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Chemical characterization of fog and rain water collected at the eastern Andes cordillera

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Abstract

During a three month period in 2003 and 2004, the chemistry of fog and rainwater were studied at the "El Tiro" site in a tropical mountain forest ecosystem in Ecuador, South America. The fogwater samples were collected using a passive fog collector, and for the rain water, a standard rain sampler was employed. For all samples, electric conductivity, pH, and the concentrations of NH_4^+, K^+, Na^+, Ca^{2+}, Mg^{2+}, Cl^-, NO_3^-, PO_4^{3-}, and SO_4^{2-} were measured. For each fog sample, a 5 day back trajectory was calculated by the use of the HYSPLIT model. Two types of trajectories occurred. One type was characterized by advection of air masses from the East over the Amazonian basin, the other trajectory arrived one from the West after significant travel time over the Pacific Ocean.

We found considerably higher ion concentrations in fogwater samples than in rain samples. Median pH values are 4.58 for fog water, and 5.26 for the rain samples, respectively. The median electric conductivity was 23 μS cm^{-1} for the fog and 6 μS cm^{-1} for the rain. The concentrations of all analysed ions were relatively low compared to other mountainous sites (Weathers et al., 1988; Elias et al., 1995; Schemenauer et al., 1995; Wrzesinsky and Klemm, 2000; Zimmermann and Zimmermann, 2002). The continent samples exhibit higher concentrations of most ions as compared to the pacific samples.

1. Introduction

Investigations of fog chemistry and cloud physics have become very important during the last decades. Nutrients and pollutants from fog and cloud water exhibit strong influence on ecosystems (Fisak et al., 2002). Especially in mountain forest ecosystems, where fog occurs very frequently, fog water is an important source of ion deposition (Gordon et al., 1994; Walmsley et al., 1996). The frequency of fog events typically increases with altitude above sea level (Olivier and de Rautenbach, 2002, Zimmermann

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and Zimmermann, 2002; Holder, 2004). Prevalent fog frequencies and high wind velocities cause more fog deposition and ion input at elevated mountainous sites than in lowlands (Minami and Ishizaka, 1996; Walmsley et al., 1996; Clark et al., 1998; Igawa et al., 2002).

The scope of this study was to describe the chemical composition of fog and rain water collected in Loja/Ecuador at the eastern Andes cordillera. A characterisation of the fog and rain chemistry of this region is not available yet. The study site is within the inner tropical convergence zone (ITC), and therefore, air masses arriving from various directions represent very different advection regimes.

2. Methods

2.1. Site description

The fog and rainwater samples were collected from November 2003 through February 2004. The collectors were established at the "El Tiro" meteorological station at 2825 m a.s.l. (3°59'45" S and 79°09'38" W), which is located in the Podocarpus National Park in southern Ecuador, South America. The sampling site is situated in the Andes highlands at the eastern Andes cordillera. The tropical climate is characterized by marginal annual, but pronounced daily, temperature fluctuations. The mean annual temperature at the "El Tiro" station is 10°C, and the annual precipitation is about 1500 mm. The wind regime at the sampling site is influenced by strong easterly trade winds (mean: 14 m s^{-1}), with superposition of regional mountain wind systems. The frequency of fog events is very high. Winds from the Amazonian lowlands advect warm and moist air masses against the Andes massif and force them to ascent and cool down adiabatically.

2.2. Fog and rain water collection

The fog water was sampled by using a Kroneis[1] passive fogwater collector (Fig. 1). This cylindrical string collector has an effective collection surface of 0.09 m^2 (diameter (0.21 m) × height (0.45 m)). The fog droplets impact on the vertical Teflon strings, combine to larger drops, run down the strings, and drip into a 500 mL Polyethylene bottle. A roof prevented rain from reaching the fog sample. The impaction of the fog droplets on the strings simulates the deposition on natural surfaces. The 460 strings are arranged cylindrically, so that the collection efficiency does not depend on the wind direction. A disadvantage of passive fog collectors is the lack of quantitative comparability of the collected fog water volumes, because there is no information about the air volume moving through the collector. Measurements of the liquid water content (LWC) were not possible. The fog collector was thoroughly cleaned once per week with de-ionized water.

The rainwater collector (Fig. 2) is a standard collector for chemical analyses, fabricated by UMS[2]. The collection surface is 314 cm^2, and the collection bottle has a volume of 0.5 L.

The fog samples were collected on a daily basis and the rain samples were taken once per week. For sample collection, the fog and rain water was taken from the respective collection bottle, quantified, and filled into 50 mL PE storage bottles. Additionally, sample aliquots for immediate pH and conductivity analyses and field blank samples were collected. All the samples were stored deep-frozen until chemical analysis in the laboratory at the University of Münster, Germany.

[1] Kroneis GmbH, Igelgasse 30-32, 1990 Wien, Austria.

[2] UMS GmbH, Gmunder Str. 37, 81379 München, Germany.

2.3. Trajectories

A common method for the identification of the origin and the pathway of air masses is to calculate backward trajectories. Trajectories are often used to study the advection of air pollutants and the coherency of the origin of air masses and their respective pollutant concentrations (Klemm et al., 1994). Air masses pick up pollutants during their travel route so that they are influenced by the geographical origin and travel path. In this study, for every fog sample a back trajectory was computed using the HYSPLIT model (Draxler and Rolph, 2003; Rolph, 2003). Each trajectory indicates the path of the respective air mass during the last 120 hours at 1 m above surface level before reaching the fog collector.

2.4. Chemical analysis

All fog and rain samples were measured for pH, electric conductivity, and the major ions NH_4^+, K^+, Na^+, Ca^{2+}, Mg^{2+}, Cl^-, NO_3^-, PO_4^{3-}, and SO_4^{2-}. The pH measurements were performed with a pH electrode (WTW pH 323) and the electric conductivity with a portable conductivity meter (WTW LF 315) in separate sample aliquots. The anions Cl^-, NO_3^-, PO_4^{3-}, SO_4^{2-} were measured by ion chromatography (DIONEX DX 100). Ca^{2+} and Mg^{2+} were determined by flame atomic absorption spectrometry (AAS). K^+ and Na^+ were measured by flame photometry, and NH_4^+ by a photometric method (Aquatec Analyser).

For the control of the quality of the analytical results, the measured and calculated electric conductivities were compared. For every fog and rain sample, the calculated conductivity x_{calc} [μS cm^{-1}] was determined by summing up the products of the specific conductivities $x_{spec,i}$ [μS cm^{-1}] and the equivalent concentrations $c_{eq,i}$ of the ions i [μeq L^{-1}]:

$$x_{calc} = \sum_i x_{spec,i} \cdot c_{eq,i}. \quad (1)$$

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Further, the ion balance was computed by summing up the equivalent concentrations of cations $i+$ and anions $i-$ of the samples. The sum of anion equivalent concentrations should be equal that of the cations according to the condition of neutrality:

$$\sum_i c_{eq,i+} = \sum_i c_{eq,i-} \quad (2)$$

3. Results

A list of all collected samples during the sampling period from November 2003 through February 2004 is given in Table 1. Three fog samples, which were collected after extended non-foggy conditions, had to be excluded from further analysis. These samples contained much higher ion concentrations that are associated with dry deposition of aerosol particles during extended dry periods on the fog collector surface before fog collection.

3.1. Origin of the air masses

By the use of the HYSPLIT model, for every fog sample, a 120 h back trajectory was calculated (Fig. 3). On the basis of these trajectories, the samples were divided into two classes. Figure 3 shows that the separation into these two groups is unequivocal. 48 fog samples were classified as "continent" samples and 8 as "pacific" samples, respectively. The continent samples originated from the Amazon basin and reached the sampling site from the East. The pathway of the continent samples is associated with south-easterly trade winds. The pacific samples travelled along the west coast of the South American continent in north-westerly direction (southeasterly trade winds as well) and turned to their right hand side before reaching the sampling site. The turnaround is suggested to result from local wind systems. The classification of the trajectories leads to the hypothesis that systematic differences between these two groups should exist concerning their chemical composition.

3.2. Chemical composition of fog water

Table 2 shows the pH, the electric conductivity, and the median concentrations of the 56 fog samples. The dominating ions are the hydrogen ions (H^+), ammonium (NH_4^+), nitrate (NO_3^-), and sulphate (SO_4^{2-}). For the fog samples, the median pH is 4.58 and the median electric conductivity is 23 μS cm^{-1}. The minimums and the maximum values of the measured data vary considerably. The minimum pH is 3.91 whereas die maximum pH is 5.61. For the conductivity and most of the measured ions, the picture is similar. Overall, the data set is very heterogeneous. The median total ion equivalent concentration is 139.4 μeq L^{-1} for the fog samples.

For the comparison of the cations and anions, an ion balance was calculated. The median of the equivalent sum of the cations (82.8 μeq L^{-1}) is larger than the respective median sum of the anions (56.6 μeq L^{-1}).

3.3. Comparison of continent and pacific samples

The results of the chemical analysis were tested statistically. A Kolmogorov-Smirnov-test leads to rejection of the hypothesis that the fog and rain data sets were normally distributed. Therefore the mean values are not further discussed. Thus, the non-parametric Bootstrap-Resampling-Method was applied to produce confidence intervals of the medians of ion concentrations. According to the statistical analysis, differences between the continent and the pacific samples do exist, but are not significant.

For the two groups (the continent and the pacific samples), ammonium, sulphate, nitrate, and the hydrogen ions are the dominating ions (Tables 3 and 4). In general, the continent samples contain higher ion concentrations (139 μeq L^{-1}) than the pacific samples (111 μeq L^{-1}). The median pH is 4.55 for the continent and 4.93 for the pacific samples. The chemical composition of the continent samples is more heterogeneous than for the pacific samples. The minimum and maximum values for the pH, the electric conductivity and for the measured ions diverge more for the continent samples. Tables 3 and 4 show the pH, the electric conductivity [μS cm^{-1}], and the median

concentrations [μeq L^{-1}] for the continent and the pacific samples.

3.4. Chemical composition of rain water

Table 5 shows the pH, the electric conductivity, and the median concentrations of the 11 rain samples. For the rain samples, SO_4^{2-} and H^+ are the dominating ions. The nitrogen compounds, NH_4^+ and NO_3^-, play only minor roles. The median value for NH_4^+ was even below the limit of quantification of 0.04 mg L^{-1}. The pH varies from 4.54 to 5.61 and the electric conductivity from 3 μS cm^{-1} to 11 μS cm^{-1}. The total median ion equivalent concentration of the rain samples is 34.4 μeq L^{-1}. The sum of the cations is 18.4 μeq L^{-1} and the sum of the anions is 16.0 μeq L^{-1}.

3.5. Comparison of fog and rain water

The chemical composition of the fog and rain samples shows pronounced differences. In Fig. 4 the median equivalent concentration of the fog and the rain samples are compared. The error bars indicate the 75% and 25% percentiles.

The total ion equivalent concentration of the fog samples is 139 μeq L^{-1} and 34.4 μeq L^{-1} for the rain samples, respectively. The median electric conductivity is about 4 times higher for the fog samples than for the rain samples. The largest differences concerning the ion composition exist for H^+, NH_4^+, NO_3^-, and SO_4^{2-}. The median concentrations of the H^+ and SO_4^{2-} are about 4 times higher in fog than in rain water. For NH_4^+ and NO_3^-, the ratios are even higher. NH_4^+ contributes 22% to the total concentration in the fog sample and is negligible in the rain samples. For the contents of PO_4^{3-}, the situation is different: the median concentration is under the detection limit for the fog samples whereas the median equivalent concentration for the rain samples is 2.4 μeq L^{-1}. The PO_4^{3-} ions amount 7% of the total median equivalent concentration in the rain samples.

Table 6 shows the contribution of the measured ions in the fog and rain samples. H^+,

NH_4^+, NO_3^-, and SO_4^{2-} dominate in the fog water. The chemical composition of the fog samples is more heterogenic than for the rain samples. The minimum and maximum values of the pH, electric conductivity, and the ion concentrations diverge more for the fog than for the rain.

4. Discussion

The analysis of the fog samples collected at the "El Tiro" site shows that there exist two classes of air mass trajectories, which are well distinguished from each other. One class represents 80% of the collected samples during the sampling period between November 2003 and February 2004. Here, the air masses travelled over the Amazonian basin before reaching the sampling site. In the other group, the air masses travel along the South American west coast over the Pacific Ocean and the coastal belt. The differences of chemical composition between continent and pacific fog water samples could not be detected as expected. There exist differences, but due to the heterogeneity of the data sets, these differences are not statistically significant. The sampling site is located about 100 km from the coast. Therefore, fog of both groups travelled over landmass on their way to the fog collector, and the pacific samples are continentally influenced as well. Therefore, a clear chemical classification of maritime and continentally influenced fog at the sampling site "El Tiro" is not possible.

Due to limited instrumentation, the liquid water content (LWC) could not be determined. Presumably, systematic differences of the LWC between the data groups might have contributed to the concentration levels by means of dilution effects.

The chemical characterization of the fog and rain samples shows distinctive differences concerning the ion composition. The most pronounced differences between the fog and the rain water exist for H^+, NH_4^+, NO_3^-, and SO_4^{2-}. These differences may result from the height of formation of the droplets as described in other studies (e.g. Bridges et al., 2002). Fog represents lower layers of the atmosphere which are stronger influenced by continental emissions. On the other hand, rain droplets, are formed at

higher altitudes where the atmosphere is less loaded with compounds originating from ground-based emissions (Bridges et al., 2002). The ions H^+, NH_4^+, NO_3^-, and SO_4^{2-}, which are the dominating ions in the fog water, are indicators for anthropogenic influence. Differences concerning the homogeneity of the fog and rain samples also may results from the characteristics of the formation. Biomass burning, industry, and traffic are suggested to be the main sources for the nitrogen compounds in the fog water. However, for the comparison of the fog and rain samples, many aspects have to be considered. The fog samples were taken on a daily basis, and the rain samples only once per week. For this reason, the rain samples represent longer time periods. Modification effects such as mixture of more concentrated with less concentrated rainwater can not be ruled out. Another reason for the differences of the chemical characteristics between the fog and rain water may be the size of the droplets. Rain drops are much larger than fog droplets and may be more diluted solutions than the fog drops. A further difference between the fog and the rain water is the unbalanced ion balance in the fog water with cations predominating over the anions. This cation surplus may result from organic acids. During the field work from November 2003 and February 2004, biomass burning was observed very close to the sampling site. Organic emissions of formic and acetic acids that could not be detected in the laboratory of the University of Münster are suggested to have caused the apparent anion deficit in the fog samples analyses. The change of land use from native forest to pasture associated with biomass burning account for an elevated ion deposition in the tropics (Keller et al., 1991). Particularly during the dry period from October through March, local emissions are transported on a regional scale and deposit on the receptor surface before they get washed out (Lanfer, 2003). In contrast, during the rainy season, local emissions rain out directly so that there is not much long-range transport. Hydrogen carbonate, HCO_3^-, which has been shown to be an important ion in tropical fogs (Schemenauer and Cereceda, 1992), may also contribute to the ion balance of the poorly buffered samples with pHs around 5. The predominance of the ions H^+, NH_4^+, NO_3^-, and SO_4^{2-} in the fog water may result from an anthropogenic influence at the sampling site. The sampling site “El Tiro” is

situated about 10 km to the east of the city Loja.

5. Conclusions

A fog water and rain water study has been conducted in a tropical mountain forest in southern Ecuador from November 2003 through February 2004. This paper presents the results of a chemical characterization of the collected fog and rain samples. The comparison of continent and the pacific fog arrived at the result that the differences are not statistically significant. The comparison of the fog and rain samples presents systematic differences concerning the pH, the electric conductivity, and most ions, respectively. The most pronounced differences exist for the virtual absence of ammonium and nitrate in rain water.

The location of the sampling site and limited instrumentation are suggested to be limiting factors for our measurements. For an unequivocal chemical classification into continent and pacific fog, the sampling site should be located closer to the Pacific Ocean to exclude any anthropogenic influence. Therefore, the "El Tiro" site is not suitable for such measurements. Due to limited instrumentation, we did not quantify the liquid water content (LWC). Therefore we can not rule out that the continent fog hold systematically less LWC and thus higher liquid water concentrations of ions. Knowledge about the LWC is important to compare our data with data from other studies. Furthermore, information about the droplet size distribution would help to interpret our data set.

The cation surplus may result from organic acids such as formic and acetic acids that were not determined. Biomass burning is suggested to be a possible source of these emissions.

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Table 1. Collected water samples during the field period from November 2003 through February 2004.

	fog water	rain water	blank
samples	59	11	6
duplicate samples	11	2	

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Table 2. Electric conductivity (μS cm^{-1}), and concentrations (μeq L^{-1}) of all the fog samples.

n=56	median	avg	σ	min	max
pH	4.58	4.46	0.41	3.91	5.61
conductivity	23	31	26	3	95
calc. Conductivity	18	27	24	2	92
H^+	26.3	34.9	28.4	2.5	123
NH_4^+	30.6	65.0	83.9	0.0	305
K^+	6.9	12.3	13.6	0.0	50.7
Na^+	8.6	19.5	25.0	0.0	129
Ca^{2+}	8.0	13.6	13.7	2.0	57.4
Mg^{2+}	2.3	6.1	8.8	0.0	43.9
Cl^-	5.5	9.1	8.8	0.9	36.1
NO_3^-	20.2	40.1	50.6	1.2	215
PO_4^{3-}	0.0	1.7	2.8	0.0	11.2
SO_4^{2-}	30.9	43.9	43.4	1.2	176
$\sum$ cations	82.8				
$\sum$ anions	56.6				

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Table 3. pH, electric conductivity (μS cm^{-1}), and concentrations (μeq L^{-1}) of the continent samples.

n=48	median	avg	σ	min	max
pH	4.55	4.42	0.41	3.91	5.61
conductivity	25	32	26	3	95
calc. conductivity	20	28	25	2	92
H^+	28.5	37.8	29.2	2.5	123
NH_4^+	28.1	64.2	82.4	0.0	305
K^+	6.9	12.5	14.2	0.0	50.7
Na^+	8.6	19.8	25.7	0.0	129
Ca^{2+}	8.0	13.6	13.9	2.0	57.4
Mg^{2+}	2.3	6.0	8.9	0.0	43.9
Cl^-	6.3	9.2	9.0	0.9	36.1
NO_3^-	21.0	40.1	50.2	1.4	215
PO_4^{3-}	0.0	1.9	2.9	0.0	11.2
SO_4^{2-}	28.8	42.6	42.0	1.2	142
$\sum$ cations	82.5				
$\sum$ anions	56.1				

Table 4. pH, electric conductivity (μS cm^{-1}), and concentrations (μeq L^{-1}) of the pacific samples.

n=8	median	avg	σ	min	max
pH	4.93	4.78	0.34	4.32	5.32
conductivity	14	17	12	5	38
calc. conductivity	10	14	11	4	34
H^+	13.2	16.7	14.3	4.8	47.9
NH_4^+	32.2	33.5	32.0	0.0	101
K^+	4.6	8.4	9.7	0.0	27.6
Na^+	4.3	10.8	13.4	4.3	43.0
Ca^{2+}	8.5	8.7	2.9	5.0	12.0
Mg^{2+}	1.7	3.5	3.9	0.8	12.5
Cl^-	5.2	5.3	3.8	0.0	13.3
NO_3^-	11.5	19.8	21.2	1.2	55.6
PO_4^{3-}	0.0	0.5	1.5	0.0	4.1
SO_4^{2-}	29.6	30.1	23.3	2.0	60.4
$\sum$ cations	64.5				
$\sum$ anions	46.3				

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Table 5. pH, electric conductivity [μS cm^{-1}], and concentrations [μeq L^{-1}] of the rain samples.

n=11	median	avg	σ	min	max
pH	5.26	5.11	0.31	4.54	5.61
conductivity	6	6	3	3	11
calc. conductivity	4	5	3	2	13
H^+	5.5	7.7	7.5	2.5	28.8
NH_4^+	0.0	3.6	11.5	0.0	38.3
K^+	2.3	7.1	9.5	0.0	30.0
Na^+	4.3	7.4	4.3	4.3	17.2
Ca^{2+}	5.0	6.8	2.9	3.5	11.5
Mg^{2+}	1.3	2.5	2.5	0.5	8.6
Cl^-	4.9	4.9	1.9	1.7	7.3
NO_3^-	0.6	1.8	2.4	0.4	8.4
PO_4^{3-}	2.4	4.4	7.5	0.0	25.9
SO_4^{2-}	8.1	7.4	4.9	1.5	14.8
$\sum$ cations	18.4				
$\sum$ anions	16.0				

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Table 6. Distribution of the median equivalent contribution to the measured ion load (%) of the fog and the rain samples.

measured ion	fog samples (n=56)	rain samples (n=11)
H^+	19%	16%
NH_4^+	22%	0%
K^+	5%	7%
Na^+	6%	13%
Ca^{2+}	6%	14%
Mg^{2+}	2%	4%
Cl^-	4%	14%
NO_3^-	14%	2%
PO_4^{3-}	0%	7%
SO_4^{2-}	22%	23%

Fig. 1. Passive fog collector.

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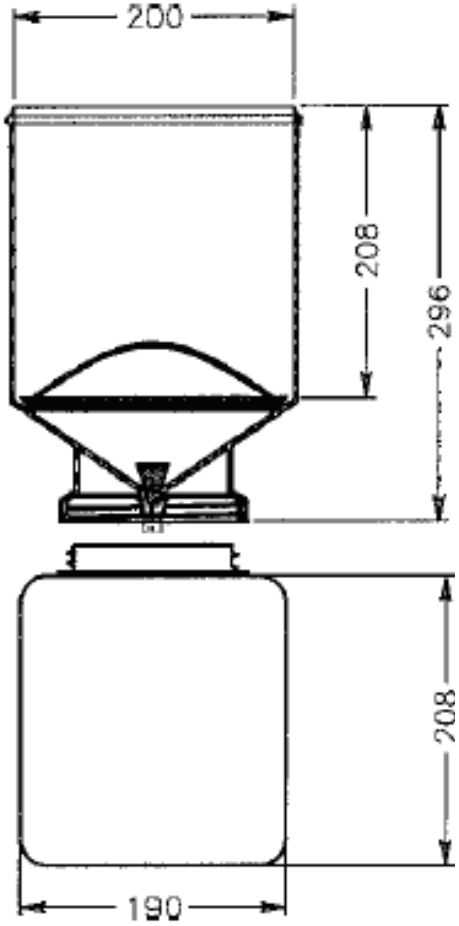

Fig. 2. Rain collector.

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2, 863–885, 2005

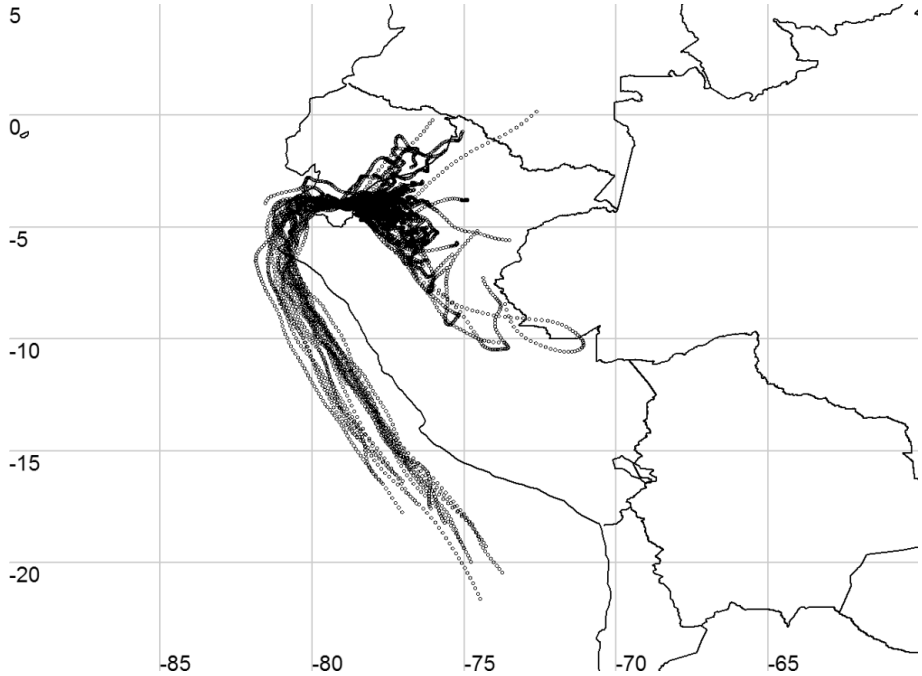

Fig. 3. 120 h backward trajectories for every collected fog sample.

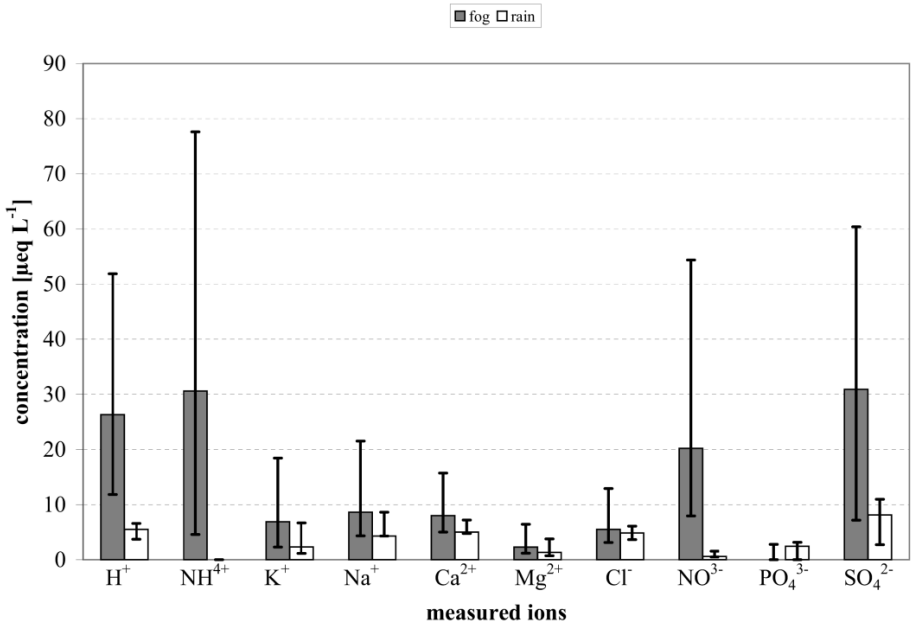

Fig. 4. Comparison of the median equivalent concentrations of the fog (grey bars) and rain (white bars) samples.