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Ion fluxes from fog and rain to an agricultural and a forest ecosystem in Europe

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Abstract

The deposition fluxes of inorganic compounds dissolved in fog and rain were quantified for two different ecosystems in Europe. The fogwater deposition fluxes were measured by employing the eddy covariance method. The site in Switzerland that lies within an agricultural area surrounded by the Jura mountains and the Alps is often exposed to radiation fog. At the German mountain forest ecosystem, on the other hand, advection fog occurs most frequently. At the Swiss site, fogwater deposition fluxes of the dominant components SO_4^{2-} (0.027 mg S m^{-2} day^{-1}), NO_3^- (0.030 mg N m^{-2} day^{-1}) and NH_4^+ (0.060 mg N m^{-2} day^{-1}) were estimated to be $<5\%$ of the measured wet deposition (0.85, 0.70 and 1.34 mg m^{-2} day^{-1}, respectively). The corresponding fluxes at the forest site (0.62, 0.82 and 1.16 mg m^{-2} day^{-1}, respectively) were of the same order of magnitude as wet deposition (1.04, 1.01 and 1.36 mg m^{-2} day^{-1}), illustrating the importance of fog (or occult) deposition. Trajectory analyses at the forest site indicate significantly higher fogwater concentrations of all major ions if air originated from the east (i.e. the Czech Republic), which is in close agreement with earlier studies.

Keywords: Fog; Acid rain; Chemical composition; Eddy covariance; Flux measurements; Deposition

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1. Introduction

Nitrogen, and to some extent also sulphur, are essential plant nutrients (Wellburn, 1994). However, if applied in too large amounts, these nutrients may disturb the natural

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cycle and eventually have detrimental effects on ecosystems (Brady and Weil, 1996). The optimum nutrient level depends mainly on the ecosystem concerned. Whereas the productivity of agroecosystems relies on relatively large amounts of nitrogen supplies, in forests, critical loads are much smaller (cf. Schulze, 1989). In case of overnutrition, sulphur and nitrogen may be lost through the soil system by leaching, which causes groundwater pollution as well as eutrophication of lakes and rivers. In addition, essential cations, in particular calcium and magnesium, may be leached from the soils as they are displaced by H^+ ions on the soil colloids (Brady and Weil, 1996). This effect is enhanced through the deposition of acid by rain and fog.

Here we report on the deposition of nutrients by fog and rain observed during two measurement campaigns in a Swiss agroecosystem and a German mountain forest ecosystem. Deposition by fog—hereafter referred to as ‘occult deposition’—was determined by combining the results of an active cloudwater collector with direct fogwater flux measurements obtained by employing the eddy covariance method. Wet deposition was measured by means of a wet-only sampler. The main objective was to determine the share of occult deposition in comparison to wet deposition measured in two different ecosystem types.

2. Experimental

2.1. Site description

2.1.1. Kerzersmoos, Swiss Plateau (KMS)

The first field campaign took place on the Swiss Plateau at Kerzersmoos at 435 m a.s.l. (46°59′40″N, 7°11′02″E) between 10 April and 31 May 2000. This site is situated in one of the most productive agricultural regions in Switzerland, on the plain between the Jura mountains and the Alps. Accordingly, the vegetation is largely dominated by arable land and grassland resulting from crop rotation. The measurements were carried out at 7.0 m a.g.l. Due to the situation of the site, fog often forms as a consequence of radiative cooling of the surface and the air masses close to the ground. Fog is most frequently observed during the cold season from September to March with a maximum occurrence between mid-September and the end of October (Wanner, 1979; Bachmann and Bendix, 1993). Persistent fog layers may exist over periods of several days.

2.1.2. Waldstein, Fichtelgebirge, Germany (WFG)

The measurements of the second field campaign were carried out at the Waldstein site at 786 m a.s.l. in the Fichtelgebirge in northeastern Bavaria in Germany (50°08′35″N, 11°52′09″E) between 27 June and 5 December 2000. The instruments were mounted on a 32-m tower ≈ 13 m above the canopy of a spruce forest. The site is situated close to the border of the Czech Republic. In the Fichtelgebirge, fog is predominantly caused by the advection of clouds. Thus, fog is usually accompanied by considerable wind speeds. Western and southwestern winds clearly dominate. Southeastern winds are less frequent. They carry air masses from northwestern Bohemia, which have been reported to be affected by emissions from lignite power plants in the Czech Republic (Matzner et al., 2001;

Wrzesinsky and Klemm, 2000; Klemm and Lange, 1999). Fog can be observed during the entire year but is most frequent between September and March. In 2000, there were 222 days with fog, totalling 1685 h with fog.

2.2. Methods

2.2.1. Wet deposition measurements

Wet deposition was measured by means of a wet-only sampler (Eugster et al., 1998; Eugster, 1999). The sampling container holds a polyethylene bucket and is coated with a thick layer of styrofoam to keep the bucket at a constant and low temperature. A reflective aluminium cover minimizes direct heating by solar radiation. A rain droplet detection sensor is used to open and close the lid (Eugster, 1999).

The samples were collected on a weekly basis at KMS whereas at WFG, they were collected simultaneously with the fogwater samples, i.e., after the occurrence of a fog event. Seven and thirty-nine rainwater samples were collected at KMS and WFG, respectively. Wet deposition was determined by multiplying the ionic concentrations measured in each sample by the amount of precipitation measured during the same period of time.

2.2.2. Occult deposition measurements

The chemical composition of the fogwater was determined at WFG by use of a modified Caltech active strand cloudwater collector (Daube, 1987) with six rows of parallel teflon strands with a calculated cut-off droplet diameter of 7.1 μm. The device was controlled by the visibility measurements of a PWD11 present weather detector (Vaisala Oy, Helsinki, Finland), using a threshold visibility of 500 m.

Fogwater fluxes were determined by means of the eddy covariance method, combining the measurements of a 3D sonic anemometer (Solent HS, Gill, Lymington, UK) with those of a cloud droplet spectrometer (Droplet Measurement Technologies, Boulder, CO, USA). Fog droplets within the diameter range 2 and 50 μm were categorized into 40 particle size bins (cf. Burkard et al., 2001). Fogwater deposition was calculated separately for the gravitational and the turbulent fogwater flux according to Beswick et al. (1991) (cf. also Kowalski and Vong, 1999). The gravitational flux was determined using the Stokes' settling velocity. The turbulent flux was calculated by means of the eddy covariance method, which relies on the observation that turbulent fluxes in the atmosphere are driven by the short-term fluctuations of the wind vector, i.e., the turbulence. Small-sized gases, particles and water droplets contained in an air parcel follow the turbulent motions of the air. The turbulent flux F_c in the vertical direction can therefore be expressed by the covariance of vertical wind speed w and liquid water content c, $F_c = \overline{w'c'}$, where the overbar denotes a temporal average (typically 30 min), and primes denote the instantaneous turbulent deviation of a measurement from its temporal mean, e.g. $w' = w - \overline{w}$. Further details are given in Burkard et al. (2002).

The total fogwater flux, i.e. the turbulent plus the gravitational flux, is calculated by summarizing the 30-min mean values for visibilities below 2000 m. Fogwater fluxes become negative, i.e. directed downwards, as soon as visibility is below 2000

m. Nutrient deposition by fog was calculated by multiplying the total fogwater flux measured during each fog event by the ionic concentrations measured in the collected fogwater. Because fogwater droplets with a diameter <7 μm were typically transported upwards rather than downwards, the 7.1-μm-cut-off of the fogwater collector is not expected to lead to an underestimation of deposited ions. Droplets with a diameter <7 μm contributed less than 1% to the turbulent fogwater flux during the WFG field campaign. The total fogwater flux (turbulent and gravitational flux) that was measured during periods with visibilities between 500 and 2000 m, which is above the threshold for switching on the cloudwater collector, on average only contributed 6% to total deposited fogwater. Although it would be desirable to collect fogwater even during times with slight fog, the collection efficiency was found to be poor for visibilities >500 m. Furthermore, contamination by dust and insects is too strong in relation to the tiny volume of water that can be sampled under such conditions.

At WFG, 56 fogwater samples were collected. In order to estimate fogwater concentrations for the KMS site, where no fogwater was collected, minimum and maximum values of the ratio between the ionic concentrations of rain- and fogwater as published by Dasch (1988), Joos and Baltensperger (1991), Zier (1992), Schemenauer et al. (1995), and Collett et al. (1999) were used in combination with ionic concentrations measured in rainwater (Table 1). Occult deposition for the KMS was calculated by multiplying the estimated ion concentrations by the total liquid water flux.

2.2.3. Analytical procedures

The rainwater samples collected at KMS were filtered through a 0.2 μm nylon filter to remove solid particulate matter. Filtrates were stored in precleaned polyethylene bottles in the freezer until chemical analysis. Concentrations of F^-, Cl^-, NO_2^-, PO_4^{3-}, NO_3^-, SO_4^{2-}, Na^+, NH_4^+, K^+, Mg^{2+} and Ca^{2+} were measured by ion chromatography using a Dionex DX120 (Sunnyvale, CA, USA).

Rain- and fogwater samples collected at WFG were stored in precleaned polyethylene bottles in the freezer. Before chemical analysis, the samples were filtered through a 0.45 μm cellulose acetate filter. The cations Na^+, K^+, Mg^{2+} and Ca^{2+} were measured by atomic emission spectroscopy (ICP-AES). NH_4^+ and the anions Cl^-, NO_3^-, SO_4^{2-}, PO_4^{3-} were measured by ion chromatography for which a Dionex DX100

Table 1
Estimated ionic concentrations (in μeq l^{-1}) of fogwater at the Kerzersmoos site, based on minimum and maximum ratios between rain- and fogwater and the weighted mean concentration measured in the rainwater according to literature values (see text)

Ion	Rain	Minimum estimate		Maximum estimate	
		Fog	Ratio	Fog	Ratio
SO_4^{2-}	22.3	66.9	3	402	18
NO_3^-	21.1	63.2	3	505	24
NH_4^+	40.3	201.4	5	1007	25

was used. The pH value was measured by a pH-electrode (WTW Sentix 21). Electrical conductivity was determined by a conductivity-electrode (WTW KLE1).

2.2.4. Trajectory calculations

Three-dimensional backward trajectories were calculated with the Hysplit-4 model from NOAA (Draxler and Hess, 1998) in conjunction with FNL archived meteorological data (Stunder, 1997). The trajectories were calculated for all fog events observed at WFG in order to verify the assignment of air mass origin to fog events.

3. Results

3.1. Data quality

Data quality was examined by considering the ion balances of the samples and, furthermore, by comparing the measured electrical conductivity with the calculated electrical conductivity. For all fogwater samples, the ion balances show a good agreement between the sum of anions and the sum of cations (median value Σanions/Σcations = 0.97, cf. Fig. 1a). The agreement between calculated and measured electrical conductivity is also very high ($r^2 = 0.974$; Fig. 1b).

With regard to the rainwater samples, where ionic concentrations are much lower, the ion balances show a less perfect agreement in that the range of Σanions/Σcations varies (Fig. 1c) with a median of 0.87. The agreement between calculated and measured electrical conductivities is only slightly lower than that for the fogwater samples, namely $r^2 = 0.936$ (Fig. 1d). One out of the thirty-nine rainwater samples had to be excluded from the discussion of ionic concentrations and nutrient deposition because the comparison of calculated and measured electrical conductivity (ratio 2.5) showed that some error in the chemical analysis must have occurred.

The results regarding the data quality of fog- and rainwater samples show that due to the higher concentrations found in fogwater, these samples could be analysed with a higher relative accuracy. In the rainwater samples, the amounts of ions (in particular of Ca^{2+} and K^{+}) were often below the detection limit. The data quality assessment

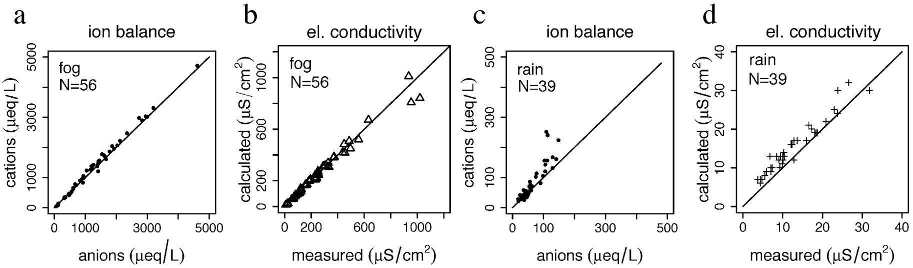

Fig. 1. Fogwater ion balance (a), electrical conductivity (b), and rainwater ion balance (c) and electrical conductivity (d) obtained at the Waldstein site.

Table 2
Comparison of the weighted means of pH and ionic concentrations (in μeq l^{-1}) in the rain- and fogwater collected between 27 June and 5 December 2000 at the Waldstein site

Ion	pH	H^+	Cl^-	NO_3^-	SO_4^{2-}	NH_4^+	Ca^{2+}	K^+	Mg^{2+}	Na^+
Rainwater	5.3	7.9	5.6	26.9	24.5	36.7	7.6	1.8	1.2	9.9
Fogwater	4.1	123.2	42.6	646.2	437.7	925.5	53.9	12.1	13.2	46.8
Ratio rainwater/fogwater	–	16	8	24	18	25	7	7	11	5

described could not be applied to the rainwater samples collected at KMS, because pH and electrical conductivity were not measured.

3.2. Fog- and rainwater chemistry

The relative ionic composition of fog- and rainwater at WFG was found to be similar. The three ions nitrate, sulphate and ammonium account for 72% and 90% of the total amount of ions in rain- and fogwater, respectively. However, absolute concentrations are much higher in fogwater than they are in rainwater. The ratios between the concentrations of rain- and fogwater, respectively, are particularly high for nitrate, sulphate and ammonium with factors of 24, 18 and 25, respectively (Table 2).

The weighted mean concentrations of nitrate (21 μeq l^{-1}), sulphate (22 μeq l^{-1}) and ammonium (40 μeq l^{-1}) of the rainwater collected at KMS are within the range of previous studies carried out at the same site (Gempeler, 1997; Nowak, 2000). They are very similar to the concentrations measured in the rainwater collected at WFG. Whereas nitrate and sulphate concentrations are slightly higher in the Fichtelgebirge (27 and 25 μeq l^{-1}, respectively), the median concentration of ammonium is slightly lower (37 μeq l^{-1}).

3.3. Occult and wet deposition

The comparison between KMS and WFG shows very similar values for wet deposition whereas for occult deposition, the calculated values show an enormous difference (Fig. 2).

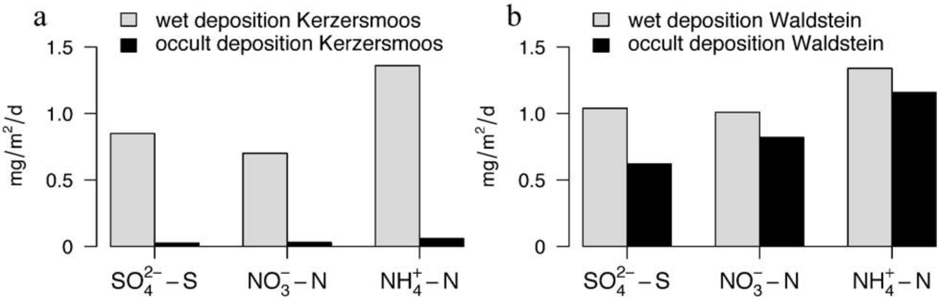

Fig. 2. Comparison of daily average wet and occult deposition of SO_4^{2-}-sulphur, NO_3^- and NH_4^+-nitrogen (in mg m^{-2} day^{-1}) at the Kerzersmoos site (measurement period 10 April to 31 May 2000) and at the Waldstein site (measurement period 27 June to 5 December 2000).

At KMS, fog only makes a small contribution to nutrient deposition (about 4% of wet deposition), whereas at WFG, sulphur and nitrogen deposited by fog amount to 60–85% of that deposited by rain (Fig. 2).

4. Discussion

4.1. Differences between sites

The differences in rainwater chemistry between the two measurement sites can be explained by the differences regarding the situation of the two sites. WFG is situated close to the border of the Czech Republic. Although the emissions of air pollutants including SO_2 decreased significantly since the 1980s, enhanced concentrations of SO_2 still occur at WFG (Klemm and Lange, 1999). Therefore, enhanced concentrations in rain and fog are to be expected there.

With regard to NH_4^+, on the other hand, the higher concentrations at KMS can be explained by the location of the site in an area of intense agricultural land use. Agriculture is an important source of ammonia (NH_3), mainly from life stock and due to fertilizing. For example, the regional NH_3 emission inventory as used by Eugster et al. (1998) indicates an average emission flux of 23 kg N ha^{-1} $year^{-1}$ from the 100 km^2 surrounding KMS.

As the precipitation amounts were similar (2.37 mm day^{-1} at KMS, 2.67 mm day^{-1} at WFG), the deposited amounts of nitrate, sulphate and ammonium reflect the same picture as the concentration measurements: slightly higher depositions of SO_4^{2-} and NO_3^- at KMS, and slightly lower deposition of NH_4^+, mainly due to differences in concentrations.

4.2. Fog- and rainwater chemistry

Fogwater chemistry at WFG was compared to rainwater chemistry. The ratios between ionic concentrations measured in fog- and rainwater show high amounts of ions in the fogwater, in particular for sulphate, nitrate and ammonium. This agrees well with earlier studies showing that ionic concentrations in fogwater are consistently higher than in rainwater (Dasch, 1988; Zier, 1992; Miller et al., 1993; Collett et al., 1993; Schemenauer et al., 1995).

In comparison to earlier studies carried out at the same site between April and October 1997 (Wrzesinsky and Klemm, 2000), and between July 1998 and March 1999 (Römpp et al., 2001), the concentrations measured between July and December 2000 are considerably higher.

4.3. Influence of air mass origin on fogwater chemistry

As the samples were mostly collected on an event basis, they could be classified according to wind direction. This classification into eastern and western fog events was based on the measured 10-min mean values previous to (2 h before the beginning of foggy conditions) and during fog. Wind speeds below 1 m s^{-1}, and conditions where measured wind direction did not agree with trajectory calculations were not taken into consideration. Thus, 39 out of 56 fog events could be clearly assigned the influence of either eastern or western air masses.

For all three major ions sulphate, nitrate and ammonium, the median concentrations in the fogwater samples are significantly higher for eastern than for western fog events. The difference is most pronounced for sulphur where the median of eastern events is more than double the median of western events (767 vs. 354 μeq l^{-1}). For nitrate and ammonium, the concentrations in fog advected from the east are 59% and 57%, respectively, higher than those in fog transported from the west. Differences in liquid water content are small and do not explain the observed differences in concentrations.

4.4. Occult deposition

Table 3 summarizes the most relevant parameters regarding occult deposition. At the WFG site, the liquid water flux measured at a height of 31.5 m was used for calculating occult deposition. Burkard et al. (2002) found that although turbulent liquid water flux changes with height, this should not dramatically change the amount of ions deposited since the most likely reason for this flux divergence appears to be evaporation, which does not alter the total mass of *nonvolatile* ions dissolved in droplets, but only increases their concentration (cf. Fuzzi, 1984). The deposition amounts of nitrogen and sulphur are roughly 19 to 27 times higher at WFG than at KMS (Table 3). The KMS measurements indicate that the fog liquid water flux is too small due to low wind speeds and short duration of fog to allow for significant nutrient inputs. Thus, in this agroecosystem, it is

Table 3
Comparison of fog flux measurements and occult deposition of NH_4^+, NO_3^- and SO_4^{2-} between the Kerzersmoos (10 April–31 May 2000) and the Waldstein site (27 May–5 December 2000)

	Unit	Kerzersmoos	Waldstein	Ratio
Number of days of campaign	days	69	161	–
Total hours with fog	h	49.5	923	–
Average hours per day with fog	h day^{-1}	0.71	5.7	8.0
Mean LWC[a]	mg m^{-3}	61.2	65.9	1.1
Mean gravitational flux[a,b]	mg m^{-2} s^{-1}	−1.34	−1.10	0.8
Mean turbulent flux[a,c]	mg m^{-2} s^{-1}	−0.31	−3.81	12.2
Mean total flux[a]	mg m^{-2} s^{-1}	−1.65	−4.91	3.0
NH_4^+-N deposition[d]	mg m^{-2} day^{-1}	0.060	1.16	19.3
NO_3^--N deposition[d]	mg m^{-2} day^{-1}	0.030	0.82	27.3
SO_4^{2-}-S deposition[d]	mg m^{-2} day^{-1}	0.027	0.62	22.1

[a] Averaged over periods with fog.

[b] Flux at measurement height; assumes constant gravitational flux with height; however, due to evaporation of falling droplets, their size and fall speed are subject to change, suggesting that the amount reaching the vegetation surface in liquid phase is less than the measurements above the vegetation.

[c] Flux at measurement height; flux divergence estimates (see Burkard et al., 2002) suggest that turbulent flux reaching the surface might be reduced to 55% and 41% at the Kerzersmoos and Waldstein sites, respectively.

[d] Averaged over 24-h periods.

justified to measure wet deposition only, thereby neglecting occult deposition as was suggested by Fuhrer (1986), based on rough estimations of deposition rates. Although our measurements took place at the end of the fog season and ions were not measured directly at KMS, it is expected that due to vegetation and predominant type of fog, year-round measurements would yield similar occult deposition rates.

The enormous difference between the two sites regarding the share of occult deposition can be explained as follows. (1) The difference in season for the two measurement campaigns is crucial regarding the frequency of fog occurrence. The WFG campaign included the period during which fog occurs most frequently. At KMS, on the other hand, measurements were carried out in the season of least fog frequency. (2) The two sites differ with regard to the predominant type of fog. At KMS, all fog events have been identified as radiation fog, characterized by small wind speeds (mean wind speed $\overline{u}$ during fog events = 1.23 ± 0.11 m s^{-1} (mean ± S.E.), friction velocity $u_* = 0.083 \pm 0.014$ m s^{-1}) and a distinct droplet size spectrum. In the Fichtelgebirge, fog is caused by the advection of clouds with considerably higher wind speeds ($\overline{u} = 3.28 \pm 0.01$ m s^{-1}, $u_* = 0.575 \pm 0.002$ m s^{-1}). This results in stronger turbulence at WFG, which in turn produces a much larger turbulent flux. (3) The difference in vegetation cover affects the process of occult deposition. Whereas at KMS, the vegetation is dominated by arable land and grassland, the WFG tower is located in a spruce forest and the measurements have been carried out just above the tree canopy. This means a considerably rougher surface and a greater leaf area index that enhances the turbulent settling of fog droplets.

4.5. Uncertainties in occult deposition estimates

Hänel (1982) emphasizes the efficiency of fog episodes in removing particulate matter, namely aerosols, from the atmosphere. Since the cloud droplet spectrometer used for this study only resolves droplets in the size range 2–50 μm in diameter, the smaller aerosol particles are not included in our deposition figures, unless they were dissolved in fog droplets. On the other end of the spectrum, droplets >50 μm that are uncommon during pure fog events, but could be important when fog and drizzle or rain occur simultaneously, were not considered either. Hence, our estimates presented here contain uncertainties that are closely related to the fuzzy definition of ‘occult deposition’, which is hard to delimit from dry aerosols at the lower boundary, or wet deposition at the upper boundary.

To fully understand microphysical processes, it would be important to obtain additional information for particles < 2 μm and droplets >50 μm. Furthermore, because the eddy covariance method requires the measurements to be performed in the atmosphere above or between vegetation elements rather than on the vegetation surface itself, uncertainties remain about the chemical transformation and evaporation or condensation of water between the measurement height and the surface where droplets are intercepted.

There is a high complexity in all these interacting processes, especially in complex terrain where fog properties may exhibit a great small-scale variation both in the horizontal and vertical direction. By using the eddy covariance method, it is assumed that such variation is chaotic, that is entirely explained by the movements of the turbulence elements—the eddies—which are randomly distributed in space and which are moved across our sensors without preferential flow pathways (cf. Taylor, 1938). This may not always be strictly correct under real-world conditions, and thus adds significant uncertainty to the quantification of deposition fluxes, as our reviewer likes to point out.

Some of the aspects of uncertainty are generic for all environmental field measurements, others are specific to the instrumentation used in this study. We take the viewpoint here that such quantifications are very helpful for ecological purposes, and most likely better than pure estimates without field measurements. The absolute accuracy of our measurements should, however, not be overestimated.

5. Conclusions

For advection fog, ionic concentrations are influenced by air mass origin. At WFG, air masses advected from the east are still considerably more polluted than air masses advected from the west. Thus, nitrate, sulphate and ammonium showed concentrations that were 57%, 117% and 59% higher, respectively, for eastern than for western fog events.

At WFG, fog adds 60–85% to measured nitrogen and sulphur deposition by rain. The much lower amount of fogwater interception compared to rainfall is compensated for by high ionic concentrations. Thus, occult deposition is on the order of wet deposition for nitrogen (namely, 85% for ammonium and 81% for nitrate), and occult deposited sulphate enhances wet deposition by 60%.

Fog was found to be a major factor in nutrient deposition to the spruce forest ecosystem under investigation. However, in case of the agroecosystem, its contribution to nutrient deposition was very small compared to that of wet deposition. In order to be able to decide whether the costly measurements of occult deposition are justified, the following key factors must be taken into consideration: fog frequency, type of fog, vegetation cover and wind speed. Our results suggest that occult deposition to forests that are frequently exposed to advection fog can double the nutrient inputs observed by wet-only deposition measurements.

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