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τίτυιο/τιτιε	PUBLICAÇÃO Nº PUBLICATION NO INPE-4062-PRE/1020 CHEMICAL COMPOSITION OF RAINWATER IN THE ANTARTIC PENINSULA	ORIGEM ORIGIN DME / DAM PROJETO PROJECT PROANTAR Nº DE PAG. ULTIMA PAG. LAST PAGE 26 VERSÃO Nº DE MAPAS NO OF MAPS
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RESUMO - NOTAS / ABSTRACT - NOTES

Rain water and fresh snow samples were collected at King George Island (Com. Ferraz Station), Northern Antarctic Peninsula, during the 1985 summer. Na † , Ca 2† , K † , Mg 2† , Cl $^{-}$, SO $_{+}^{2}$, and NO $_{3}$ were analysed in these samples. Ionic concentrations varied from hundreds to a few tenths of ppms. The results showed marine influence in the concentration of elements and compounds, and also in the ionic ratios determined. This influence was expected since the sampling station was located near the coast and close to sea level. The composition of rain water reflected marine composition for all elements, Ca 2† and K † included, the only exception being NO $_{3}$ which showed distinct results, as noted in other papers related to Antarctic snow sampling. An excess of about 20% in SO $_{+}^{2}$ concentration was also found in the samples.

— OBSERVAÇÕES/REMARKS

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Abstract

Rain water and fresh snow samples were collected at King George Island (Com. Ferraz Station), Northern Antarctic Peninsula, during the 1985 summer. Na^+ , $\mathrm{Ca^2}^+$, K^+ , $\mathrm{Mg^2}^+$, $\mathrm{C1}^-$, $\mathrm{S0_4^2}^-$, and $\mathrm{NO_3^-}$ were analysed in these samples. Ionic concentrations varied from hundreds to a few tenths of ppms. The results showed marine influence in the concentration of elements and compounds, and also in the ionic ratios determined. This influence was expected since the sampling station was located near the coast and close to sea level. The composition of rain water reflected marine composition for all elements, $\mathrm{Ca^2}^+$ and K^+ included, the only exception being $\mathrm{NO_3^-}$ which showed distinct results, as noted in other papers related to Antarctic snow sampling. An excess of about 20% in $\mathrm{SO_4^2}^-$ concentration was also found in the samples.

Introduction

Increase in emissions of atmospheric pollutants and related changes in atmospheric chemical composition have stimulated scientific work on trace gases, particulate matter and rain water even in remote regions of the world (Galloway et al., 1982; Galloway and Gaudy, 1984). Antarctica, with almost 14x10⁶Km² covered by snow and ice, is a remote region which, in principle, can provide chemical data of an atmospheric environment considered virtually with no anthropogenic effect due to greatdistances from pollution sources ,and also because of the few scattered stations and bases in its territory which produce little contamination. These conditions led to studies of aerosols and glacial chemistry with analysis of recent and old layers of snow (Nguyen et al., 1974, Hogan and Barnard, 1978, Maenhaut and Zoller, 1979, Ono et al., 1981, and Bouton and Lorius, 1979, Boutron, 1980, Boutron and Leclerc, 1984, Delmas et al., 1982, Legrand and Delmas, 1984, Murozomi et al., 1969, Warburton et al., 1980, among others).

Studies performed in ice cores tend to correlate results of chemical composition of snow with the atmospheric conditions at the time of precipitation (e.g., Lorius et al, 1985). This subject is still under consideration according to some authors (Legrand and Delmas, 1984). They have shown that a Cl-excess exists in snow,

possibly due to HCl emissions ,and that maximum values of SO_4^2 and NO_3^- were observed during summertime although their occurence pattern is not synchronous. Delmas et al. (1982) also found Cl⁻, SO_4^2 and NO_3^- in excess, what was explained as being also from gas derived origin and not only marine. Boutron and Leclerc (1984) observed that concentrations of 22 elements and compounds in a ice block more then 1200 years old were similar to those found in recent precipitation and suggested that the South Pole area had little effect of modern global pollution. All the above quoted authors acknowledge that influences in snow composition from continental origin (soils, e.g.) can be neglected.

It is also important to notice that the first step towards an understanding of the chemistry of continental snow is the study of precipitation at sea shores (Delmas et al., 1982) and that very limited data (if any at all) exist for Antarctica. In this study the chemical composition of fifteen samples of rain water collected during the 1985 summer at the Brazilian station Com. Ferraz, King George Island, is presented.

Sampling Environment and Methods

The Antarctic continent, with $14 \times 10^6 \, \mathrm{km}^2$, covering latitudes from 90°S to 60°S in all longitudes, and with an average elevation of 2,300m, presents very distinct meteorological characterisitics. In the case of precipitation, the amounts vary from those typical of deserts, as in the central plateau, to the 2,320mm/year registered at the North of the Antarctic Peninsula (Schwerdtfeger, 1984).

In general, the study of liquid precipitation in Antarctica is possible at the Peninsula. In the continent itself low temperatures produce snow or ice cristals. At Estação Comandante Ferraz (ECF) — see map on figure 1, King George Island, South Shetland Islands, NW of the Northern tip of the Antarctic Peninsula (62°05'S, 058°23'W), environmental conditions are different. Mean annual temperature at ECF during 1948—1960 was —2,5°C; from December through March monthly means were always above 0°C, reaching +1,5°C in February (Schwerdtfeger, 1984). Therefore, at least for four months in the year such temperatures favor liquid precipitation, which is associated with relatively warm and humid air masses from NW. Precipitation data for the ECF region is presented in Table 1.

During February 1985, which covers most of the samples collected for this study, the total rainfall at ECF registered in 18 days was 31,8mm (Diniz, 1985) with a maximum of 9,9mm in one single day. In seven of these days light snow was mixed with rain and it melted in contact with the protective screen of the collector surface. The collecting period was considered a little dryer than the average, but not excessively. It is also important to remember that rain at ECF is normally originated from cold frontal and low pressure systems, associated with strong and gusty winds which always introduce significant errors in rainfall measurements. Average wind speed was 5,2m/s and gusts of 20m/s were frequent. Easterly winds prevailed during 33% of the period. Southerly winds were the least frequent, with 6% of persistence. The mean temperature for February 1985, was 2,0°C with a minimum recorded of -5,2°C, both within expected values for the season.

The rain collector used was an acrylic hemisphere with 0,23m of collecting surface covered by a nylon screen. A polyethylene hose connected the bottom of the bowl to a polyethylene bottle which was replaced after each precipitation event. The bowl was attached to a tripod 1,20m above the ground. The collector was placed at about 20m above sea level and 80m from the shore. A meteorological station which provided precipitation, wind and temperature data was located at the same place. Before the

experiment all parts were washed with detergent, tap water, 50% Nitric Acid and deionized water, and were stored in plastic bags to avoid contamination. The rain water samples were protected from light and stored at about 2 C in a refrigerator. They were transported from ECF to INPE's laboratory (São José dos Campos, SP, Brazil) inside coolers with no significant temperature change during the 48hs trip. The cations (Na⁺, Ca²⁺, K⁺and Mg²⁺) were determined using a Varian Atomic Absorption Spectrophotometer (AA1475). Anions (SO²₄, NO₃, and Cl⁻) were measured with a Dionex Ion Chromatograph (2010i). Potentiometry was also used to measure Cl⁻ concentrations. Data are accurate and precise to 10%. The sampling period extended from January 28, to February 20, 1985. The minimum precipitation registered was 0,1mm, for sample 16, and the maximum, 1,2mm for sequential samples 14 and 15. Light snow was observed during sapis 5, 16 and 17.

Discussion of Results

Figure 2 shows the precipitation distribution records during the sampling period and the sample identification numbers (Si, i=1 to 17). Rain data for sample S13 was not registered. Table 2 presents extremes, means and standard deviations of concentration values for the anions and cations analysed. The results obtained for each sample are shown in Figure 3. It may be noticed in this

figure that the highest concentrations were determined in sample S1, obtained after three days without rain (seeFigure 2), except for NO, which showed a maximum in S3.

Samples S1, S3, and S4 correspond to a total rainfall of 2.2mm. S1, the initial precipitation, accounts for 0.7mm and showed Na^+ , Mg^{2^+} and SO_4^2 concentrations corresponding to 87% of the total contents of these ions when the three samples are considered together. For CI^- and NO_3^- , the percentages were 90% and 36%, respectively. In S3 and S4, concentrations were about 10% of the total contents obtained in the three samples for CI^- and NO_4^+ , 12% for Mg^{2^+} and $\mathrm{SO}_4^{2^-}$, and 6% for NO_3^- . Therefore, from these values, one may conclude that as rainfall progresses an important effect of dilution of ion concentrations is observed.

A second maximum was obtained after two days without precipitation, for sample S5 - see Figures 2 and 3. For the four samples amounting to 11,8mm and collected during 3 days, the initial 0,7mm showed a concentration of 68% for the total of Na⁺, Mg^{2+} and NO_3 -contents, 66% of C1⁻, and 71% of SO_4^{2-} . Considering the uncertainity of the measurements, it is possible to infer that for the above two sampling sequences the initial 0,7mm contributed to about 90% of the total ion concentrations (except for NO_3^-) in the first one, and 70% for the second one.

The maximum values found are very high, particularly for Cl⁻ and Na⁺, which are even higher than those found by Buat-Ménard (1970) for rain samples in the North-Atlantic ocean. These values are also higher than typical values for the Brazilian coast (Tavares et al., 1983, Ferreira and Moreira-Nodemann, 1985) and for other remote regions of the globe (Galloway et al., 1982).

On the other hand, minimum concentrations were registered in S15, with NO_3^- as an exception again. This sample amounted to 7,1mm, gathered after six consecutive rainy days, what explains the lower concentrations determined and indicates the occurrence of the washout effect in the atmosphere.

Table 2 also presents volume weighted means, arithmetic means and standard deviations for each ion analyzed. Apparently the wide variation in concentrations and also the relative small number of samples resulted in large values for the standard deviations. These variations may also explain the difference between arithmetic and weighted means. For this analysis the weighted mean seem to be a better representation than the arithmetic mean since it reduces the effect of the extremes.

The volume weighted means are also high, and even higher than in results from continental snow obtained before (Legrand and Delmas, 1984, Delmas et al., 1982, Warburton et al., 1980, and

others).

From the mean values of Table 2, in $\mu eq/1$, we have:

$$[Na^{+}] + [Ca^{2+}] + [K^{+}] + [Mg^{2+}] = 1,238 \mu eq/1$$

$$[Cl^{-}] + [SO^{2}] + [NO_{3}] = 1,311 \mu eq/1$$

The excess of 0,073 μ eq/l, resulting from the difference of the anion and cation concentrations, can be explained either by uncertainty in the measurements or by the lack of H and NH results. We also notice that among cations, Na corresponds to 78% of the total, and Mg²⁺, to 18%. Among anions, Cl⁻ represents 87% of the total, SO_4^{2-} to 10%, and 2% to NO_3^{-} .

However, if excess Sulfate and Chloride are calculated from the following known relations,

$$[SO_{4 \text{ ex}}^{2}] = [SO_{4}^{2}] - 0,12 [Na^{+}], \text{ and}$$

$$[C1^-]_{ex} = [C1^-] - 1,17 [Na^+],$$

using the weighted means, an excess of 22,9 μ eq/l results for SO_4^2 (1,09ppm), and 30 μ eq/l for CT (1,06ppm). In the case of SO_4^{2-} this

determined value represents an excess of 16% above the uncertainty of the measurements (10%). In the case of Cl⁻, the excess is less than 3%, within expected variations.

Calculations of the enrichment factor, E, was determined from:

E = (X/R) rain water/(X/R) sea water, where

(X/R) sea water is the ratio $Cl^-/Na^+ = 1.8$, and also $SO_4^{2-}/Na^+ = 0.25$, for sea water, and (X/R) rain water refers to the same elements for rain water.

For Cl $^-$ a value close to unit was estimated, 1,03 for the arithmetic mean and 1,08 for the weighted mean, indicating that no excess exists in the rain. Regarding SO_4^2 , and using the same process as above, 1,5 and 1,2 were calculted, respectively. These values also agree with previous conclusions whereupon: a slight excess of non maritime origin SO_4^2 exists in Antarctic rain.

The marine influence for all ions but NO_3^- is clear when Figure 3 is examined. Such effect was expected since the sampling site was close to the shore. The fact that all maximum values were found in S1, and that minimum values in S15 also corroborates this interpretation.

Table 3 shows ionic ratios obtained from the arithmetic means and respective standard deviations using Na⁺ as reference element. This table also shows the ionic ratio calculted from the weighted means (V.W.M.). The same ratios in sea water, from literature, are given for comparison. Ionic ratios are close to those of ocean results (except for $\mathrm{SO_4^2-/Na^+}$), although with relatively high standard deviations due to the small number of samples collected. The weighted means of the Ca²⁺/Na⁺ ratio were smaller than values for the ocean. From ionic ratios in Table 3 we also notice that the contribution of terrestrial origin is negligible. For ionic ratios calculted with weighted means the SO_4^2 -/Na relation was 20% higher than typical ocean values; this also confirms the 50_4^{2-} enrichment in the precipitation. Galloway and Gaudry (1984) also found a 20% excess in the ${\rm SO}_4^{2-}/{\rm Na}^+$ ratio in the Indian Ocean. Some authors (Delmas et al., 1982, and Legrand and Delmas, 1984, e.g.) suggested the effect of other sources based on the ${\rm Cl}^-$ and ${\rm SO}_4^{2-}$ excesses, but this work can only confirm a slight excess of SO_4^2 . No evidence of Cl excess was found in our samples.

Mean values for NO_3^- were high and above those of other regions (Galloway et a., 1982). As it can be seen in Figure 3 this ion shows a distinct pattern from the others and any influence from the sea is highly improbable. It is also known that NO_3^- presence in

oceans is negligible. NO_3^- excess in continental Antartic snow was found by Delmas et al.(1982), Legrand and Delmas(1984), and Parker and Zeller(1982), who also neglected any marine origin. It has been suggested that NO_3^- formation could be possibly tied up to solar activity, being produced in the high troposphere or stratosphere (Parker et al., 1978). Our results confirm an NO_3^- excess in Antarctic rainfall and that this ion does not seem to be of marine origin. Pollution sources are probably also out of question following the recent work of Boutron and Leclerc (1984), where many elements and compounds were determined in pre-historic Antarctic ice (including NH_4^+ , but not NO_3^-) showing that the Southern Hemisphere is still little affected by global pollution, including Sulfur compounds. Further work is necessary in order to reach any conclusion about the sources of NO_3^- found in the Antarctic atmosphere

Conclusions

Ions in Antarctic rain samples presented very high concentrations for maximum values and for weighted means. Such levels are higher than those of other remote regions of the globe and of continental snow in the South Pole.

Because the sampling site was close to the shore the ionic

composition of the rain reflects that of sea water, except for NO_3^- A slight excess of SO_4^{2-} was determined. Among the cations, Na^+ and Mg^{2+} represented 78% and 18% of the total mass, respectively. For the anions, Cl^- amounts to 87% of the total, SO_4^{2-} 10%, and NO_3^- 2%. Ionic ratios for Ca^{2+} and K^+ in relation to Na^+ showed that any contribution from the continent was negligible.

The calculation of the the enrichement factor and Cl⁻excess has indicated that no excess of this ion exists; ionic ratios also corroborated this conclusion. A 20% excess of SO_4^{2+} was found in the rain samples. A relatively high NO_3^- excess and SO_4^{2-} enrichment determined in the samples appear to originate from other sources and not from the sea. Due to the non-synchronous behavior of these ions in rain water, this sources may not be the same.

Rain sampling at ECF was continued during the 1986 summer and snow samples will be collected during the winter. Such additional samples will probably help to interpret the ${\rm NO_3}^-$ and ${\rm SO_4^2}^-$ presence in Antarctic precipitation.

Acknowledgments

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TABLE 1. Precipitation data in mm for the ECF region (Schwertfeger, 1970: Arctowski, 1978/9)

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indre i. Frecipication data in mm lor the Eur region (Schwertieger, 1970; Arctowski, 1978/9)	LOCATION	Deception Island 62 59'S,60 43'W (150Km S of ECF)	King George Island 1978– Arctowski Station (8Km S of ECF) 1979

volume-weghted means of the composition of 15 precipitation events; TABLE 2. Minima, maxima, arithmetic means, standard deviations and values in mg/1, or as stated.

	Na+	Ca ²⁺	**************************************	ca^{2+} K^{+} Mg^{2+}	C1_	so2-	NO3
Maximum	520.00	14.00	21.00	55.00	912.00	123.00 12.50	12.50
Minimum	0.22	0.04	0.002	0.08	0.53	0.14 0.013	0.013
Mean, Arith.	51.36	1.24		5.88	92.64	14.08	3.04
Std.Deviat.	130.87	3,65	5.31	13.80	228.97	30.77	5.50
V.W.M.	22.09	0.52	0.81	2.67	40.91	6.64	1.70
V.W.M., ueq/1 971.96	971.96	26.00	21.06	218.94	21.06 218.94 1145.48	139.44	27.2

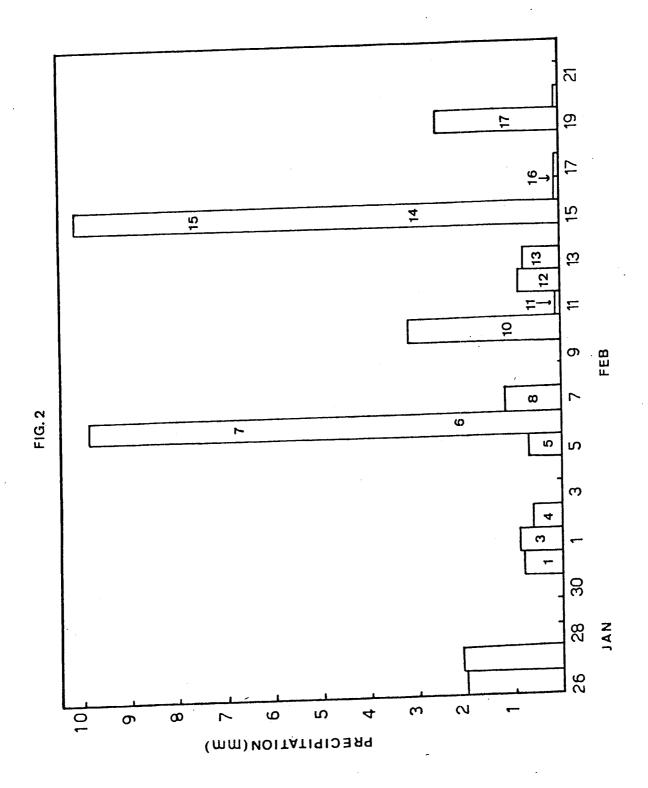
TABLE 3. Ionic ratios for sea-water, rain water and volume weighted means(V.W.M.).

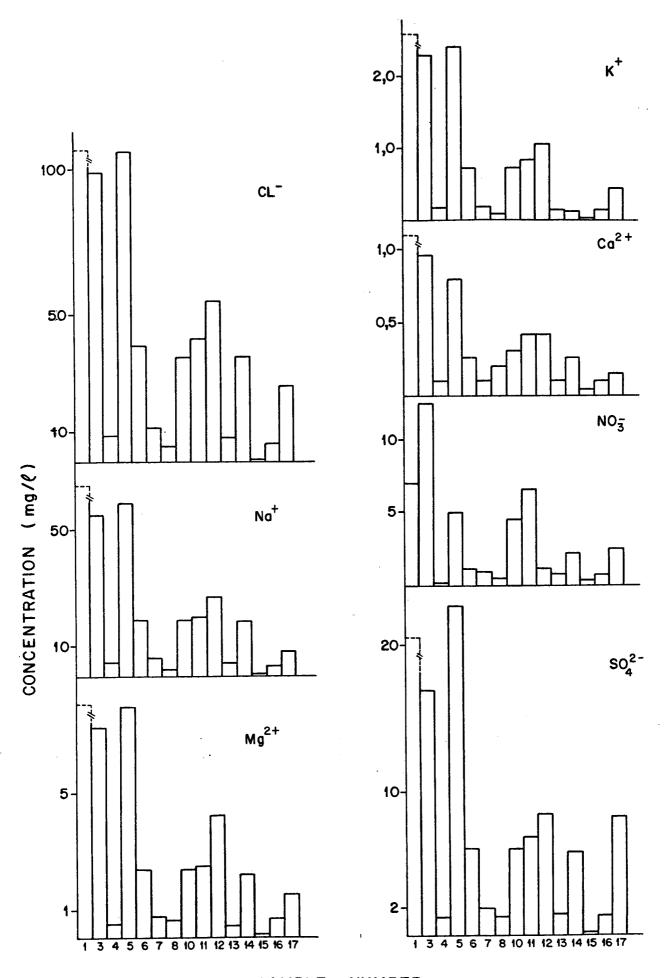
Figure Captions

Figure 1. Location of the Comandante Ferraz Station.

Figure 2. Rainfall distribution for the sampling period.

Figure 3. Ionic concentrations in mg/l for each sample.





SAMPLE NUMBER

L.M.Moreira-Nordeman

Chemical Composition of Rain in the Antarctic Peninsula Figure 1.

L.M.Moreira-Nordeman

Chemical Composition of Rain in the Antarctic Peninsula Figure 2.

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Chemical Composition of Rain in the Antarctic Peninsula Figure 3.

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