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Acid Deposition

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ACID PRECIPITATION RESEARCH IN BRAZIL: A SHORT REVIEW

Alvaro Ramon Ceolho Ovalle and Emmanoel Vieira da Silva Filho
Department of Geochemistry, Fluminense Federal University,
Rio de Janeiro, Brazil

INTRODUCTION

The acid precipitation phenomenon has been extensively studied in various regions of the northern hemisphere, where anthropogenic influences have disrupted natural biogeochemical cycles. The impact of this acidity ranges from acidification of lakes, rivers, groundwater and soils, to changes in agricultural and forest crop productivity (Mason & Seip, 1985).

The acidity of rainfall is associated with carbon, sulphur and nitrogen oxidation, principally from industrial and fossil fuel combustion sources. These anthropogenic emissions may induce a disequilibrium in the atmosphere redox balance, a system more susceptible to these modifications because of its lower dimensions when compared to the hydrosphere and lithosphere (Stum et al., 1987).

In Brazil industrialisation and urbanisation processes have accelerated in the last 30 years, with installation of industries and urban centre growth concentrated along the Brazilian Atlantic Coast (Fig. 1). However, this development has occured without considering the full environmental impact. At this time gaseous, particulate and liquid effluents were freely discharged in soils, rivers and the atmosphere. In view of their more dynamic nature, materials injected into the atmosphere induce long distance impacts, making atmospheric pollution a chronic problem, principally at urban centres and industrial zones concentrated in Sao Paulo and Rio de Janeiro States (Fig. 1).

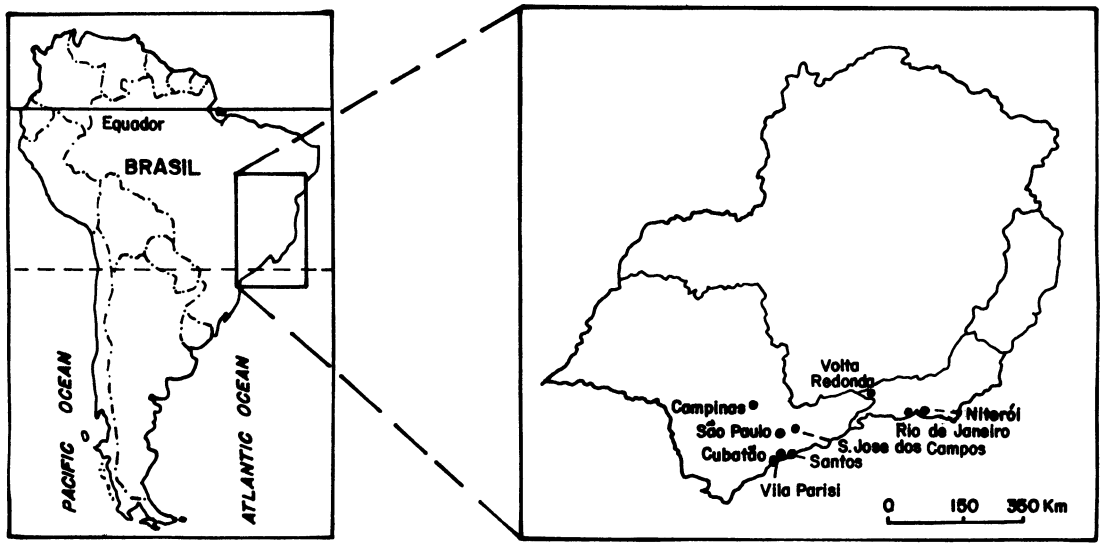

FIGURE 1 - Map of the Southeastern region of Brazil, showing principal areas of precipitation chemistry research.

Air quality has become a serious issue in some areas, such as Sao Paulo and Cubatao cities, where this problem assumed critical proportions, with serious damage to water resources, vegetation and population (CETESB, 1981; Kucinski, 1982; Monteleone Neto, 1982; and Queiroz Neto et al.,1984).The serious health implication of air pollution in these areas has meant that gaseous and particulate matter has been researched and controlled more fully than has acidic precipitation.

The first record of acidic precipitation in Brazil appeared in the literature at the end of the 1970's as part of nutrient cycling studies in forested areas, but only in the 1980's does the literature begin to contain specific papers concerning precipitation chemistry in urban centres and industrial zones.

The objective of this short review is to critically discuss the data available concerning acid precipitation in Brazil, classifying the various levels of the phenomena ranging from the detection of the problem to free acidity budget.

Methodology Used in Studies of Precipitation Chemistry

A critical aspect in analysing precipitation chemistry in Brazil is the absence of a standard method, which usually involves collection apparatus and periodic sampling. Collectors used vary from automatic wet - only samplers (Stallard and Edmond, 1981) to acrilic (Ferreira and Moreira - Nordemann, 1985) and plastic funnels (Silvo Filho, 1985). The latter are permanently opened and the results reported as bulk precipitation. As dry deposition plays an important role in the atmospheric inputs in Brazilian Atlantic Coast ecosystems (Ovalle et al., 1987), results obtained from bulk and wet - only collectors must be critically compared, as dry deposition may have great spatial variation, related to the nature of material injected into the atmosphere and local meteorological conditions.

The data discussed here have a sampling interval varying from single events (Mello & Motta, 1988) to weekly integrated samples (Silva Filho, 1985), and the duration of the study ranging from a few days (Stallard & Edmond, 1981) to some years (Ovalle et al., 1987). Variability of chemical composition of rain water either between events or between years indicates that comparison of data from different locations must be done carefully.

The Detection of the Problem

As precipitation chemistry is intimately related to atmospheric composition, air quality becomes a starting point to understand the acidity of rain water. An evaluation of atmospheric SO_2 content in Brazilian natural, industrial and urban areas is presented in Table 1.

High values for industrial and urban areas when compared to natural regions and rural sites in the United Kingdom, attest the critical state of the atmospheric problem in Brazil. These data indicate the relative position of South America in terms of global man - made emissions of sulphur dioxide.

The Brazilian scientific community involved with atmospheric pollution was alerted to acidity of rain water when mineral cycling studies in forested ecosystems on the Brazilian Atlantic Coast reported pH values ranging from 3.8 to 5.5 (Meguro et al., 1979; Silva Filho et al., 1984a,b; Silva Filho, 1985). These studies were carried out in forested areas between the metropolitan areas of Sao Paulo and Rio de Janeiro States. The frequency that precipitation pH lower than 4.0 was detected leaves no doubt about the magnitude of the problem, principally because these lower values are frequently associated to low volume events (< 10 mm/day), resulting in strongly aggressive solutions (Silva Filho et al., 1987). These data led to a pilot study of acidity related to fog in Rio de Janeiro city, a frequent phenomenon associatedwith coastal montane forested ecosystems during winter. Pedlowski (1986) showed that fog pH ranges between 3.5 and 7.5, with weighted mean of 4.9. Despite the tentative nature of the research, it is clear that there is an acid component present in the fog in the Rio de Janeiro metropolitan region. A study of precipitation chemistry in Cubatao, a highly industrialised zone with significant environmental problems, also reported the problem (Moeira - Nordmann et al., 1983).

Table 2 summarises the pH range in precipitation chemistry in some Brazilian areas. With the exception of the Amazon Forest, the other areas are localised in a zone of some 500km along the

Table 1 : Annual Mean Sulphur Dioxide in different Brazilian regions ; (after Moreira - Nordmann , 1987) and Rural sites in United Kingdom (United Kingdom Review Group on Acid Rain, 1987)

Location	ug SO_2 / m^3	Source
Sao Paulo	44.3	CETESB, 1985
Rio de Janeiro and Volta Re donda	60 - 80	FEEMA, 1984
Rural areas of Sao Paulo State	3.0	CETESB, 1985
Vila Parisi (1984*)	36	CETESB, 1985
Cubatao (1984*)	50	CETESB, 1985
Natural regions	0.15	Lawson & Winchester, 1978
Rural sites in United Kingdom	1.3 - 16.5	United Kingdom 1987

*** = mean value**

Table 2 : pH ranges in precipitation of some Brazilian areas

Location	pH range	Source
Rio de Janeiro	3.8 - 5.4	Silva Filho, 1985
Sao Paulo	3.8 - 6.8	Alonso et al., 1985
Sao Paulo	3.8 - 4.6	Meguro et al., 1979
Cubatao	3.7 - 4.7	Moreira - Nordeman et al., 1983
Niteroi	4.3 - 5.3	Mello & Motta, 1988
Amazon Forest	4.7 - 5.7	Stallard & Edmond, 1981

Brazilian Atlantic Coast (Fig.1), characterising acid precipitation as a well defined regional problem, associated to urban centres and highly industrialised zones.

In the Amazon region acid precipitation with pH values close to 4.0 have been reported (Ungemach, 1969; Brinkmann & Santos, 1973; apud Stallard & Edmond, 1981), but the authors assumed these values as natural in origin, as there is no plausible pollution source to which one could attribute this acidity. Large - scale deforestation in the Brazilian Amazon frequently associated with large scale burning is a potential source of atmospheric CO_2 and other gases (Malingreau and Tucker, 1988),that must be taken into account when assessing precipitation chemistry in the Amazon region.

Precipitation Chemistry and Sources of Elements

Table 3 summarises precipitation chemistry data available in the literature for various areas in Brazil, including natural regions (Amazon), urban centres (Rio de Janeiro, Niteroi, Sao Paulo, San Jose dos Campos, Salvador and Santos), and high industrialised areas (Cubatao and Vila Parisi). The Amazon, as would be expected for a natural region with minimum marine influence, presents low values for all chemicals and could be used as a background in discussion.

In a meq basis SO_4 and NO_3 together have a variable contribution to total anions in precipitation: Amazon - 34%; Santos - 47%; Cubatao - 69%; Sao Paulo - 87%; Vila Parisi - 93%. In the Amazon with no plausible pollution source, chloride is the principal anion, whereas in the other locations with high anthropogenic contribution, SO_4 and NO_3 tend to be dominant. Vila Parisi is a critical case, where SO_4 alone corresponds to 88% of total anions in precipitation.

In a general view we can separate a coastal group with high ionic content precipitation including Salvador, Rio de Janeiro, Niteroi, Cubatao, Vila Parisi and Santos, and an interior group with low ionic content precipitation comprising Sao Paulo, Sao Jose dos Campos and the Amazon.

Table 3 : Chemical composition of precipitation in some Brazilian regions; values in umol/l mean/standard deviation); Amazon data are mean value for the whole basin, except snow samples.

Local	Na	K	Ca	Mg	Cl	NH_4	NO_3	SO_4	pH
Amazon	13/13	1.0/0.7	1.0/1.1	1.2/1.5	15/15	0.8/1.3	2.1/1.7	5.3/2.8	5.1/0.2
Sao Paulo	16.5/41.3	5.9/10.2	23/20	12/18	20/33	46/31	35/22	21/35	4.9/0.7
San José	3.5/---	2.6/---	5.7/---	1.7/---	7.3/---	23/---	-------	-------	4.4/---
Salvador	89/67	6.1/4.6	23/18	10/7.4	111/89	3.9/3.3	-------	-------	5.7/0.3
Rio de Janeiro	130/117	15/8.7	16/8.5	21/23	158/121	-------	-------	-------	4.6/0.4
Niterói	196/278	22/18	-------	21/28	140/200	19/12	6.1/5.0	22/27	4.7/0.3
Santos	221/326	18/19	9.7/10	45/54	296/423	26/41	25/27	61/91	6.4/---
Cubatao	144/148	113/128	15/15	45/45	166/166	72/89	40/26	85/93	4.2/---
Vila Parisi	139/113	143/205	893/1929	193/317	330/420	200/222	84/124	563/599	6.2/0.4

In the coastal group Salvador and Santos present normal pH values, with the latter showing SO_4 and NO_3 contents which could justify a higher acidity in precipitation. Data concerning sulphur and nitrogen compounds in Rio de Janeiro city are not available, but proximity with Niteroi and similar composition with respect to other species, suggest that Niteroi data could be used as an indicator for Rio de Janeiro metropolitan region. SO_4 content in Niteroi precipitation indicates that SO_2 emission from combustion of fossil fuel is the principal precursor of acidity in this area, with NO_3 having a minor contribution. On the other hand Salvador data reflects an insignificant anthropogenic contribution.

Cubatao and Vila Parisi are characterised by an intense industrial activity, with abnormal rain water ionic content for various species. Vila Parisi precipitation presents SO_4 and NO_3 contents that could justify a lower pH than those measured, but unfavourable topographic conditions for pollutant dispersion and the variety of materials injected in the atmosphere with further interaction with rain water generate a solution so complex that pH values tend to neutrality. On one occasion after 20 rain free days, long pH values up to 9.0 were measured (Moreira - Nordmann et al., 1986).

The interior group contains the Amazon data that represent a natural condition, whereas Sao Paulo and San Jose dos Campos present acid precipitation with high values for SO_4, NO_3 and NH_4, principally at Sao Paulo, related to the diverse nature of its sources. The metropolitan regions have a strong contribution from combustion of fossil fuel, and also from a great number of industrial areas with significant emission of atmospheric pollution. SO_4 values for these two locations are underestimated in view of analytical methods used (Moreira - Nordmann et al., 1985).

The sources of elements in precipitation can be grouped into general categories of marine and non - marine (including anthropogenic). Generally a reference element is used to quantify the marine

contribution to precipitation chemistry. Data presented in Table 3 have a strong anthropogenic component that alters Cl content in precipitation collected in Cubatao and Vila Parisi, (Miller et al., 1985; apud Moreira - Nordmann et al., 1986). Sodium is also affected, but to a lesser extent than chlorine. Thus we used sodium as the reference element for evaluation of the proportion of marine source in precipitation chemistry.

In Table 4 we can observe that all ratios are above sea salt values, except calcium and chloride in the Amazon Basin, suggesting a significant contribution of a terrestrial source in precipitation chemistry. Various ratios present abnormal values, in particular those related to Vila Parisi for all chemicals, Sao Paulo for K, Ca, Mg and SO_4, San Jose dos Campos for K, Ca, Mg and Cl, Cubatao for K, Mg and SO_4 and the Amazon for SO_4. Except for the Amazon where the SO_4 ratio could be related to a great extent to natural emissions from soils and vegetation, the others reflect a strong anthropogenic contribution. The sulphur ratio shows extremely high values for Vila Parisi, Sao Paulo and Cubatao.

Table 4 : Mole Ratio between various elements and Na, compared to sea salt ; data from Table 3 used in calculations.

	Na	K	Ca	Mg	Cl	SO_4
Sea salt	1.000	0.021	0.119	0.022	1.173	0.060
Amazon	1.000	0.076	0.076	0.092	1.154	0.408
Sao Paulo	1.000	0.357	1.393	0.727	1.212	1.273
San Jose	1.000	0.743	1.628	0.486	2.086	-------
Salvador	1.000	0.073	0.277	0.120	1.337	-------
Rio de Janeiro	1.000	0.115	0.123	0.161	1.215	-------
Niteroi	1.000	0.112	-------	0.107	0.714	0.112
Santos	1.000	0.081	0.044	0.203	1.333	0.275
Cubatao	1.000	0.785	0.104	0.313	1.153	0.590
Vila Parisi	1.000	1.029	6.424	1.388	2.374	4.050

Long Distance Tranport and Acidity Deposition

Despite the importance of the long distance transport of acidity in the regionalisation of acid precipitation phenomena, little effort has been made to quantify the magnitude of this process in Brazil.

A single and superficial study of this aspect was carried out in Rio de Janeiro city by Silva Filho et al.(1987). They observed an association between events of acidity deposition in coastal forested ecosystems, and the passage of cold fronts, that originate in the south of the continent. During their pathway to the north, the fronts pass over the industrialised regions of Sao Paulo State before reaching Rio de Janeiro. Residence time in the atmosphere of between 2 to 5 days for nitrogen and sulphur oxides (Irwin & Williams, 1988; Babich et al., 1980; Likens & Borman, 1974) indicates that, besides a local contribution, long distance transport of acidity is probably occuring in Brazil's Atlantic Coast.

Silva Filho (1985) in a one year study of atmospheric inputs to a premontane forested ecosystem, showed that free acidity deposition varied weekly from 0.02 to 2.9 keq/ha/year with a mean value of 0.58 keq/ha/year. The deposition was characterised by occasional extremely high values, for example, the four weeks of higher precipitation volume were responsible for 13% of total annual flux. Comparing two collectors 1,500m distant and with 600m of elevation difference, he also noted that acidity deposition was spacialy uniform, despite fractioning observed for other elements such as chlorine, sodium and calcium.

Brown et al (1985) collected 180 precipitation samples at Tijuca National Park and reported a range of + 0.4 to + 12.5 % suggesting that despite a local component rain water suffered long distance transport before precipitation, acting as a mass transport agent for pollutants injected into the atmosphere.

Free Acidity Budget and Possibles Effects

The effect of acid precipitation on tropical ecosystems, particularly in Brazil, has not been evaluated. An outline of the possible environmental consequences of this phenomenum in Brazil, can be made by grouping data concerning Tijuca National Park in Rio de Janeiro city. By using data from Ovalle et al., 1987 , Silva Filho, 1985 and Ovalle, 1985, we can compose a free acidity budget based on pH measurements of rain and stream water in a 350 ha forested catchment.

The weekly fluxes are characterised by long periods of low values punctuated by occasional extreme values associated with precipitation pulses higher than 80mm/day.

During an 80mm storm, the values found were (eq/ha/year): atmospheric input = 11.7 and stream output = 0.55. Assuming this event as representative of the pulses > 80mm/day, and its annual frequency of occurrence along the studied period, they are responsible for 16 - 36% of annual free acidity input and > 95% of output measured.

Figure 2 shows weekly pH variations in rainwater, throughfall and stream water at Tijuca National Park. It is clear that during its course to stream channel, rainwater acidity decreases,

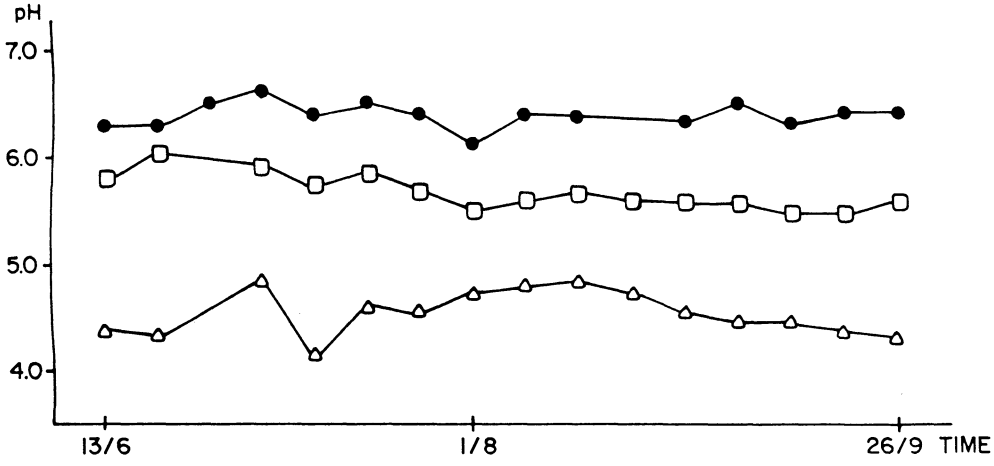

FIGURE 2 – pH weekly variation of rainwater(△), throughfall (□) and stream water (●) Tijuca National Park, Rio de Janeiro between June and September of 1984.

suggesting that this forested ecosystem acts as a natural filter. Silva Filho & Ovalle (1984) show that interaction with vegetation cover provides a pH change from 4.7 to 5.6, associated with a strong increase in water ionic content. They suggest that despite a simple washing of dry deposition from leaf surfaces, anionic exchange between rainwater and foliage could be responsible for acidity neutralisation, as calcium content in throughfall shows an increase of an order of magnitude. As forested ecosystems are associated with nutrient depleted soils, high losses of essential elements such as calcium could not be expected.

The acid nature of the soils (pH close to 4.0), the slow rate of weathering reactions (Ovalle et al., 1984), and mean pH of stream water (6.1), suggest that the neutralisation of acid precipitation at tree canopy level is an important mechanism in the filter action in these forested ecosystems.

Meguro et al. (1979) also report a similar behaviour between rainwater and throughfall in a secondary forested area at Sao Paulo city metropolitan area, suggesting that Brazilian coastal forested ecosystems may act as a natural filter for acid precipitation. The cost of this mechanism for forest nutrient cycling and consequently productivity has not been evaluated.

CONCLUSION

At present acid precipitation research in Brazil is in an initial stage, with data concerning acid precipitation being by - products of institutional research programmes. With the identification of the

problem, appropriate research programmes are being formulated.

The major problem at this stage is related to the techniques of collection, preservation and analysis of rainwater samples that must be standardised in order to permit a regional approach to precipitation chemistry research in Brazil, and avoiding dispersion of efforts incomparable data, and optimising the often limited financial support.

In this context a programme was initiated in 1988 involving 6 different research centres distributed along the Brazilian coast, using the methodology suggested by Galloway and Likens (1978), and now used in a worldwide programme of precipitation chemistry. The starting point is to analyse the extent of the problem, and to create an interchange between emergency research groups and experienced ones.

In terms of environmental impacts the absence of data is almost total. The absence of long - term monitoring programmes investigating the dynamics of the diverse ecosystems existing in Brazil does not permit a realistic evaluation of the impacts. As acid precipitation effects are long - term, and although the effects would be small for some ecosystems they must be seen as additional added stress to ecosystems. The effects in most cases are unrecognised and often confounded by other sources of pollution (Likens & Borman, 1974). Other critical environmental problems such as the rate of forest clear - cutting turn studies of acid precipitation - vegetation interaction into a paleo - ecological exercise. Industrial pollution and cultural euthrophication of surface waters, make impacts of acid rain on aquatic resources a minor part of the complex environmental problems in Brazil.

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