (Hauck, 2008).

6.5.2.2 Eutrophication risk

Although sulphur deposition has been suppressed in many countries due to emission control of SO_2, nitrogen deposition, the other acidification source, has been remained constant or increasing since its emission control has generally been delayed. Although the risk of acidification by NO_3^- and NH_4^+ is acknowledged by researchers, nitrogen deposition is relatively less highlighted because nitrogen is an essential nutrient for plant growth. In natural ecosystems, nitrogen is usually the key factor limiting plant growth, which means excess nitrogen may be acquired by vegetation, and reduces the risk of soil acidification. However, high nitrogen loads induce another environmental risk, namely the decline of biodiversity. We understand that maintaining biodiversity is important for human existence, as well as for ensuring the function of suitable nutrient cycling (Vitousek *et al.*, 1995; Matson *et al.*, 2002). Even if the nitrogen level rises only slightly, chronic high N deposition may considerably and plastically affect the diversity of plant species (Phoenix *et al.*, 2006). The effects of excess nitrogen seem to differ among ecosystems such that opposing trends; namely increased and decreased diversity, were reported following field observation and some experimental studies. However, high N deposition surely influences biodiversity and nutrient cycling in ecosystems (Bobbink *et al.*, 2010) as shown in Figure 6.5.5.

In Japan, Itoh *et al.* (2004) reported that N concentrations in head stream waters collected from maintains around the Kanto Plain were significantly higher compared with those from elsewhere, which suggested that the high N load in the Kanto Plan exceeded the N demand of forest growth. This phenomenon is known as so-called “nitrogen saturation” in the forest ecosystem. Nakahara *et al*. (2010) also reported on progressive soil acidification and increased N and S levels in stream water in Lake Ijira catchment (see 6.2.2.2), central Japan, where acidic deposition levels were observed to be the highest in Japan. However, the N demand of trees and their physiological responses to high N levels vary among species (Wallace *et al*., 2007). Nagakura *et al*. (2004) reported that transpiration of the Japanese cedar increased under high N load compared to that of the Japanese cypress. High N deposition stimulates transpiration from cedar needles, even in dry soil moisture. This water stress might be the cause of cedar decline observed in the Kanto Plain in the 1980s.

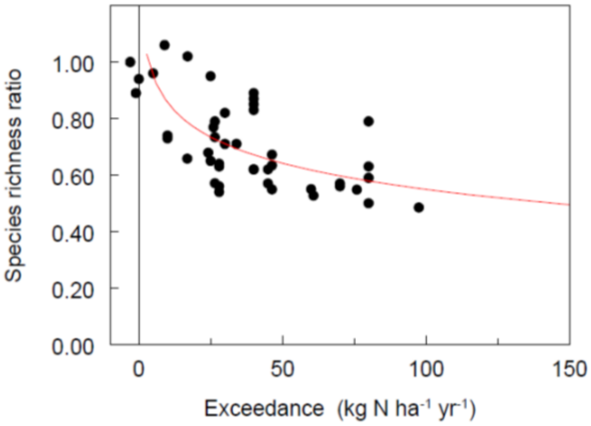


Figure 6.5.5. Decrease in the species-richness ratio (the ratio of the mean number of grass species in N treatment and the control) with nitrogen loads exceeded in European grassland experiment. *(Bobbink et al., 2010).*

Outside industrial areas, the application of fertilizer to farmland is a source of nitrogen emissions, which are also related to nitrogen saturation. In China, significant soil acidification due to increasing application of fertilization has been widely observed (Guo *et al*., 2010). In Japanese rural areas, Baba *et al*. (2001) reported that high N deposition was observed in a forest near the livestock industry. These findings suggested that the risk of eutrophication had spread to local and suburban areas due to modern agricultural management systems.

6.5.2.3 Identification of areas susceptible to acid deposition

In spite of the policy of reducing pollutant emissions, our environment still faces the gradual risk of acidification and eutrophication. The only way we can detect the effects on biodiversity and other environmental risks in the ecosystem is to monitor the condition of vegetation, soil, streams and lakes in the area. To determine changes in biodiversity, grass and herb species should be monitored, as well as tree decline. However, we must strive to reduce the cost of monitoring because polluted air and precipitation are widespread. Accordingly, in addition to monitoring of air and precipitation chemistry, changes in vegetation cover and soil characteristics must be examined. Combining this information, hot spots of endangered ecosystem can be identified, which facilitate a cost-effective monitoring system by focusing on specific areas (Takahashi *et al*., 2001). For instance, Figure 6.5.6 shows the distribution of forest soils with low exchangeable base and high exchangeable Al, which may be sensitive to acid deposition.

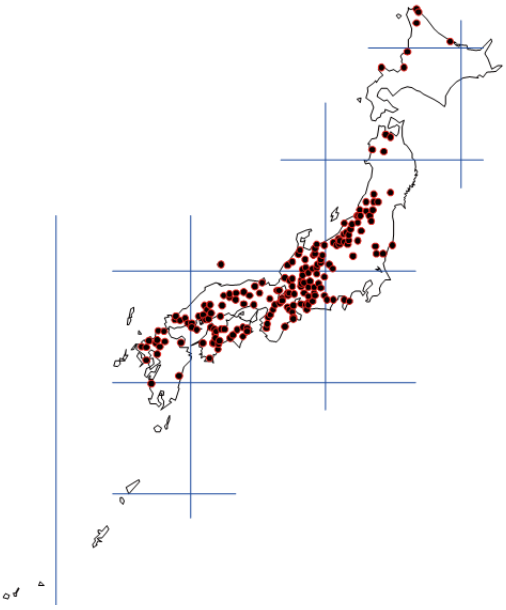


Figure 6.5.6. Distribution of forest soils with low exchangeable base and high exchangeable Al. *(Takahashi et al.,2001)***.**

To identify susceptible areas, we must know the exact distribution of vegetation, soil, topography, and geology as well as the deposition of acid substances. Some species may have tolerance to acidic soil, which can be determined by the natural distribution of the tree species. However, acidic deposition may enhance the stress of these trees, which may increase susceptibility to other stresses such as exposure to high ozone concentrations. Figure 6.5.7 shows a trial of this kind of risk map as reported by Hicks *et al*. (2008).

Areas where soil base saturation will be reduced to less than 20% within 20 years are distributed from South China to the Indochina peninsula (Figure 6.5.7). This area is projected to show high economic growth in future, which may accelerate the time course of acidification faster than expected by the model. In addition, because soil phosphate is often a factor limiting vegetation growth in tropical areas (Elser *et al*., 2007), increasing nitrogen deposition may induce a nutrient imbalance in natural vegetation, which may also change vegetation structure and compositions. Therefore, we should update the map based on the latest information from monitoring data and some models to reduce the risk of environmental deterioration.

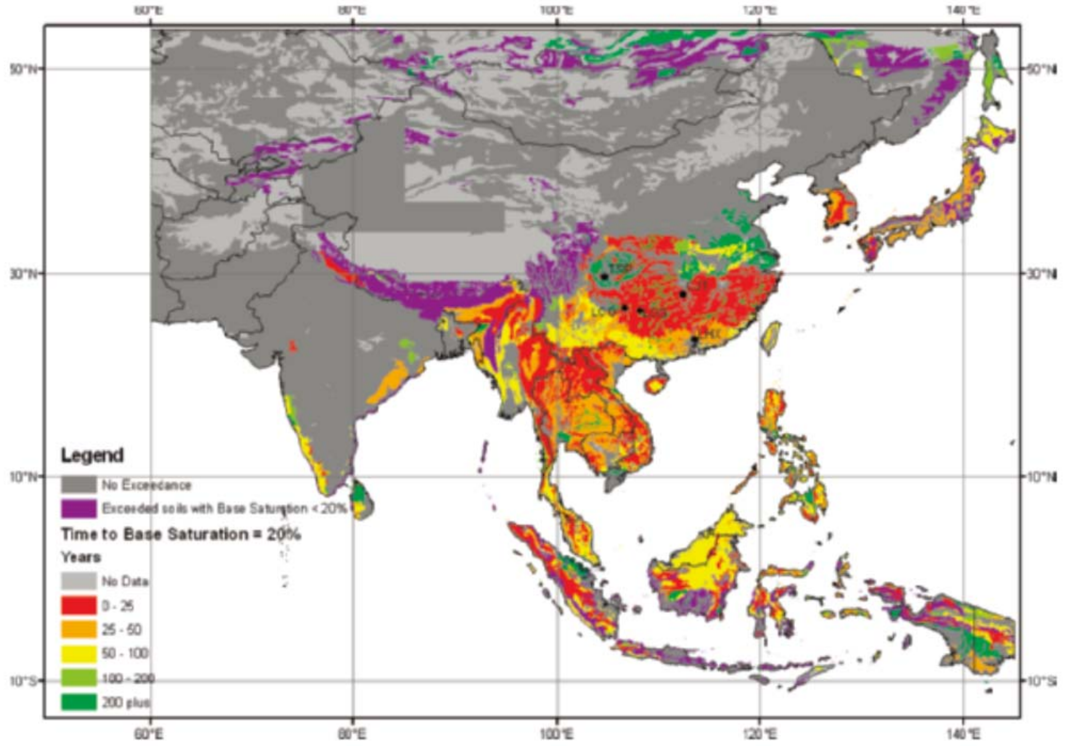


Figure 6.5.7. Case map of the area susceptible to decline of soil base saturation under acidic deposition by the model under the scenario of emission reduction. *(Hicks et al., 2008).*

6.5.3 Environmental risk assessment of ozone and PM

6.5.3.1 Effects of O_3 and PM on human health

Ozone is a highly oxidative compound formed in the lower atmosphere, and owing to its highly reactive chemical properties, it is harmful to vegetation, materials and human health (WHO 2003, 2005). The following effects are acute responses to the exposure to ozone (WHO, 2008); adverse effects on pulmonary function and respiratory symptoms, lung inflammatory reactions, increase in medication usage, hospital admissions and death rates. The reduced lung function, development of atherosclerosis, development of asthma and reduction in life expectancy, are likely associated with ozone exposure as chronic effects, which are less conclusive, however, mostly due to limited number of epidemiological studies (WHO, 2005).

Particulate matter (PM) is stressed on its significance since exposure to PM in ambient air has been linked to a number of different health outcomes due to the ability to be inhaled into respiratory system. WHO (2006) reported that the effects related to short-term exposure are lung inflammatory reactions, respiratory symptoms, adverse effects on the cardiovascular system, increase in medication usage, hospital admissions and premature mortality; the effects related to long-term exposure are increase in lower respiratory symptoms and chronic obstructive pulmonary diseases, reduction in lung function, and life expectancy.

To provide adequate protection of public health, WHO set the Air Quality Guideline (AQG) value for ozone at 100 $\mu g.m^{-3}$ (50 ppb) for an 8-hour daily average, which is estimated to cause 1–2% increase in daily mortality (WHO, 2005). The Clean Air Act of the U.S. required the U.S. Environmental Protection Agency (USEPA) to set National Ambient Air Quality Standards (NAAQS) for ground-level ozone at 75 ppb for an 8-hour daily average (3-year average of the 4th-highest daily maximum 8-hour average must not exceed 75 ppb) (USEPA, http://epa.gov/air/criteria.html).

WHO set AQG value of 10 $\mu g.m^{-3}$ for annual mean of $PM_{2.5}$ to represent the lower end of the range over which significant effects on survival have been observed in the American Cancer Society (ACS) Study (WHO, 2005; Pope *et al.,* 2002). AQG of $PM_{2.5}$ is also set for value of 25 $\mu g.m^{-3}$ for 24-hour mean (99th percentile (3 days $year^{-1}$)), since, at low levels, there is less concern about remaining episodic excursions. The Clean Air Act of the US required USEPA to set NAAQS for $PM_{2.5}$ at 15 $\mu g.m^{-3}$ for annual mean (3-year average must not exceed), and 35 $\mu g.m^{-3}$ for 24-hour (3-year average of the 98th percentile must not exceed) (USEPA, http://epa.gov/air/criteria.html).

6.5.3.2 Risk assessment of ozone and PM on human health in East Asia

There are some studies on the risk assessment of human health for air pollutants including East Asian region. These studies are based on statistically considerable relationships identified between elevated concentrations of ozone and premature mortality which is considered the most robust, reliable and significant indicator (WHO, 2008; Lippmann, 2009). Positive correlations and statistically significant relationships have been identified between $PM_{2.5}$ concentrations and mortality, lung cancer, and reduced lung function in human populations (Pop III *et al*., 2002; Pope III and Dockery, 2006; Lippmann, 2009).

There are several indices for assessing effect of ozone on human health such as; SOMO35, accumulating ozone concentrations above a threshold of 35 ppb; EU60, using a threshold of 60 ppb; and USEPA80, evaluating the exceedance of 80 ppb. Ellingsen *et al.* (2008) found, by using an 18 ensemble of global chemistry transport models, that SOMO35 probably the most robust air quality index when evaluating results from individual models.

To evaluate the global mortality caused by long term exposure to ozone and $PM_{2.5}$ for the year 2000, Anenberg *et al.* (2010) used global atmospheric chemical transport model (CTM) simulations and health impact functions. Anthropogenic ozone was associated with an estimated 0.7 ± 0.3 million respiratory mortalities (6.3 ± 3.0 million years of life lost) annually. Anthropogenic $PM_{2.5}$ was associated with 3.5 ± 0.9 million cardiopulmonary and 220,000 ± 80,000 lung cancer mortalities

(30 ± 7.6 million years of life lost) annually. Figures 6.5.8 and 6.5.9 show the estimated annual mortalities attributed to ozone and $PM_{2.5}$, respectively. Mortality estimates were reduced approximately 30% when it is assumed that low-concentration thresholds of 33.3 ppb for O_3 and 5.8 $\mu g.m^{-3}$ for $PM_{2.5}$.

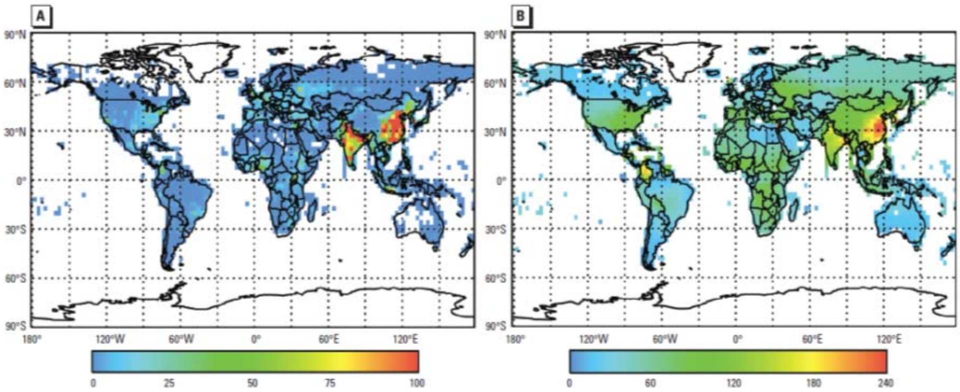


Figure 6.5.8. Estimated annual premature mortalities attributed to anthropogenic O_3 when no upper or lower concentration threshold is assumed, for respiratory mortalities per 1,000 km^2 (A) and rate of respiratory mortalities per 10^6 people (B). *(Anenberg et al. 2010).*

WHO estimated that 373 thousand deaths were caused by ambient particulate matter in the year 2000 within WPR (East Asia and Pacific) region as a result of cardiopulmonary disease, lung cancer and Acute Respiratory Infection (ARI) in children aged 0–4 years, combined (WHO, 2004).

Liu *et al.* (2009) estimated global annual premature mortalities (for adults age 30 years and above) due to inter-continental transport of $PM_{2.5}$ to be nearly 380 thousand in 2000. Approximately half of these deaths occurs in the Indian subcontinent (IN), mostly due to aerosols transported from Africa and the Middle East (ME). Approximately 90 thousand deaths globally are associated with exposure to foreign (i.e., originating outside a receptor region) non-dust $PM_{2.5}$. More than half of the premature mortalities associated with foreign non-dust aerosols are due to aerosols originating from Europe (20), ME (18) and East Asia (EA, 15); and nearly 60% of the 90 thousand deaths occur in EA (21), IN (19) and Southeast Asia (16) (in thousands).

6.5.3.3 Ozone effects on crops

Ozone causes reductions in crop yield, tree growth and carbon sequestration, and to modify species composition (Ashmore, 2005; USEPA, 2006). In terrestrial ecosystems, the most important direct effects of ozone are those on leaf physiology and plant growth; many other indirect effects on ecosystems flow from these primary direct effects (The Royal Society, 2008).

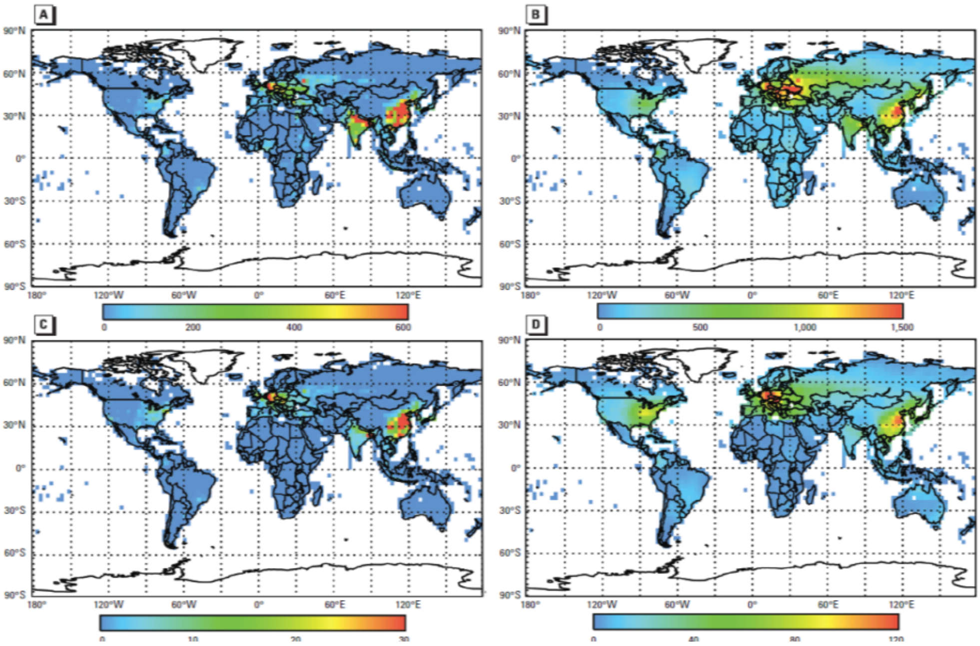


Figure 6.5.9. Estimated annual premature mortalities attributed to anthropogenic $PM_{2.5}$ when no upper or lower concentration threshold is assumed, for cardiopulmonary mortalities per 1,000 km^2 (A), rate of cardiopulmonary mortalities per 10^6 people (B), lung cancer mortalities per 1,000 km^2 (C), and rate of lung cancer mortalities per 10^6 people (D). *(Anenberg et al. 2010)***.**

Except Australia, ground level ozone has already exceeded the threshold for damaging sensitive vegetation in all regions including Asia (Ellingsen *et al.*, 2008). To estimate the ozone effect on crop yield, there are many studies implemented in East Asia, and many researchers identified positive correlations (yield response functions) between losses of crop yields and elevated ozone concentration indexes; such as 7/12-h mean, AOT40, SUM06 and W126 (Kobayashi *et al.*, 1995; Mills *et al.*, 2007; Ellingsen *et al.*, 2008). The yield response functions for ozone differ from North America with Asia, and some of crops (wheat and rice) showed a greater sensitivity in Asia (Emberson *et al.*, 2009). Recently accumulative stomatal ozone flux (AFstY) indicator has been used in Europe to consider real ozone uptake by plant (ICP M&M, 2004).

Using the integrated assessment approach including regional/global CTM simulation, studies estimated the yield loss in East Asia and Global scale. Aunan *et al.* (2000) estimated the crop (rice, wheat, soybean and corn) yield loss in China in 1990 and 2020 based on 3 different indices that yield loss of spring wheat using 7/12 hrs d^{-1} mean (M7/M12), AOT40 and SUM06 are 3.2%, 0.1% and 9.1% in 1990, and, 8.2%, 29.3% and 29.3% in 2020, respectively. Wang *et al.* (2007) showed, using M7 and M12, that in 1990 China, Japan and South Korea lost 1–9% of their yields of wheat, rice and corn, and 23–27% of their yield of soybeans. In 2020, yield loss is projected to increase to 2–16% for wheat, rice and corn and 28–35% for soybeans, with an associated value of 1990 US$ 3.5, 1.2 and 0.24 billion in 1990, and increasing economic costs by 82%, 33%, and 67% in

2020 for China, Japan and South Korea, respectively. Nawahda *et al*. (2011) indicated, based on AOT40 index that in 2000 there are losses of approximately 30.5%, 10.4%, 2.8%, and 19.0% of total yields for wheat, rice, maize, and soybeans in China, respectively. Similarly, in 2005 the losses are approximately 47.2%, 12.5%, 5.6%, and 27.1%. Avnery *et al*. (2011) estimated, under the IPCC SRES A2 and B1 scenario, substantial reductions in yields in 2030: in the A2 scenario yield loss of wheat ranges from 5.4 to 26%, 15–19% for soybean, and 4.4–8.7% for maize with total global agricultural losses worth \$17–35 billion USD_{2000} annually, in B1 scenario 4.0–17% for wheat, 9.5–15% for soybean, and 2.5–6.0% for maize, with total losses worth US\$12–21 billion annually. Figure 6.5.10 shows the estimated yield loss based on B1 scenario.

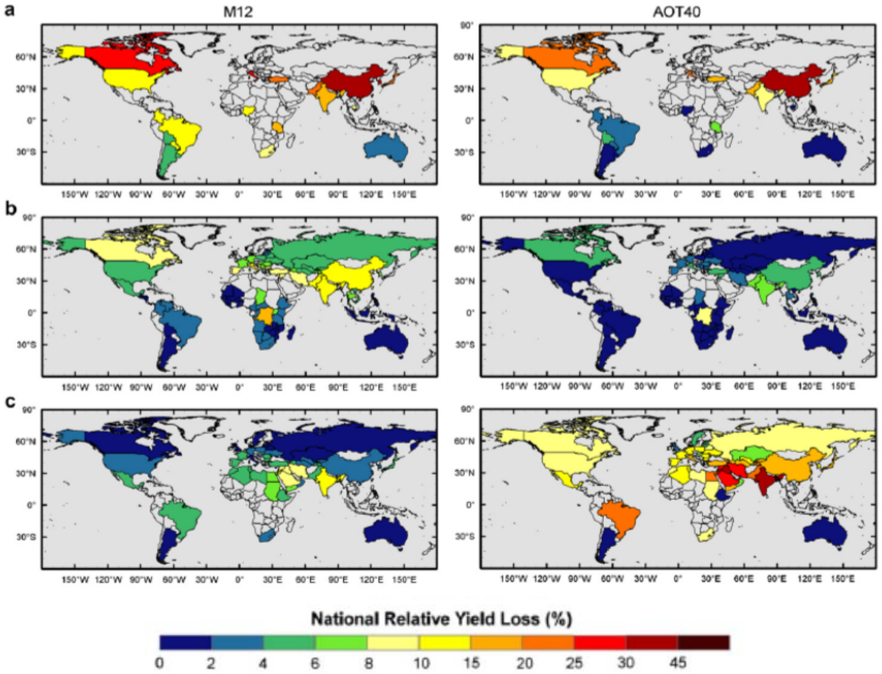


Figure 6.5.10. National relative yield loss under the 2030 B1 scenario according to the M12 (left panels) and AOT40 (right panels) metrics for (a) soybeans, (b) maize, and (c) wheat. *(Avnery et al. 2011).*

The above health risk assessment studies are mainly based on the epidemiological researches in Europe and the U.S., therefore, it should highly conduct the epidemiological research in East Asia reflecting regional specific ozone/PM concentrations and their responses so that the uncertainty in the risk estimation can be reduced. The development of the fundamental data sets such as crop distribution and calendar is crucially important for the more reliable risk assessment of crop yield loss as well as exposure-response function of crops in East Asia.

6.6 Other international initiatives on air pollution

6.6.1 WMO/GAW

The Global Atmosphere Watch (GAW) programme of WMO is a partnership involving 80 countries, which aims to provide reliable monitoring data and information on the chemical composition of the atmosphere, its natural and anthropogenic change. (http://www.wmo.int/pages/prog/arep/gaw/gaw_home_en.html). The rationale for the Global Atmosphere Watch is the need to understand and control the increasing influence of human activity on the global atmosphere.

GAW focuses on global networks for GHGs, ozone, UV, aerosols, reactive gases, and precipitation chemistry. Among them "precipitation chemistry" and "reactive gases" networks would be most relevant to EANET. Surface ozone is also discussed in the "reactive gases" activities, in addition to "ozone" activity which is more focused on stratospheric ozone. There are four baseline stations of WMO/GAW in East Asia, Mt, Waliguan (China), Minamitorishima (Japan), Danum Valley (Malaysia) and Bukit Kototabang (Indonesia), of which the last two are in common to EANET sites. Figure 6.6.1 and 6.6.2 shows the WMO/GAW monitoring sites for precipitation chemistry and surface ozone, respectively. In addition to these networks, the Urban Research Meteorology and Environment (GURME) project (http://www.wmo.int/pages/prog/arep/gaw/urban.html) is also relevant to activities related to EANET. It aims to collect information and have capabilities that are essential to the forecasting of urban air pollution and the evaluation of the effects of different emission control strategies.

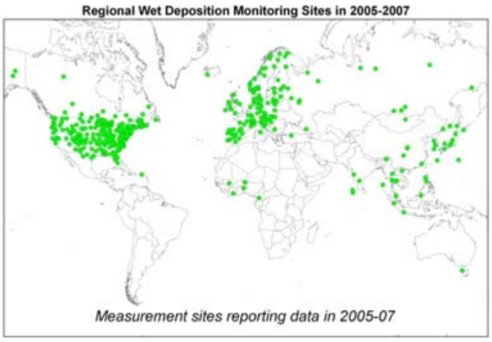


Figure 6.6.1. WMO/GAW sites for precipitation chemistry.

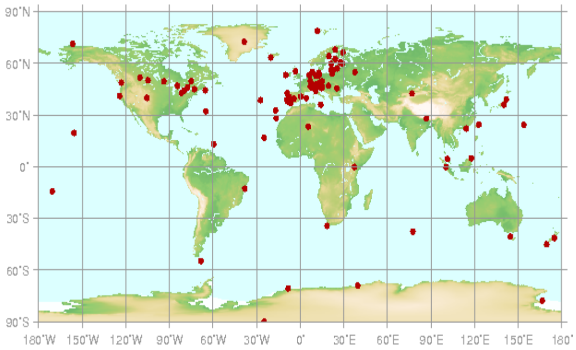


Figure 6.6.2. WMO/GAW sites for surface ozone.

6.6.2 TF HTAP

To develop a fuller understanding of intercontinental transport of air pollution in the Northern Hemisphere, the Executive Body of the UNECE Convention on Long-range Transboundary Air Pollution (LRTAP Convention) established the Task Force on Hemispheric Transport of Air

Pollution (TF HTAP) in 2005 (http://www.htap.org/). The main aim of TF HTAP has been identified as to plan and conduct the technical work necessary to develop a fuller understanding of the hemispheric transport of air pollution for consideration in the reviews of protocols to the Convention. The Task Force organized workshops and task force meetings since 2006 to prepare the 2010 assessment report. During the discussion ozone and particulate matter, mercury and persistent organic pollutants (POPS) have been identified to be important for intercontinental transport, and included in the 2010 report (http://www.htap.org/). Although none of the Asian countries joins the LRTAP, government officials and scientists of Asian countries have been invited to the meetings, and a few scientists from Japan and China joined as an editor and authors of the report.

One of typical outputs in the report is the source-receptor relationship of surface O_3 among the four continents, North America (NA), Europe (EU), East Asia (EA), and South Asia (SA). To quantify the importance of the intercontinental transport, the relative annual intercontinental response metric representing the ratio of the response in a particular region due to the combined influence of sources in the three other regions was defined. The relative annual intercontinental response for annual average surface O3 over each region is shown in Table 6.6.1. They varies from 43% for Europe, to 32% for NA and SA. This indicates that in all four regions, emission changes in the three other source regions are 50–75% as important as emission changes over the receptor region itself.

Table 6.6.1 Annual and spatial mean surface O_3 response (ppbv) to 20% decreases in anthropogenic precursor emissions (NOx, CO, NMVOC, plus aerosols and their precursors). Values are mean (median) ± one standard deviation across the 15 models that conducted the regional perturbation simulations (SR6). Bold font denotes responses to foreign emission perturbations that are at least 10% of the response to domestic emission perturbations. Also shown is the relative annual intercontinental response for each receptor region defined as the ratio of the total response in mean surface O_3 due to changes in the other three source regions compared to that due to changes in all regions.

Table 6.6.1 Annual and spatial mean surface O_3 (ppbv).

	Receptor Region			
Source Region	NA	EU	EA	SA
Annual mean decrease				
NA	1.04(1.03)±0.23	0.37(0.37)±0.10	0.22(0.24)±0.05	0.17(0.19)±0.04
EU	0.19(0.18)±0.06	0.82(0.68)±0.29	0.24(0.24)±0.08	0.24(0.25)±0.05
EA	0.22(0.23)±0.06	0.17(0.17)±0.05	0.91(0.86)±0.23	0.17(0.17)±0.05
SA	0.07(0.07)±0.03	0.07(0.07)±0.03	0.14(0.13)±0.03	1.26(1.18)±0.26
Relative annual intercontinental response				
	32%	43%	40%	32%

6.6.3 LTP

The Long-range Transboundary Air Pollutants in Northeast Asia (LTP) project was commenced in 1995 under the initiative of Korean government as a joint venture by China, Japan and Korea, for collaborative research on long term monitoring and a model study for quantifying the source receptor relationship of acidic pollutants over the Northeast Asia. Sulphur and total nitrate deposition simulations were carried out by three models; CMAQ (Byun and Dennis, 1995) by China, RAQM (Regional Air Quality Model; An *et al*., 2002; Han *et al*., 2006) by Japan, and CADM (Comprehensive Acid Deposition Model; Lee *et al*., 1998) by Korea. Three countries individually compiled their own emission inventories of SO_2 and NOx within their countries in cooperation with administrative agencies, and merged as illustrated in Figure 6.6.3 (NIER, 2005; NIER, 2008).

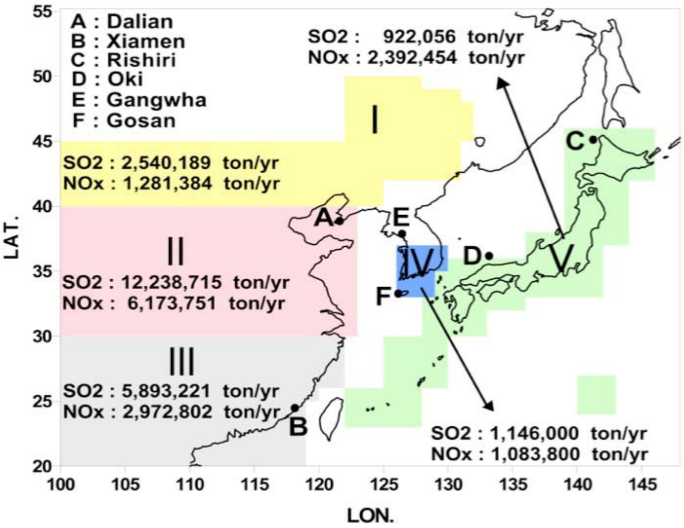


Figure 6.6.3. LTP emission for SO_2 and NOx prepared by China, Japan, and Korea. The five subdivided regions (Region I-V) for the analysis of statistics and the location of the six wet deposition monitoring sites.

The annual total sulphur depositions shown in Figure 6.6.4 simulated in three models yielded nearly identical sulphur deposition patterns with the pattern correlation of 0.91±0.07, reflecting the patterns of emission strength as in Figure 6.6.3. Annual total sulphur depositions over China, Japan, and Korea for 2002 totaled at 7,066, 7,794 and 6,851 kton S, as determined by CMAQ, RAQM and CADM, respectively, with only small deviations of 5~7% among three models (Table 6.6.2). The resultant ensemble mean deposition fraction among total emitted sulphur amounts over the whole domain reached 62.1, 68.6 and 60.3% in the three models, which in turn indicates a transported fraction of 31~40% outside of the Northeast Asia.

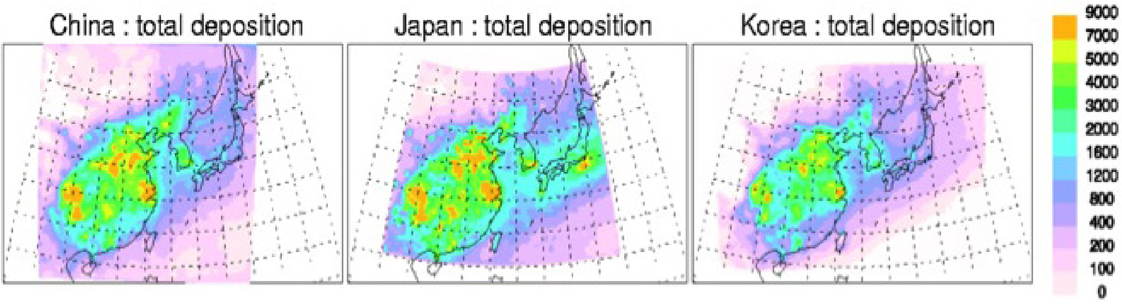


Figure 6.6.4. Distribution patterns of annual total sulphur deposition (Smg.m^{-2}) as simulated by CMAQ, RAQM, and CADM.

The model intercomparison study and quantifying the source-receptor relationship is being underway among three countries for nitrate depositions.

Table 6.6.2. Ensemble mean wet and dry depositions of sulphur (mean ± Standard deviation) (kton-S) from three models during 2002 over the five subdivided regions indicated in Figure 6.4.1.

Region / Season		I	II	III	IV	V
Annual total	Total	705.0(±170)	3282.7(±251)	2290.8(±446)	207.7(±56)	754.2(±400)
	Wet	410.3(±122)	1497.4(±369)	1394.6(±316)	73.0(±35)	463.8(±305)
	Dry	294.8(±50)	1790.3(±243)	896.3(±186)	134.6(±22)	282.0(±117)

6.7 Conclusions

As has been seen in this Chapter, substantial numbers of peer reviewed journal papers have been published in the last few years to elucidate cause and response of regional acid deposition and air pollution and their environmental impacts in East Asia based on observational and modeling studies. Most of those studies reviewed in this Chapter are deeply related to EANET from the viewpoints that EANET monitoring data have been fully used for validation of model simulations or the results were obtained in the EANET "additional activities" by collaboration of scientists of participating countries. These kinds of scientific investigations are crucial for EANET for passing messages of environmental implication of the monitoring data to policy makers and public.

It may be concluded that the EANET in the first decade has been successful in the sense that the monitoring data of acid deposition and atmospheric concentration of ozone have been widely used in international science communities, which means that the data quality of EANET has been accepted to be high enough to be used for scientific research. More active integration of emission inventory, model simulation and monitoring would be a key for future successful operation of EANET.

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Chapter 7. Summary and Recommendations for Future Activities

7.1 Introduction

EANET celebrated its 10-year anniversary in 2010. During the last ten years, air pollution in East Asia attracted much higher attention than before within EANET countries and international community of scientists and policy makers. This is because recent emission amount of air pollutants in East Asia rival those of Europe and North America, and causes more serious urban and rural air pollution as well as contributing hemispherical air pollution and global climate change. This would affect human health, agriculture and natural ecosystems in East Asia, as well as affect hemispherical air pollution and global climate change. Such environmental risk of acidification and eutrophication, and high concentrations of ozone and aerosols are concerned in this second Periodical Report on the State of Acid Deposition in East Asia. This Report presented the analysis of monitoring data focusing on the second five years, 2005-2009, but also including whole of the data obtained since 2000. Related Scientific studies mostly using the EANET monitoring data were also introduced to strengthen raison d'etre of EANET monitoring.

In this chapter, short summary of contents and recommendations for future activities in each topic are presented.

7.2 Summary

7.2.1 Quality assurance and quality control (QA/QC)

The Inter-laboratory Comparison project has been carried out once in every year using common artificial rainwater, standard-spiked collection filter, treated soil and artificial inland water which have been prepared by the Network Center. These samples have been shipped to the laboratories in charge of EANET monitoring and requested to analyze by their adopted analytical method and to submit to the Network Center. Such Inter-laboratory Comparison project has been implemented for the last 12th, 5th, 11th and 10th Inter-laboratory Comparison project for wet deposition, dry deposition, soil and inland aquatic environment, respectively. Visit and inspection of the monitoring sites are requested to be implemented in principle every two years by the National Center staffs, trying to solve any problems of each site through discussion with National QA/QC managers and local staffs.

QA/QC of air concentration monitoring has been long delayed compared to rainwater monitoring for wet deposition. Expert group on preparation of technical manual for air concentration monitoring was established in 2010, and this working group is engaged in development of the manual. When this manual is completed and distributed to EANET participating countries, each county can follow the harmonized condition of air concentration monitoring to assure data quality.

7.2.2 Monitoring of rainwater concentration and wet deposition

The five-year averaged pH of rainwater at the whole EANET sites ranged from 4.4 to 6.2. Among these sites, 26 out of 42 sites (62%) recorded pH lower than 5.0, which is proposed as a guide for "acid rain". As a cause of "acid rain", sulphuric and nitric acids due to anthropogenic emissions of sulphur dioxide (SO_2) and nitrogen oxides (NOx), respectively, have been identified as major acidifying species although organic acids due to biogenic emissions have also been found to be important in areas sufficiently distant from anthropogenic sulphur and nitrogen sources, and particularly in tropical and sub-tropical region. The five-year averaged rainwater pH lower than 4.6 occurred at specific urban sites in Malaysia, China (Chongqing) and Indonesia, and several rural/remote sites in Republic of Korea and Japan. In contrast, average rainwater pH higher than 6.0 occurred in China (Xi'an) and Mongolia (Ulaanbaatar). The high pH values are due to higher contribution of alkaline species such as ammonia (NH_3) from agriculture and calcium carbonate as soil dust, respectively.

In the sites of EANET region, anthropogenic sulphate called non-sea salt sulphate (nss-SO_4^{2-}) concentration is higher than that of nitrate (NO_3^-), which means the emission of SO_2 is still the major contributor to "acid rain". This is particularly true in the inner land of China where the contribution of NO_3^- is less than 20% of the sum of nss-SO_4^{2-} and NO_3^-. In contrast, NO_3^- is relatively high at a Malaysian urban site and Thailand sites. NO_3^- contributes more than 40% of the sum of nss-SO_4^{2-} and NO_3^- in these sites. Since emission control of sulphur is in general precedent to nitrogen control, relative importance of nitrogen as acidifying species in rainwater will be increasing in future in East Asia.

As for alkaline species, contribution of NH_4^+ is higher than nss-Ca^{2+} (non-sea salt calcium emitted from terrestrial sources) at many (more than 90%) EANET sites except for sites influenced by high soil dust, particularly due to yellow sand. As for nitrogenous species, it should be noted that the concentration of NH_4^+ is higher than NO_3^-at 78% of the sites. Particularly high concentration of NH_4^+ in rainwater has been observed at some inner land sites in China (Xi'an and Chongqing). It should be noted that although NH_4^+ is alkaline in the atmospheric species, it turns to NO_3^- in the soil releasing H^+ after deposition and contributes to acidification.

Five-year averaged wet deposition has been calculated by multiplying rainwater ion concentrations by precipitation amount. Annual precipitation amount in East Asia ranges from over 2,000 mm in most of Southeast Asia and Japan to be less than 500 mm in Russia, Mongolia, and Xi'an, China. Top three highest depositions of hydrogen ion (H^+) are seen in Malaysia (Petaling Jaya), Japan (Ijira), and Indonesia (Jakarta). Deposition of nss-SO_4^{2-} was the highest in China (Chongqing), and that of NO_3^- was the highest at the urban site in Malaysia (Petaling Jaya) reflecting the high concentrations in rainwater and precipitation amount.

From the point of ecosystem impact, effective hydrogen ion deposition (H^{+}_{eff}) defined by the sum of H^{+} and NH_4^{+} is more important for soil acidification since NH_4^{+} should be treated as acidifying species in soils. Higher five-year averaged deposition of H^{+}_{eff} was observed at most of Southeast Asian urban sites and Chongqing, China.

In addition to acidification, eutrophication (nitrogen saturation of soil and water) are of concern from a view point of ecosystem impacts in East Asia. For evaluating the risk of nitrogen saturation, the sum of NO_3^{-} and NH_4^{+} depositions (ΣN) have been evaluated. The higher deposition of ΣN was observed in Southeast Asian urban area, inner land of China and Japan (Ijira).

Even if all the data of the whole period (2000-2009) are taken into account, ten years' record is in general not enough to discuss clear trends of ion concentrations in rainwater and wet deposition. Nevertheless, statistically significant cases are seen at some sites for some species. Bangkok site significantly showed increasing trend for NH_4^{+} both in concentration and deposition, which leads to the increasing trends of ΣN and H^{+}_{eff}. Some sites in Xiamen and Chongqing in China showed increasing trends for NO_3^{-} concentration. Urban sites at Petaling Jaya in Malaysia and Jakarta in Indonesia displayed some decreasing trends for nss-SO_4^{2-} in deposition and/or concentration.

7.2.3 Monitoring of air concentration and dry deposition

Atmospheric concentrations of gaseous pollutants (SO_2, NOx, HNO_3, NH_3 and O_3), and aerosol ion components and mass concentration (PM_{10}/$PM_{2.5}$) have also been monitored in EANET. In Northeast Asia, the gaseous and aerosol species, SO_2, HNO_3, NH_3, nss-SO_4^{2-}, NO_3^{-}, and NH_4^{+}at urban and rural sites in China is notably high compared to other area. Among the remote sites, the gaseous components are high at Jeju, Republic of Korea and Terelj, Mongolia compared to other area, and aerosols components are high at Jeju in Republic of Korea, and Oki, Yusuhara, and Hedo in Japan. In Southeast Asia, the gaseous species are high in Petaling Jaya, Metro Manila and Bangkok than others, and aerosol components are notably high in Hanoi. At remote sites, Nakhon Ratchasima and Kanchanabri in Thailand showed higher concentrations of gaseous and aerosol concentrations. The mass concentration of PM_{10} has been observed in China, Japan, Republic of Korea and Thailand. The concentration is considerably high at sites in China followed by in Thailand and at Jeju, Republic of Korea. At most of Japanese sites, the average monthly PM_{10} concentrations in the latter 5 years (2005-2009) were lower than those in the former 5 years (2000-2004). $PM_{2.5}$ has been monitored only at Oki and Rishiri, Japan among the EANET sites.

Ozone (O_3) concentration has been monitored in Japan, Republic of Korea, Thailand and one Russian site (Mondy). The common seasonal variation was observed at these sites, namely, the highest in spring, the lowest in summer, and the second high in autumn. The maximum concentrations in spring at most of the sites in Japan exceeded 50 ppb in monthly average and even higher than 60 ppb at several sites. The monthly average O_3 concentrations in the latter 5 years (2005-2009) were higher than those in the former 5 years (2000-2004) at most sites in Japan.

Five-year mean annual dry deposition amounts for sulphur and nitrogen compounds were calculated using the atmospheric concentration data of gases (SO_2, HNO_3, NH_3) and aerosols (SO_4^{2-}, NO_3^- and NH_4^+), and deposition velocity for each species estimated from meteorological and land use data. Estimate was made only for Japanese site due to availability of hourly meteorological and land surface use data. The ratio of dry deposition to wet deposition ranged from comparable to less than 30%. Higher total (dry and wet) deposition amounts of nitrogen compounds were observed at Ijira and Banryu, which are classified as rural and urban sites, respectively. Total nitrogen depositions were lower at Rishiri and Ogasawara, which are located far from major emission sources of nitrogen oxides. Since the atmospheric lifetime of nitrogen compounds is generally shorter than that of sulphur compounds, the regional characteristics of the total deposition amounts for nitrogen compounds would be more affected by local emission sources.

7.2.4 Monitoring of ecological impacts

Atmospheric deposition may cause acidification and/or eutrophication of ecosystems. An increasing trend on pH of soil was observed in several plots (e.g. Los Baños in Philippines, Kanchanaburi in Thailand), for a declining trend during the last decade was observed in one plot (Imsil in Republic of Korea). But the changes in pH do not correlated to those of acid and/or base cations concentration, suggesting effects of other factors, such as organic substances. Nutrient imbalance may be a cause of changes in the soil chemical properties.

No decline in tree growth and species of understory vegetation was observed during the reporting periods 2000-2009. Overall forest function and structure seemed to be still maintained. Tree decline symptoms observed were mainly caused by pest infection although direct effects of air pollutants such as ozone on trees are also still unclear in the region.

A significant decrease in soil C/N was observed in several plots (Jinyunshan, Xiaoping, Zhuxiandong in China, Bogor in Indonesia, and Banryu in Japan). Simultaneously, NO_3^- concentration of inland water increased in their vicinities (Jinyunshan Lake, Zhuxiandong Stream, and Patengang Lake near Bogor). Too much NO_3^- in soil solution, which is indicated by low C/N, may flow into the stream or lakes and lead to NO_3 increase in the inland waters. It may be possible that high N deposition in these areas caused the phenomenon above. This status in a forest is called “nitrogen saturation”, and terrestrial eutrophication, soil acidification and fresh water eutrophication may occur in stages and would cause harmful impacts to ecosystems.

A declining trend on lake/stream water pH was observed in several sites during the period from 2000 to 2009 and concentrations of SO_4^{2-} increased simultaneously in these sites (Jinyunshan Lake, Xiaoping Dam, and Jiwozi River in China, Patengang Lake in Indonesia, and Semenyih Dam in Malaysia). The acidification may be related leaching of SO_4^{2-} from the ecosystems. Moreover, as mentioned above, an increasing trend on NO_3^- was also observed in one of these sites (Jinyunshan Lake).

The regular monitoring on soil and inland water showed certain symptoms of eutrophication and/or acidification in several sites/plots as mentioned above. Actually, deposition loads of S and N were very high in these areas, although clear evidence of relationship with atmospheric deposition has not been shown. Therefore, a catchment-scale analysis was also promoted to clarify relationship between atmospheric deposition and changes in ecosystems and detailed mechanisms. The form of analysis took from the integrated monitoring including evaluation on atmospheric, vegetation, soil, and inland water depositions with qualitatively and quantitatively. More precise evaluation can be done near future by promotion of the regular catchment-scale monitoring.

7.2.5 Emission inventories

Several regional scale emission inventories in Asia has been published and highly utilized as model input for the analysis air concentrations and depositions of acids, ozone, and their precursors. Both global and regional emission inventories has revealed that anthropogenic emissions in Asia are larger than those in Europe and North America today and will continue to increase in the future. In fact, recent tropospheric satellite observations have demonstrated that the increasing of NO_x emissions in China have been accelerated impressively since 2000 showing somewhat slowing tendency of increasing rate in very recent years. On the other hand, SO_2 emissions in China have decreased since 2007 due to the penetration of fuel-gas desulphurization (FGD) equipment in the power plants.

National emission inventories in EANET member countries have been listed for the first time in this Report. It is mentioned that there are advantage and disadvantage of inventories developed by researchers and government. The advantage of researcher-based inventories is their transparency identifying sources of used parameters, and in many cases published in open literature. The advantage of government-based inventories is that in addition to their official status, they sometimes are based on more detailed country specific information such as emission factors and activity data which are not easily available to researchers.

7.2.6 Modeling activities

Substantial modeling studies have been published in peer-reviewed international science journals, which used EANET monitoring data of acid precipitation/deposition and ozone. The use of monitoring data in scientific papers for validation of models and for analysis to give environmental implication is a measure of merit of monitoring system. In this sense EANET has been very successful as an acid deposition and air concentration monitoring system. Global and regional chemical transport models validated by EANET observational data are much more reliable in simulation in East Asian as compared to 10 years ago when EANET data was not available.

Since the EANET sites are still very sparse in a regional perspective, chemical transport modeling is the only way to give total view of spatial distribution of concentrations, depositions, and temporal variations of monitored species. Global model revealed that annual deposition for NO_3^- (HNO_3 and aerosol nitrate), NHx (NH_3 and NH_4^+) and SO_4^{2-} in Northeast Asia is the highest in the world. General characteristics of high wet deposition distribution of sulphate and nitrate in China from Sichuan basin toward middle and lower reaches of Yangtze River is well represented by regional models agreeing with the spatial patterns of wet deposition synthesized by observational data. Modeling studies on sulphur and nitrogen focusing on Southeast Asia is so far very scares, and the model predicts concentrations in Southeast Asia less accurately, that is related to larger uncertainty in the emission estimates and simulated precipitation fields. High total depositions of sulphur (> 1gS m^{-2} yr^{-1}) are found in Southern China, Northern Vietnam, parts of Java, and around major cities in Thailand, Malaysia and Singapore. Comparison of the modeled atmospheric concentration of SO_2 and SO_4^{2-} concentration in precipitation with the observational data of EANET and others showed in general much poorer agreement than in Northeast Asia. In addition to the model uncertainty in the reproduction of precipitation, the uncertainty of some of observed values of SO_2 either by a filter pack or passive sampler in this region has also been noted. It is recommended that more efforts should be done in Southeast Asia for better tuning of models and for improving quality of monitoring data for air concentrations using automatic instruments.

Substantial number of studies on spatial distributions and seasonal variations of O_3 has been studied using global and regional chemical transport models particularly for Northeast Asia. Spring maximum and summer minimum of O_3 often observed in mid/high-latitude in the East Asia has been well reproduced in general, and could be interpreted as the combination of the factors of in-situ photochemical production, including inter- and intra-continental long range transport, stratospheric intrusion, and meteorological influence of monsoon. Long-term (ca. ten years) increasing and inter-annual variability of surface ozone concentrations around Japan have been reproduced by a regional model, RAMS/CMAQ. A substantial part of increasing has been ascribed to the increase of anthropogenic emissions of precursors in Northeast Asia, and inter-annual variability has been ascribed to surface pressure anomaly over the Pacific Ocean east of Japan.

Source-receptor relationships of total sulphur, total nitrogen, and O_3 in Northeast Asia have been published in several peer-reviewed papers. Particularly for O_3, the ratios of domestically produced, trans-boundary transport in East Asia, hemispherical “background” composed of intercontinental transport and stratospheric intrusion has been quantified for the first time using a global chemical transport mode.

7.2.7 Risk assessment of acidification and eutrophication

The declines of forests have been reported in several countries but the causes remain difficult to be identified, although air pollution and acid rain have often been suspected. In 1980s, soil acidification was thought to be the key factor behind forest decline because aluminum toxicity in acidic soil harms fine roots and tree growth. However, the decline involves several factors; for example, insect and fungal diseases, meteorological damages such as frost and drought, air pollution and acidic deposition, ozone exposure, and so on. Now we recognize that multiple causes and complex mechanisms are involved in ecosystem damage. Unfortunately, information remains limited on the mechanisms of the decline and death of forests.

In the industrial complex of Yeosu City in Republic of Korea, the decline of the red pine was observed over a decade and the long term trend of tree ring chemistry was studied. A significant relationship between pine tree decline and soil nutrient availability has been studied and concluded that Mg deficiency and excess Al were possible causes of the decline of pine trees. In the urban environment of the Seoul metropolis, the ecological degradation of the soil and vegetation structure was also reported. Many reports on forest declines in China. For example, in Chongqing city, with the worst polluted air in Southern China, Masson pine forests have sustained considerable damage. Similarly, Masson pine trees in Guangdong province, Southern China, were damaged after the open-door policy triggered an economic boom. As for soil acidity, increases in the Al/(Ca+Mg) molar ratio due to acid deposition were observed in Chinese forest soils. In Russia, the risk of lake acidification, lichen growth as an indicator of forest decline, In Mongolia, tree decline and the influence of air pollution on lichen were reported in forest stands.

Although sulphur deposition has been suppressed in many countries due to emission control of SO_2, nitrogen deposition, the other acidification source, has been remained constant or increasing since its emission control has generally been delayed. High nitrogen loads induce another environmental risk, namely the decline of biodiversity. In Japan, it was reported that N concentrations in head stream waters collected from maintains around the Kanto Plain were significantly higher compared with those from elsewhere, which suggested that the high N load in the Kanto Plain exceeded the N demand of forest growth, implying so-called “nitrogen saturation” in the forest ecosystem. Progressive soil acidification and increased N and S levels in stream water has been reported in Lake Ijira catchment, involving an EANET ecological site in central Japan, where acidic deposition levels were observed to be the highest in Japan. Transpiration of the Japanese cedar increased under high N load was compared to that of the Japanese cypress, and it was found that high N deposition stimulates transpiration from cedar needles, even in dry soil moisture.

7.2.8 Risk assessment of ozone and PM

Owing to its highly reactive chemical properties, ozone (O_3) is harmful to vegetation, materials and human health. Particulate matter (PM) is stressed on its significance since exposure to PM in ambient air has been linked to human health. Acute and chronic effects of ozone and PM include lung inflammatory reactions, respiratory symptoms, and adverse effects on the cardiovascular system, increase in medication usage, hospital admissions and premature mortality. To provide adequate protection of public health, WHO set the Air Quality Guideline value for ozone at 100 μgm^{-3} (50 ppb) for an 8-hour daily average. The Clean Air Act of the U.S. required the U.S. Environmental Protection Agency (USEPA) set National Ambient Air Quality Standards for ground-level ozone at 75 ppb for an 8-hour daily average. WHO set $PM_{2.5}$ Air Quality Guideline value of 10 μgm^{-3} for annual mean, and 25 μgm^{-3} for 24-hour mean. The Clean Air Act of the US required USEPA to set National Ambient Air Quality Standards for $PM_{2.5}$ at 15 μgm^{-3} for annual mean and 35 μgm^{-3} for 24-hour.

To evaluate the global mortality caused by long-term exposure to ozone and $PM_{2.5}$ for the year 2000, global atmospheric chemical transport model simulations and health impact functions were used. Anthropogenic ozone was associated with an estimated 0.7 ± 0.3 million respiratory mortalities annually. Anthropogenic $PM_{2.5}$ was associated with 3.5 ± 0.9 million cardiopulmonary and 220,000 ± 80,000 lung cancer mortalities annually. WHO estimated that 373 thousand deaths were caused by ambient particulate matter in the year 2000 within West Pacific Region (East Asia and Pacific) as a result of cardiopulmonary disease, lung cancer and Acute Respiratory Infection (ARI) in children aged 0–4 years, combined.

Ozone causes reductions in crop yield, tree growth and carbon sequestration, and to modify species composition. In terrestrial ecosystems, the most important direct effects of ozone are those on leaf physiology and plant growth; many other indirect effects on ecosystems flow from these primary direct effects. Using the integrated assessment approach including regional/global chemical transport model (CTM) simulation, studies estimated the yield loss in East Asia and Global scale. An example of estimates showed that 1–9% of their yields of wheat, rice and corn, and 23–27% of their yield of soybeans were lost in China, Japan and South Korea with an associated loss value of 1990 US$ 3.5, 1.2 and 0.24 billion in China, Japan, and Korea, respectively, in 1990.

7.3 Recommendations for future activities

7.3.1 Quality assurance and quality control (QA/QC)

While the technical manuals for each monitoring activity have been revised, the less reliability of the compiled data has still been remained since there have been much differences in the QA/QC procedure among each participating country. The common QA/QC procedures are strongly recommended to obtain more comparable and reliable dataset. In order to EANET data to be more useful for scientific analysis, monitoring protocols including sampling frequencies, etc., should be harmonized to follow the protocol specified in technical manuals. In order to innovate the transparency of monitoring techniques of participating countries, National Monitoring Plan including site selection, sampling and analytical methodologies, monitoring interval, and so on, should be prepared with certain intervals and submitted to Network Center to be shared.

Data check procedures should be established including data screening confirming matching to the range of Data Quality Objective (DQO), data evaluation for ion balance (R_1), conductivity balance (R_2), and data completeness. As for air concentration monitoring proper calibration and maintenance of the analytical instruments for gases and particulate matters (PM) monitoring are necessary for future activities. Particularly for ozone monitoring, calibration system in EANET-wide traceable to international standard for automatic instruments is strongly recommended. Repeated measurement for the evaluation of the reproducibility for soil and vegetation monitoring and duplicate sampling of lake/river water, and on-site measurements of pH and electric conductivity (EC) are recommended for inland aquatic environment monitoring.

7.3.2 Monitoring of rainwater concentration and wet deposition

As of 2009, there were 54 wet deposition monitoring sites and 44 air concentration monitoring sites operating in the EANET network. Number of site increased during past five years, eight sites for wet deposition and nine sites for air concentration for dry deposition, respectively. However, monitoring sites are still concentrated in some areas. In order to clarify the status of the atmospheric environment in the whole EANET region, establishment of additional monitoring sites should be recommended. Nomination of domestic monitoring sites is one of the possible ways for establishing new EANET site. Geographic, climatic, and ecological situation as well as the location of historical monuments or nature valuables should be taken into account when selecting monitoring area.

One of the importance purposes of atmospheric monitoring system such as EANET is to continue the monitoring for a long period of time in order to elucidate the longer-term trend of atmospheric environment. Wet deposition monitoring manual was revised in 2010 adding new analytical items such as hydrogen carbonate and organic acid. By following the manual, it is expected to monitor rainwater property more accurately especially in the inland and southern part of EANET region. In contrast, since sample collection procedure such as frequency and recommended instrument was not changed, long-term trend of wet deposition could be obtained with high reliability. However, long-term monitoring will bring a lot of problems. Circumstances around the monitoring sites on local and regional scales should be checked continuously. Maintenance of the equipment is also important to keep same condition for sample collection and analysis. Moreover, it is the most important to keep the motivation of researchers and technicians through periodical data evaluation and training program.

7.3.3 Monitoring of air concentration and dry deposition

QA/QC of air concentration monitoring is one of the most important issues in EANET. Currently, there is no comprehensive manual of air concentration monitoring in EANET, however, Expert Group on Preparation of Technical Manual for Air Concentration Monitoring was established in 2010 to develop the manual. When this manual is completed and distributed to EANET participating countries, each county can follow unified condition of air concentration monitoring to assure data quality. Establishment of traceability of automatic air monitoring instruments is another important issue. Management of certified standard gases, calibration of O_3 by international traceable standard and inter-laboratory comparison with reference method of PM monitoring should be promoted in future.

Filter pack method has been adopted for gas and aerosol monitoring in EANET. However, it has been well known that the filter pack method has the artifact problems. For example, volatilization of particulate NH_4NO_3 and NH_4Cl after collection on a filter could occur especially in warmer condition. This may cause underestimation of particulate matter concentration and overestimation of gaseous species concentrations. In order to avoid such artifact, annular denuder and/or diffusion scrubber may be coupled with filter pack. Application of these new innovative methods should be considered in near future to the EANET manual monitoring. Meanwhile, introduction of automatic instruments for real time air concentration monitoring should be encouraged in order to capture air quality more accurately.

Expert Group on Dry Deposition Flux Estimation established in 2007 under the Task Force on Dry Deposition Monitoring has prepared Technical Manual on Dry Deposition Flux Estimation, which was adopted by the Scientific Advisory Committee and endorsed by the Intergovernmental Meeting in 2010. Inferential method was formally adopted for dry deposition monitoring in EANET. In the inferential method dry deposition flux is calculated based on the observed air concentration of particular gaseous or particulate species by multiplying deposition velocity (V_d). Deposition velocity in turn is estimated from meteorological parameters and land surface characters for each species. The real time monitoring of meteorological parameters is a must for the estimation of dry deposition as well as land use data of the monitoring sites. Unfortunately, such parameters are neither measured at the EANET sites nor reported to EANET data report except for a country, which hindered to give dry deposition flux data in the whole of EANET region. It is a first priority to measure and report such parameters for the calculation of dry deposition. Further, since multiplication of real time air concentration to deposition velocity based on real time meteorological parameters is thought to improve the accuracy of dry deposition flux estimation, real-time measurements of air concentrations of gases by automatic instrument is highly recommended.

Careful selection of air concentration monitoring sites is recommended considering site representativeness and compatibility for air quality and dry deposition flux measurement. It is hoped that technical manual for air concentration monitoring under preparation would give a clear guideline.

7.3.4 Monitoring of ecological impacts

In order to promote scientific analysis related to ecological monitoring in EANET, catchment-scale monitoring and analysis is desirable rather than spot monitoring of soil and vegetation or inland aquatic monitoring. In the catchment-scale monitoring, biogeochemical cycles of elements e.g. sulphur, nitrogen, etc., are discussed using the monitoring data of atmospheric deposition, soil and inland aquatic in the same catchment area trying to relate any impact on vegetation. In this respect, it is highly recommended that atmospheric deposition monitoring is to be made in the same catchment area, and both wet and dry deposition flux can be estimated as an input parameter according to the *Guideline for Catchment-scale Monitoring in East Asia* recently prepared and approved in EANET.

Since there is indication showing nitrogen saturation in the EANET ecological monitoring data, not only acidification but eutrophication should be concerned in future monitoring and analysis. Task Force on Soil and Vegetation Monitoring has been trying to identify the area susceptible to acid deposition and air pollution based on soil, vegetation, and geological features. Actual hotspots should be identified with the maps on acid deposition or air pollution in addition to the information above. Taking such hotspots into consideration, a strategic monitoring framework should be designed in near future.

Direct effects of air pollutants on trees should also be worried in some areas of EANET region. Particularly, EANET monitoring data shows that ozone concentrations is high and still increasing in some region, the relationship between ozone and tree decline should be concerned. Nevertheless, automatic instrument data on air pollutants are very limited in forest areas. It is recommended that concentration of ozone and tree health condition should be monitored simultaneously in forest areas using the automatic instrument to capture higher concentration during daytime.

7.3.5 Emission inventories

It is hoped that emission inventories of each participating country will be prepared and open to scientists for the use of interpretation of spatial distribution, temporal variation and long-term trend of atmospheric and rainwater compositions and depositions. It is recommended that the researcher-based and government-based inventories will be harmonized as much as possible through cooperation between administrative agencies and inventory experts in each country.

There have been substantial international efforts to harmonize global emission inventories of short-lived air pollutants and long-lived greenhouse gases reflecting concern on the relevance between air pollution and climate change. Based on these emission inventories discussions on co-benefit of simultaneous reduction of air pollutants and greenhouse gases have been started. It is recommended that EANET would concern on this issue including the discussion of interaction of acid deposition and air quality with climate change.

7.3.6 Modeling activities

Modeling activities by use of both global and regional chemical transport models validated by EANET monitoring data should be encouraged to scientists in participating countries in order to get better understanding on the cause of spatial and temporal variability of monitoring species.

Ensemble mean of multi-models is thought to be more robust than a single model prediction for spatial and temporal variation and source-receptor relationships. MICS-Asia (Model Inter-comparison Study for Asia) is an initiative by volunteered scientists from Northeast/Southeast Asia, North America and Europe for inter-comparison of chemical transport models applied for East Asia. MICS-ASIA Phase III is now under planning phase to start form 2011 including both global and regional models. It is highly recommended to the scientists in relevant countries to join the project in order to discuss the present status of atmospheric environment in more transparent way.

7.3.7 Risk assessment of acidification and eutrophication

After the ten years operation of EANET monitoring for acid deposition, the critical evaluation of the potential environmental risk of acidification in East Asia is necessary. The evidence of forest tree decline has been reported at least some areas in China and Japan but the cause has not been clearly identified with consensus among scientists. Environmental acidification issue has a long history, and the similar situation has been experienced in Europe and North America, and tremendous efforts have been exerted to study and identify the cause. It is recommended to conduct a thorough review of their experiences contrasting to the situation in East Asia for the purpose to accentuate possible potential risk of acidification in East Asia.

Soil eutrophication and nitrogen saturation have been raised relatively new as a cause of forest tree decline. Since the increase of emissions of nitrogen oxides are predicted in East Asia in contrast to the more successful control of emissions of sulphur in near future, the EANET monitoring and the elucidation of “hot spots” for potential environmental risk is recommended to consider not only acidification but also eutrophication. In this regards, it is recommended that accuracy of emission inventories of ammonia whose contributions to both soil acidification and eutrophication are substantial in East Asia, will be improved.

7.3.8 Risk assessment of ozone and PM

There is naturally a large uncertainty for the estimate of the health and agricultural impacts of O_3 and PM. More extensive epidemiological studies are necessary in each EANET counties since most of previous studies are based on the data obtained in Western countries. Also since these estimates are largely depend on spatial distributions of O_3 and $PM_{2.5}$ concentrations obtained by chemical transport models. Due to course resolution of these models they are not necessarily reproduce near surface concentrations accurately. Not only the EANET monitoring data of O_3 and $PM_{2.5}$ for using of the national monitoring network, but also their human and agricultural impacts are recommended to estimate to be more accurately.

Risk assessment of acidification and eutrophication, and ozone and PM is thought to be useful for disseminating common understanding on the importance of atmospheric pollution issues among EANET communities, and is encouraged to be studied in each participating countries.

7.3.9 General recommendation for EANET

The globalization of air pollution calls for new issues such as the link of urban air pollution with regional and hemispherical air pollution, and interlinks between air pollution and climate change. Acid deposition is thus closely linked to many other environmental issues. Acid deposition shares precursor emissions with ozone and aerosols/particulate matter so that mitigation of acid deposition should have co-benefit with mitigation of other atmospheric pollutants. The air pollutants causing acid deposition, such as ozone and aerosols contribute to climate change and climate change affects acid deposition and its effects through e.g. precipitation change. Therefore, extended assessment of the state of acid deposition including other relevant air pollution and climate change should be correlated as well to improve EANET interaction with emerging issues.

An urgent need to secure the achievements to sustain the development of EANET is raised, along with the demand to strengthen the basis of the network operation. For this purpose, the following targets are to be achieved at next stage.

- Improvement of acid deposition monitoring including other relevant components such as ozone and particulate matter (PM) with increasing transparency;
- Extended assessment of the state of acid deposition including other relevant atmospheric pollutants;
- Promotion of research activities including modeling and emission inventories;
- Establishment of an epistemic community and promotion of public awareness to achieve common understanding on atmospheric pollution;

- Strengthening policy relevance of activities providing policy advise and information based upon sound science and assessment;
- Strengthening technology support and capacity by enhancement of cooperative efforts among participating countries;
- Reviewing the present status of atmospheric environment in East Asia and discussing future expansion of the scope;
- Enhancement of collaboration with outside organizations.

To achieve the goals, multi-layer stakeholders, i.e. policy makers, the scientists, the business and the public must commit themselves to an improved ethic of stewardship.

List of the Secretariat of the Drafting Committee for the Preparation of the Second Periodic Report

Asia Center for Air Pollution Research (ACAP)
1182 Sowa, Nishi-ku, Niigata-shi, 950-2144
Japan
Tel: +81-25-263-0550
Fax: +81-25-263-0566
E-mail: eanet@acap.asia

Name	Position	Department
Dr. Hajime Akimoto	Director General	
Dr. Jesada Luangjame	Deputy Director General	
Mr. Makoto Hayashi	Deputy Director General	
Mr. Jiro Sato	Assistant Deputy Director General	
Dr. Ken Yamashita	Head	Planning and Training Department
Mr. Shiro Toda	Senior Researcher	Planning and Training Department
Ms. Kozue Kasahara	Administrative Staff	Planning and Training Department
Ms. Junko Fujita	Administrative Staff	Planning and Training Department
Dr. Tsuyoshi Ohizumi	Head	Atmospheric Research Department
Dr. Keiichi Sato	Senior Researcher	Atmospheric Research Department
Dr. Jyunichi Kurokawa	Senior Researcher	Atmospheric Research Department
Dr. Yayoi Inomata	Researcher	Atmospheric Research Department
Mr. Kenichi Koide	Researcher	Atmospheric Research Department
Mr. Tomokazu Nagai	Researcher	Atmospheric Research Department
Dr. Hiroyuki Sase	Head	Ecological Impacts Research Department
Dr. Naoyuki Yamashita	Researcher	Ecological Impacts Research Department
Mr. Shigeki Uchiyama	Researcher	Ecological Impacts Research Department
Dr. Takuya Shiozaki	Head	Data Management Department
Mr. Ryuta Shirai	Researcher	Data Management Department
Ms. Ayako Aoyagi	Researcher	Data Management Department
Ms. Kumiko Nakamura	Researcher	Data Management Department