PERGAMON

AE International – Europe

Atmospheric Environment 37 (2003) 2979–2990

ATMOSPHERIC ENVIRONMENT

www.elsevier.com/locate/atmosenv

Vertical fogwater flux measurements above an elevated forest canopy at the Lägeren research site, Switzerland

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Received 13 December 2002; received in revised form 14 March 2003; accepted 19 March 2003

Abstract

During the winter of 2001/2002 wet and occult deposition measurements were performed at the Lägeren research site (690 m a.s.l.) in Switzerland. Two types of fog were observed: radiation fog (RF) and fog associated with atmospheric instabilities (FAI). The deposition measurements were performed above the forest canopy on a 45 m high tower. Occult deposition was measured by means of the eddy covariance method. Due to the large differences of microphysical properties of the two fog types, the liquid water fluxes were much higher (6.9 mg m^{-2} s^{-1}) during RF than during FAI (0.57 mg m^{-2} s^{-1}). Fogwater concentrations were considerably enhanced during RF compared with FAI. The comparison of fog and rain revealed that fogwater nutrient concentrations were 3–66 times larger than concentrations in precipitation. The considerably larger water fluxes and nutrient concentrations of RF resulted in much higher nutrient deposition compared with FAI. In winter when RF was quite frequent, occult deposition was the dominant pathway for nitrate and ammonium deposition. Daily fluxes of total inorganic nitrogen were 1.89 mg m^{-2} d^{-1} by occult and 1.01 mg m^{-2} d^{-1} by wet deposition. The estimated contribution of occult deposition to total annual nitrogen input was 16.4% or 4.3 kg N ha^{-1} yr^{-1}, and wet deposition contributed 26.5% (6.9 kg N ha^{-1} yr^{-1}). As a consequence, critical loads of annual N-input were exceeded, resulting in a significant over-fertilization at the Lägeren site.

Keywords: Fog; Chemical composition; Eddy covariance; Flux measurements; Deposition; Annual nitrogen input

1. Introduction

The interception of fog and cloud droplets by vegetation has widely been recognized as a significant component of the hydrologic budget of mountain forests that are frequently immersed in dense fog (Dasch, 1988; Schemenauer and Cereceda, 1994; Walmsley et al., 1996; Bruijnzeel, 2001). Fog is a cloud in contact with the ground that reduces visibility to ⩽1000 m. In industrialized countries, nutrient and pollutant concentrations in fogwater can be up to 100 times higher than in rainwater, and therefore fogwater fluxes can also contribute significantly to the deposition of acidity, nitrogen, sulphur, and other ionic compounds (Waldman et al., 1982; Fuzzi et al., 1996; Choularton et al., 1997; Hameed et al., 2000; Wrzesinsky and Klemm, 2000; Thalmann et al., 2002), even in cases where fog is not as frequent as in tropical cloud forests.

Direct measurements of these fluxes are a demanding task, and therefore first estimates are mostly obtained by deposition modelling, as it was for example done for balsam fir forests of the northern Appalachian mountains by Lovett (1984). In recent years the eddy covariance (EC) method has been applied to measure the deposition of fog droplets directly (Beswick et al., 1991; Vong and Kowalski, 1995; Vermeulen et al., 1997). Kowalski and Vong (1999) found that the surface uptake measured by EC was much smaller than model predictions, raising the question whether earlier studies have significantly overestimated ion deposition to ecosystems.

Here we report on direct EC fogwater flux measurements performed over a mixed forest in Switzerland

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1352-2310/03/$ - see front matter
doi:10.1016/S1352-2310(03)00254-1

within the "Fog Interception and Nutrient Inputs to Montane-Subalpine Areas in Switzerland" (FINIMSAS) project. The measurements were carried out in order to quantify current fogwater deposition (hereafter denoted as occult deposition) of major inorganic ions during one cold season, and to compare this deposition with conventional wet-only deposition measurements.

Excessive nitrogen deposition may have harmful effects on the vegetation. For instance, ammonium can cause decreasing base cation supply due to occupation of exchange capacity in soils. Furthermore, nitrate, as well as ammonium if subsequently nitrified (= oxidized), can add to acidification of the soil or increase cation leaching, which may also result in malnutrition. Additionally, increased uptake of nitrogen by forest can result in nutrition imbalance if not accompanied by higher base cation uptake (Schulze et al., 1989; Katzensteiner, 2000). Possible effects of such processes on individual plants include increased susceptibility to frost, herbivory and biological stress factors (beetles, bacteria, viruses, fungi), and to air pollutants such as ozone (Collett et al., 1990). Typical symptoms may include chlorosis, necrotic lesions ("rust") or discoloring. Excess N uptake can also disturb the balance of whole plant communities and may cause loss of species diversity or shifts in species composition of ecosystems (Eugster, 1999). MacDonald et al. (2002) showed that the N-input to European forests by throughfall (wet + occult + dry deposition) ranges from less than 1 kg N ha^{-1} yr^{-1} to more than 60 kg N ha^{-1} yr^{-1}. The contribution of occult deposition was not quantified in their study, but according to several studies (e.g. Fuhrer, 1986; Dasch, 1988; Fuzzi et al., 1996; Wrzesinsky and Klemm, 2000; Thalmann et al., 2002), it is obvious, that the variability in N-input to forests in Europe can be partly explained by the absence or occurrence (with variable frequencies and characteristics) of fog, especially at elevated sites.

With our measurements we addressed the question how relevant the ammonium, acid and other ion loads from fog that are believed to be one possible cause of novel forest decline (Nihlgård, 1985) actually are for the mixed forest under investigation.

2. Experimental

2.1. Site description

The Lägeren research site is situated at 47°28′49″N, 8°21′05″E at 690 m a.s.l. on the south slope of the Lägeren mountain (866 m a.s.l.), approximately 15 km northwest of Zurich, Switzerland. The south slope of the Lägeren mountain marks the boundary of the Swiss Plateau, which is bordered by the Jura and the Alps. This site became a permanent station of the Swiss air quality monitoring network (NABEL) in 1986. The vegetation cover around the research site is mixed forest dominated by beech and Norway spruce.

2.2. Flux measurements

Fogwater collection and liquid water flux measurements were performed on a 45 m tall tower at 35 and 45 m, respectively (5 and 15 m above the forest canopy). Fogwater flux measurements took place between 23 May 2001 and 10 April 2002, whereas fog- and rainwater sampling started on 21 September 2001 and continued until the end of the flux measurements.

The vertical flux of fog droplets above a forest canopy is governed by two primary transport processes. The main process is turbulent diffusion of smaller droplets that are suspended in the atmosphere which leads to impaction of the droplets on structural elements of the vegetation. The other process is gravitational settling of the larger droplets (droplet diameters > 10 μm). The relative importance of both processes is largely determined by the size distribution of the fog droplets and the strength of turbulent mixing. The turbulent part of the liquid water flux can be measured directly, whereas the gravitational part can be calculated using Stokes's settling velocity (Beswick et al., 1991). The turbulent flux was calculated by means of the EC 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 (30 min in our case), and primes denote the instantaneous turbulent deviation of a measurement from its temporal mean, e.g. $w' = w - \bar{w}$. Details on EC data processing are described in Burkard et al. (2002).

To yield ion fluxes from EC liquid water flux measurements we took fogwater samples, analyzed them for major inorganic compounds as described in Section 2.4, and multiplied concentration measurements with the liquid water fluxes. In another field campaign we found that there remain some uncertainties with regard to changes of the liquid water flux with height (Burkard et al., 2002). But the deposition of ions does not appear to be affected, because the total mass of nonvolatile ions dissolved in fog droplets is conserved (Burkard et al., 2002). Further details on the ion flux measurements were described by Thalmann et al. (2002).

2.3. Instrumentation

EC fluxes were measured with a three-dimensional ultrasonic anemometer (model 1199 HSE with a built-in inclinometer, Gill Ltd., Solent, UK) and an active high-speed FM-100 cloud particle spectrometer (Droplet Measurement Technologies, Inc., Boulder, CO, USA). Its principle of operation is described in detail in Burkard et al. (2002). Fog droplets within the diameter range 2 and 50 μm were categorized into 40 size bins. The anemometer and the FM-100 were operated at a sampling rate of 12.5 Hz, in order to resolve most of the frequency spectrum of turbulent motion.

A modified Caltech-type Active Strand Cloudwater Collector (CASCC) (Daube et al., 1987; Demoz et al., 1996) was constructed for this field experiment. The 50% collection efficiency of this CASCC was determined to be 6.6 μm according to Demoz et al. (1996), with an overall collection efficiency of 93%. The CASCC was triggered by the visibility measurements obtained from a present weather detector (model PWD11, Vaisala, Helsinki, FI; Nylander et al., 1997). Whenever the visibility dropped below 500 m the fogwater collector was switched on and fogwater samples were collected during time intervals of 6 or 12 h. Rainwater was sampled by a wet-only sampler (Eugster et al., 1998).

Ancillary meteorological data were measured every 10 s employing a data logger (Campbell Scientific, Inc. model CR10X) and stored as averages every 10 min.

All measurement devices were connected via digital serial data lines (RS422) to a laptop. The data were transferred weekly to a SUN workstation and were then evaluated and processed by the in-house software CONVERTALL version 11.08.[1] A detailed description of the basic concepts of this software can be found in Eugster (1994).

2.4. Analytical procedures

Precipitation- and fogwater samples were collected in precleaned polyethylene bottles. Immediately after measuring pH and electrical conductivity, the samples were stored at −65°C until chemical analysis of the major inorganic ions F^-, Cl^-, NO_2^-, PO_4^{3-}, NO_3^-, SO_4^{2-}, Na^+, NH_4^+, K^+, Mg^{2+}, and Ca^{2+}. The pH was measured using an electrode (Single Pore pH Electrodes, Hamilton, Switzerland), and electrical conductivity was determined by a conductivity cell (TetraCon WTW, Germany). The pH electrode was frequently calibrated with two buffered solutions with pH 4.0 and 7.0, and the conductivity cell was zeroed with demineralized water. After filtration through 0.2 μm nylon filters the fog and rainwater samples were analyzed by ion chromatography using a Dionex DX120. During the field campaign, several blank samples were collected in order to estimate the contamination of samples due to the collector or sample handling. No significant contaminations were observed, neither before cleaning the fogwater collector nor after cleaning of the CASCC by spraying demineralized water into the running collector.

The quality of the chemical analysis was assured by running ≈10 standard samples in each batch together with the field samples. Data quality was finally examined by considering the ion balances of the samples, and by comparing the measured with the calculated electrical conductivity. The percentage difference threshold of the ion balance and the conductivity for a given sample was 25%. No samples had to be rejected on this evaluation basis.

[1] The C source code can be obtained freely from the authors.

3. Results

3.1. Classification of fog events

Two types of fog were observed at the Lägeren research site: (1) Radiation fog (RF), which forms when radiative cooling reduces the air temperature to or below its dewpoint, especially at night and early in the morning. (2) Fog associated with atmospheric instability, hereafter denoted as FAI, occurs during situations with a low cloud base due to cooling and moistening by evaporation of falling rain drops (e.g. Curry and Webster, 1999, p. 168) or forced lifting, usually associated with frontal passages or thunderstorms. The microphysical characteristics of the two fog types differed significantly due to their very different origins (Table 1). Therefore, the data were analyzed separately for both fog types.

During the 323 days of our field campaign we observed 151 days with at least 30 min of fog with a visibility ⩽1000 m (47% of all days). During the period when chemical samples were available (21 September 2001–10 April 2002), we counted 101 days with fog out of 201 days. Although the period with the highest fog frequency was observed during the time when the cloudwater collector was in place, the fog frequency during summer was remarkable. Summer fog was generally a result of frontal passages and thunderstorms when our site was immersed in various types of clouds. Two distinct periods with very high fog frequencies occurred: one during November 2001 (both types of fog with equal share) and one during the first half of January 2002 (exclusively RF). In the following analysis a 'fog event' is defined as the period during which the visibility was ⩽1000 m for at least 2.5 consecutive hours.

3.2. Characteristics of fog events

The duration of fog for various visibility categories is presented in Table 2. Dense fog (visibility ⩽200 m) accounted for about 60% of foggy time periods. The two different fog types (RF and FAI) were identified

Table 1
Median liquid water content (LWC), droplet size (VMD), streamwise wind speed (U) and direction (θ, φ), median liquid water fluxes, and friction velocity (u_*) for the two types of fog at the Lägeren research site during the full period 23/05/2001–10/04/2002

	RF		FAI-events	
Threshold visibility	$\leqslant$500 m	$\leqslant$1000 m	$\leqslant$500 m	$\leqslant$1000 m
Duration (h)	227.0	258.0	487.5	563.0
LWC (mg m^{-3})	160.5	143.5	73.0	63.0
Visibility (m)	93.3	103.0	129.7	147.7
$\sigma_L/\bar{L}$[a] (-)	0.56	0.61	0.43	0.48
VMD[b] (μm)	14.36	13.93	10.78	10.43
U (m s^{-1})	0.55	0.58	2.50	2.50
θ[c] (°)	121.2	118.3	256.5	256.6
φ[d] (°)	−13.5	−12.2	−11.0	−10.9
Turbulent flux (mg m^{-2} s^{-1})	−4.92	−4.42	−0.33	−0.18
Gravitational flux (mg m^{-2} s^{-1})	−2.52	−2.26	−0.44	−0.36
Total flux (mg m^{-2} s^{-1})	−7.81	−6.90	−0.80	−0.57
u_* (m s^{-1})	0.18	0.18	0.49	0.49

[a] LWC standard deviation/mean LWC; dimensionless (cf. Kowalski et al., 1997).
[b] Droplet volume mean diameter.
[c] Horizontal wind direction (° from North).
[d] Vertical wind attack angle (° from horizontal plain).

Table 2
Duration of fog with atmospheric instability (FAI), radiation fog (RF), and overall fog (in four density classes and for the meteorological definition of fog with visibility $\leqslant$1000 m) for the period 21/09/2001–10/04/2002

Visibility (m)	FAI events (h)	(%)	RF Events (h)	(%)	Overall fog (h)	(%)
$\leqslant$100	115	2.4	124.5	2.6	240.5	5.0
100–200	134	2.8	64	1.3	206.0	4.2
200–500	99.5	2.1	38.5	0.8	164.0	3.4
500–1000	49	1.0	31	0.6	115.0	2.4
$\leqslant$1000	397.5	8.2	258	5.3	725.5	15.0
	(days)	(%)	(days)	(%)	(days)	(%)
Foggy days	50	24.8	25	12.4	101	50.0

FAI and RF based on a minimum event duration of 2.5 h. Foggy days refers to all days with at least one half hour of fog.

with the help of: (a) weather charts (surface and 500 hPa), (b) meteorological soundings (2× daily, 180 km west of Lägeren), (c) the weather reports of the Swiss Meteorological Institute (SMI), and (d) data from nearby meteorological stations of SMI. Except for three events, the identification could easily be made. Table 1 gives an overview of the characteristics of the two types of fog. RF events are associated with a significantly greater median liquid water content (LWC; 143.5 mg m^{-3}) due to the wider droplet spectrum (during RF: significant contribution of droplets > 20 μm; during FAI: mostly droplets < 20 μm) and a larger droplet volume-mean diameter (VMD; Table 1) compared to FAI events (63 mg m^{-3}). The mean LWC of FAI at Lägeren was close to the value of 72 mg m^{-3} that we observed at the Waldstein research site (Germany, 786 m a.s.l.; Burkard et al. 2002; Thalmann et al., 2002).

3.3. Liquid water fluxes

The difference in liquid water fluxes between RF and FAI was more pronounced for turbulent than for gravitational fluxes (Table 1) because of the large fraction of upward turbulent fluxes during FAI. Similar upward fluxes were found by Kowalski and Vong (1999). Considering FAI, median gravitational fluxes were even higher than median turbulent fluxes.

Although RF was less frequent than FAI at Lägeren, its overall influence was much stronger due to the greater water fluxes and hence greater total water input (Table 1). The cumulative total water input by FAI was 2.7 mm which is only 37% of RF fogwater input (7.3 mm). The very short fog events ($\leqslant$2.5 h) were of little importance, for they contributed only 3% to the total fogwater input. This confirms that our choice of the minimum duration threshold of 2.5 h for defining a fog event was adequate.

Of the total RF fogwater input 33% were due to gravitational settling compared to 64% during FAI events. In absolute numbers (Table 1), however, gravitational settling during FAI is less important. Conditions with a visibility $\leqslant$500 m accounted for 93–96.5% of the fluxes observed at visibilities $\leqslant$1000 m, indicating that with the threshold visibility of 500 m to start the fogwater collector the water input by fog was largely captured.

3.4. Rainfall and meteorological conditions

With regard to the amount and frequency of rainfall, two periods can be distinguished: from June to September rainfall at Lägeren was high with > 80 mm collected each month. From October to March, precipitation was significantly lower than during the first period. During the time when the cloudwater collector was operational, 300.7 mm of total precipitation or 1.5 mm d^{-1} were measured, roughly a factor of 30 compared to the 10.3 mm (0.05 mm d^{-1}) of fogwater input.

December and January were influenced by a strong cold air high pressure system over western Russia. RF events in January 2002 were relatively long and frequent. Another phase with frequent RF occurred from the beginning of October to the second part of November 2001. In contrast to the RF events, the FAI events were widely spread over the whole campaign with a maximum of occurrence during fall.

The wind patterns at the Lägeren were completely different during the two types of fog. Stronger winds from the W were almost exclusively observed during FAI events. Nevertheless the mean wind speed of 2.5 m s^{-1} (Table 1) is still quite low. In contrast, during RF events the wind was mostly blowing from E with a mean wind speed of only 0.6 m s^{-1}.

3.5. Ion concentrations in fog

The concentrations varied enormously between fog events: Conductivity, as a measure of total ion loading of a water sample, ranged from 34.5 μS (FAI) to 1017 μS (RF; Table 3). Several other studies of rather continental experimental sites (e.g. Joos and Baltensperger, 1991; Collett et al., 1993; Thalmann et al., 2002) found a similarly high variability. Most ions (except Mg) showed significantly lower concentrations during FAI compared to RF. The maximum and median Mg concentrations were higher during FAI. Weighted mean values (weighted by LWC) of RF were between 1.1 (protons, not shown) and 4.9 (fluorid) times higher than FAI means. Concentrations of the three dominant ions ammonium, nitrate, and sulphate were around a factor 2.5–3 higher during RF. Higher concentrations during RF than FAI can be explained by suppressed vertical mixing and reduced horizontal transport with subsequent accumulation of pollutants in the shallow

Table 3
Ion concentrations (μeq l^{-1}), pH and conductivity Lf (μS) of RF and FAI

Ion	RF				FAI-event			
	Min.	Max.	Median	Weighted mean[a]	Min.	Max.	Median	Weighted mean[a]
F^-	2.3	128.9	68.4	60.5	b.d.	63.5	7.7	12.4
Cl^-	20.4	322.6	109.9	109.3	15.5	546	35.6	50.1
NO_3^-	195.6	4039	1201	1477	117.7	3012	427	623
SO_4^{2-}	147.3	3623	678	1058	87.6	2632	247.6	338
Na^+	11.0	971	166.1	218	10.5	615	68.8	83.8
NH_4^+	362.3	5165	1548	1945	162.8	3130	605	738
K^+	13.1	261.4	93.1	92.5	12.3	140.4	25.7	33.2
Mg^{2+}	b.d.	88.7	4.4	10.5	b.d.	106.3	4.7	7.9
Ca^{2+}	28.7	1156	49.9	121.4	5.1	630	40.7	69.3
pH	3.7	5.8	3.9	4.3	3.3	6.6	4.5	4.6
Lf	72.9	1017	323	389	34.5	771	125.3	162.1

Medians are computed from event-based median values, i.e. events are not weighted by their duration (b.d. indicates values below the detection limit).

[a] Weighted by the LWC.

atmospheric boundary layer. This behavior was also described in other studies (Collett et al., 1993; Fuzzi et al., 1996). Although weighted mean pH values were similar for both fog types, FAI showed a larger variability than RF. Sites with similar values of NO_3^- or SO_4^{2-} but with lower NH_4^+ concentrations due to lower emissions from agriculture show significantly lower pH values (as low as 2.7), e.g. in north-eastern USA (Jagels et al., 1998) or eastern Canada (Schemenauer et al., 1995).

3.6. Ion concentrations in rain

The ionic composition of the rainwater is dominated by ammonium, nitrate, sulphate and sodium with weighted mean concentrations of 29.6, 27.2, 30.3, and 32.5 μeq l^{-1}, respectively. These four major ions with similar relative fractions contribute more than 70% to the total ionic concentration in precipitation at Lägeren. Additionally, the concentration of chloride (14.6 μeq l^{-1}) is quite high and in combination with the high sodium concentration gives an indication of the origin of the clouds. The median pH value was 5.3, which is in the range of natural background levels (Seinfeld and Pandis, 1998).

As a result of in-cloud and below-cloud scavenging (Alastuey et al., 2001), the ionic composition of rainwater is a mixture of local and distant influences. Whereas the ions dissolved in fogwater of FAI at Lägeren mainly originate from rather close-by sources despite the mostly far-away origin of the associated air masses, the ionic composition of rainwater does indeed show a pronounced large-scale component with a clear dominance of sea-salt ions. The share of chloride and sodium is much larger in precipitation than fog, and the share of ammonium is significantly smaller. This indicates that the pollutants in the atmospheric boundary layer are more efficiently incorporated into fog (by in-cloud scavenging, Collett et al., 1993) than into rain drops (by below-cloud scavenging).

The imbalance of sodium and chloride may result from sodium sulphate aerosols, formed by the reaction of sodium chloride with sulphuric acid. Since atmospheric residence times of the two precursor species SO_2 and NO_2, and their oxidative derivates are within the same range (Seinfeld and Pandis, 1998), the dominance of sulphur must be attributed to a relatively strong distant source, namely the ocean (e.g. sea spray). According to Lin et al. (1999), who refer to Keene et al. (1986), the sea salt contribution of sulphate in precipitation was calculated to be 14% (median value), and thus is much higher than for fog (FAI: 3%; RF: 1.5%). Precipitation concentrations at Lägeren were within the same range as measurements from relatively clean recording stations such as mid or eastern Ireland (Aherne and Farrell, 2002).

3.7. Deposition of nutrients by fog and rain

Nitrogen and sulphur were of special interest in this study because these elements are believed to be responsible for excessive nutrient inputs and acidification in forests (Nihlgård, 1985). Despite the significantly lower overall duration of RF (227 h, cf. Table 1) compared to FAI (349.5 h), deposition by RF was between a factor 1.4 (phosphate) and 12.5 (fluoride) higher. Deposition of nitrate, sulphate, and ammonium by RF was 5.5, 7 and 6.4 times greater than by FAI. This is due to the combined effect of considerably higher ion concentrations and larger liquid water fluxes during RF. The ratios of FAI vs. RF deposition strongly depended on the relative occurrence of RF, which may vary considerably between years (Tables 4, 5). Thus, the ratios reported here are likely to be subject to strong inter-annual variations. Average occult deposition per day for the various ions is shown in Table 6. Because of droplet-size dependent fog chemistry, these estimates are subject to error that cannot be quantified. According to Collett et al. (1993, 1999) and Seinfeld and Pandis (1998), ammonium, nitrate and sulphate are enriched in small droplets, whereas soil dust cations such as calcium and magnesium are enriched in the larger droplets (larger CCN). Since small droplets are less efficiently collected by a CASCC than larger ones there may be a systematic bias in our flux estimates. However, we expect this error to be relatively small.

3.8. Total nitrogen deposition

Occult nitrogen deposition measured between 21 September 2001 and April 2002 was 2.4 and 1.5 times greater than precipitation inputs of reduced and

Table 4
Comparison of the ratios of weighted mean fogwater vs. rainwater concentrations between Lägeren (this study; RF = radiation fog, FAI = fog associated with atmospheric instability) and studies at Bern (Switzerland), Waldstein (Germany), and Clingmans Peak (North Carolina)

Ion	Lägeren		Bern[a]	Waldstein[b]	Clingmans peak[c]
	RF	FAI			
NO_3^-	54	23	35	24	13
SO_4^{2-}	35	11	60	18	13
Cl^-	7	3	77	8	32
Na^+	7	3	23	5	26
NH_4^+	66	25	74	25	14
Ca^{2+}	11	6	110	7	33
H^+	11	10	3	16	6

[a] Fuhrer (1986).
[b] Thalmann et al. (2002).
[c] Dasch (1988).

Table 5
Comparison of weighted mean concentrations of major ions, and pH of fog associated with atmospheric instability (FAI) and radiation fog events

	NO_3^-	SO_4^{2-}	NH_4^+	pH
FAI				
Lägeren (This study)	623	338	738	4.6
Mount Rigi, Switzerland (Collett et al., 1993)	520	430	1100	5.2
Waldstein, Germany (Thalmann et al., 2002)	646	438	926	4.1
Mont Tremblant, Québec (Schemenauer et al., 1995)[a]	170	339	239	3.7
Radiation fog (RF)				
Lägeren (this study)	1200	678	1548	4.3
Lägeren (Joos and Baltensperger, 1991)[b]	547	660	1408	5.2
San Joaquin Valley, California (Collett et al., 1999)	483	117	1008	6.5
Mantova, Italy (Fuzzi et al., 1996)	1594	2724	5771	6.6

[a] Unweighted means.
[b] Recomputed from original data, considering stationary measurements only.

Table 6
Ionic fluxes per day by occult and wet-only deposition in μeq m^{-2} d^{-1} and as percentage of total wet + occult deposition between 21/9/2001 and 10/4/2002

Ion	Occult		Wet	
	(μeq m^{-2} d^{-1})	(%)	(μeq m^{-2} d^{-1})	(%)
F^-	2.4	32	5.0	68
Cl^-	4.6	17	21.6	83
NO_3^-	58.8	61	38.2	39
SO_4^{2-}	38.8	50	38.4	50
Na^+	8.2	14	51.6	86
NH_4^+	75.9	70	31.9	30
K^+	3.6	26	10.2	74
Mg^{2+}	0.4	19	1.9	81
Ca^{2+}	4.7	23	15.7	77
H^+	3.7	23	12.5	77

oxidized nitrogen, respectively (Fig. 1). The contribution of occult deposition of reduced nitrogen to total wet + occult input is greater than that of oxidized nitrogen due to the stronger ammonium enrichment in fogwater compared with nitrate.

Total atmospheric nitrogen deposition was estimated for the Lägeren research site to amount to 26.2 kg ha^{-1} yr^{-1} (Table 7), which is clearly above the average N-input (16.8 kg ha^{-1} yr^{-1}) to European forests (MacDonald et al., 2002). The relative contributions of the various deposition pathways to total annual atmospheric nitrogen deposition is shown in Table 7. Compared to wet deposition fog adds 62% nitrogen to the annual budget, and 187% to the seasonal budget during the winter half year. The estimated share of occult deposition in the total atmospheric nitrogen deposition budget is 16.5%. These numbers reflect the fact that RF is almost absent during the warm season when precipitation inputs are largest.

4. Discussion

4.1. Fogwater

In agreement with several studies (Collett et al., 1993; Schemenauer et al., 1995; Fuzzi et al., 1996; Wrzesinsky and Klemm, 2000; Thalmann et al., 2002), fogwater composition was dominated by ammonium, nitrate, and sulphate. Their median contribution to the total ionic strength was the same for both types of fog with approximately 85%, and their relative contribution was constant throughout the events. This points to the influence of anthropogenic ammonium-containing aerosols and gases of mostly local to regional origin that are dissolved in fogwater. In contrast, wet-only and throughfall water (data not shown) show a more even contribution of the various ions. A fog study conducted in 1985/1986 21 km west of Lägeren (Sigg et al., 1986) showed a similar persistence of the ratios between NH_4^+, NO_3^- and SO_4^{2-}, despite variable proportions of their gaseous precursors (NH_3, NO_2, SO_2) in the atmosphere. Sigg et al.'s (1986) hypothesis for this behavior was the relatively constant proportions in the precursor aerosols. They assumed that the aerosols provide a substantial fraction of these components in fogwater. Still, Joos and Baltensperger (1991) showed that at least 40±6% of nitrate and 20±10% of ammonium concentrations in fogwater at Lägeren sampled during the winter of 1986/1987 could be attributed to gas phase reactions. From this we deduce that a combination of relatively constant precursor aerosol concentrations and complex interactions between the precursor gases was most likely

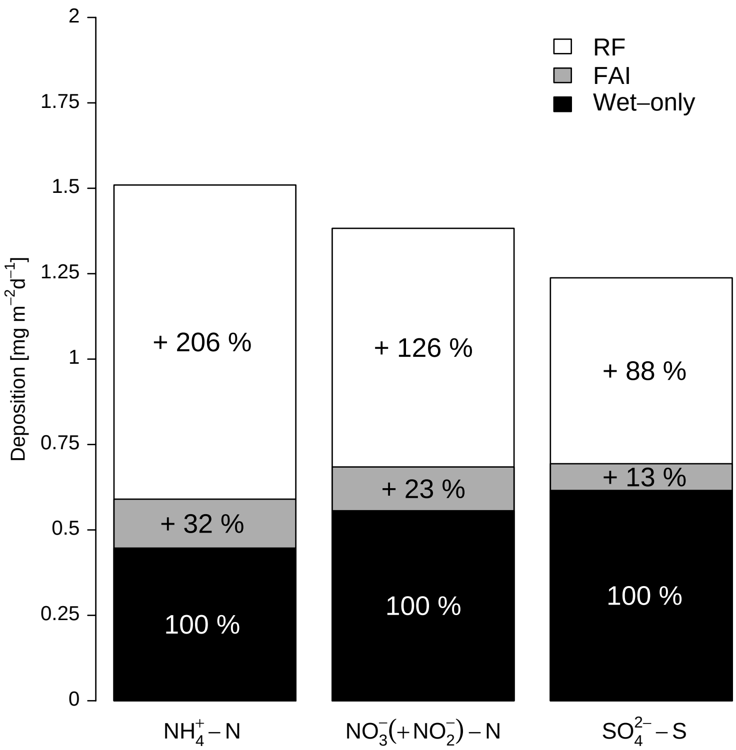

Fig. 1. Wet and occult reduced/oxidized nitrogen and sulphur input per day (mg m^{-2} d^{-1}) between 21/9/2001 and 10/4/2002. The numbers are based on wet-only deposition as 100%.

Table 7
Estimated relative (and absolute) contribution of the different deposition pathways to total annual atmospheric nitrogen deposition

Deposition	Percentage	kg N ha^{-1} yr^{-1}
Wet and occult N deposition	42.9%	11.2
By FAI fog	3.9%	1.0
By RF	12.5%	3.3
By precipitation	26.5%	6.9
Dry N deposition	57.1%	15.0
Gaseous components	48.3%	12.7
Aerosol particulate matter	8.8%	2.3
Total N deposition	100.0%	26.2

Wet deposition was measured between 25 July 2001 and 30 April 2002 and then scaled up to annual values using median concentrations in combination with measured precipitation. “Gaseous dry” and “aerosol particulate” are taken from Eugster (1999) for a representative area approx. 100 km SW of Lägeren. Critical load as estimated by Rihm (1996) for calcareous forests in Switzerland.

responsible for the constant ratios observed during our campaign.

The concentrations of the three major ions during FAI were similar to values observed by Collett et al. (1999) and Thalmann et al. (2002) (Table 5). The concentrations of ammonium and sulphate were somewhat lower than on Mount Rigi, whereas nitrate concentration was higher. The difference in nitrate and ammonium concentrations can partly be explained by differences in emission from the close-by intensive agriculture around Mount Rigi, as opposed to the proximity of Lägeren to major traffic roads and the Zurich international airport.

At Waldstein, where FAI was the only type of fog occurring, concentrations of ammonium and sulphate were significantly higher compared with this study, whereas nitrate concentrations were in the same range. Measurements of Schemenauer et al. (1995) on a rather remote mountain ridge of similar height in southern Québec with no important upwind emission sources showed a different picture with concentrations consistently lower than at Lägeren and sulphate as the dominant ion.

RF concentrations at Lägeren were relatively high compared with other studies (cf. Table 5). Except for the study from southern Québec, the proportions of the three major ions at all locations showed a major influence of ammonium and a minor contribution of sulphate. At Lägeren the ratio of ammonium versus nitrate was considerably lower (Table 5). In other words, agriculture has a greater relative importance than traffic in other regions compared with the Lägeren site. Consequently, the pH values at Lägeren were lower due to a smaller neutralization effect by ammonia.

4.1.1. Rainwater

Weighted mean ionic concentrations in our rainwater samples were in the range of the measurements at the Waldstein research site in northeastern Bavaria, Germany (Thalmann et al., 2002). The median share of sodium and chloride at Lägeren amounts to roughly 34% of the total amount of ions in rainwater and to approximately 7% in fogwater (FAI and RF almost equal), respectively. This clearly illustrates the large-scale influence of air masses advected from the North Atlantic as it is assumed that sodium has no other significant source (Lin et al., 1999). The results from Waldstein support this assumption: the contributions of chloride and sodium were about 2.5 to 3 times lower than at Lägeren (Thalmann et al., 2002), which is most likely due to the more continental character of the research site. The difference of the relative contribution of sodium and chloride between rain and fog was very similar to the Lägeren results, which suggests a rather local to regional character of non-sea-salt ions in fogwater.

4.2. Comparison of ion concentrations in fog and rain

Fogwater concentrations at Lägeren were between 3 and 66 times higher than rainwater concentrations (Table 4). The enrichment factor between fogwater and precipitation was highest for ammonium, underlining the rather local influence of ammonia emissions from farming and livestock keeping. Nitrate was also strongly enriched, which is indicative of mainly local to regional NO_2 emissions. Enrichment factors of the two ions were almost identical for FAI in this study and the Waldstein campaign (Thalmann et al., 2002) irrespective of absolute concentrations. In other words, for similar fog types at different locations, the fog(FAI)-to-rain concentration ratio of ammonium may be quite constant, with rainwater concentrations around 4% of fogwater concentrations. Sulphate enrichment in rainwater can partly result from the longer atmospheric residence time (and thus vertical and horizontal transport distance) of SO_2 during dry periods compared to ammonia and NO_2 because of its lower reactivity (Seinfeld and Pandis, 1998). Table 4 indicates that the enrichment of protons is not necessarily correlated with the enrichment of other ions. The enrichment factors at Clingmans Peak (Dasch, 1988) vary much less than at the other sites. This is most likely due to the high elevation of Clingmans Peak. In summary, fogwater enrichment factors described in literature are in a similar range as the ones we found at Lägeren. However, the enrichment in radiation fogwater seems to be at the high end of the range. Enrichment is strongest for the most important anthropogenic constituents NO_3^-, SO_4^{2-} and NH_4^+, and lowest for ions associated with long-range transport such as Na^+ and Cl^-.

4.3. Deposition of nutrients by fog and rain

Daily occult deposition, measured by the same setup at the Waldstein research site in Germany (Thalmann et al., 2002) was about 33% higher for most ions. Only sodium deposition was higher at Lägeren, possibly because of a greater influence of the ocean source. The larger deposition fluxes at Waldstein reflect the 3.75 times greater fogwater flux per day due to a considerably higher fog frequency. Indirect cloudwater deposition estimates from throughfall measurements, as performed by Dasch (1988) at Clingmans Peak, North Carolina, showed values in a similar range for most ions, except for sulphate and protons. The difference in the latter two is due to a strong enrichment of sulphate in fogwater collected in the 1980s, which led to a much higher acid deposition flux. The good agreement between the other ions is surprising because Dasch (1988) estimated a much higher cloudwater flux (0.5 mm d^{-1}).

4.4. Total nitrogen deposition

A comparison of nitrogen and sulphur deposition fluxes found in the literature is shown in Table 8. At Waldstein (Thalmann et al., 2002), fogwater fluxes and precipitation amounts were between 45% (wet NO_3^--N) and 200% (wet NH_4^+-N) larger than at Lägeren, which is due to the significantly greater water fluxes. Another recent study at Zöbelboden (900 m a.s.l), eastern Austria (Katzensteiner, 2000), revealed much larger wet deposition fluxes due to higher concentrations and greater water fluxes as well. At Whiteface Mountain (1000 m a.s.l), New York, Miller et al. (1993) found similar nitrogen fogwater inputs in the late 1980s but with a predominance of NO_3^--N over NH_4^+-N compared to Lägeren. Sulphur inputs were much higher with SO_4^{2-}-S being the most important inorganic input of the three major species.

Still, annual atmospheric nitrogen deposition at Lägeren is smaller than at many other places in the industrialized world. For instance, Katzensteiner (2000) estimated 20 kg ha^{-1} yr^{-1} nitrogen input by wet deposition alone, compared to the 6.9 kg ha^{-1} yr^{-1} at Lägeren. Nevertheless, current loads at Lägeren considerably exceed the critical load for calcareous forests in Switzerland of 15 kg ha^{-1} yr^{-1} (Rihm, 1996). According to the modelling approaches by Eugster (1999), dry deposition alone would amount to roughly the critical load and all additional wet and occult nitrogen input contributes to eutrophication.

Table 8
Comparison of occult and wet deposition of anorganic nitrogen and sulphur between various studies (mg m^{-2} d^{-1}), n.d. indicates "not detected"

	NH_4^+-N		NO_3^--N		$\sum$ nitrogen		SO_4^{2-}-S	
	Occult	Wet	Occult	Wet	Occult	Wet	Occult	Wet
Lägeren (this study)	1.06	0.45	0.83	0.56	1.89	1.01	0.62	0.62
Waldstein (Thalmann et al., 2002)	1.16	1.36	0.82	1.01	1.98	2.37	0.62	1.04
Whiteface Mt. (Miller et al., 1993)	0.79	0.74	0.68	1.18	1.48	1.92	1.64	2.33
Zöbelboden (Katzensteiner, 2000)	n.d.	3.26	n.d.	2.22	n.d.	5.48	n.d.	2.19

Deposition studies at several sites in eastern Austria by Katzensteiner (2000) showed that with a deposition of more than 15–25 kg N ha^{-1} yr^{-1}, nitrogen export from the system takes place, depending on soil and vegetation type. At Lägeren, the upper threshold value is slightly exceeded. The high pH values found in throughfall water, however, suggest that soil acidification may not be of primary concern at the Lägeren site, whereas aboveground nitrogen uptake and subsequent mutational imbalance may be critical.

5. Conclusions

Two different types of fog were observed at Lägeren: radiation fog (RF) and fog associated with atmospheric instability (FAI). It was shown that these two fog types exhibit large differences with respect to fog microphysical properties. Basically, RF showed much larger median fogwater fluxes than FAI, and concentrations were significantly enhanced in RF compared with FAI. The relative fogwater composition seemed to be determined mostly by local to regional emissions, whereas absolute concentrations were dependent on the synoptic weather pattern. Long range transport appeared to be less important. The dominant ions in fogwater were nitrate, sulphate, and ammonium.

Nutrient concentrations found in precipitation at Lägeren were relatively low. Fogwater enrichment factors were between 3 and 66. Enrichment was highest for derivates of locally generated emissions with relatively short atmospheric residence times such as ammonium. As opposed to fogwater, dominant ions in precipitation were sulphate and sodium. This was attributed to a more pronounced large-scale influence on precipitation at Lägeren from the North Atlantic.

The considerably larger water fluxes and nutrient concentrations of RF resulted in much higher nutrient deposition compared with FAI. Although average daily occult deposition was relatively low compared with other studies, its contribution to total wet plus occult deposition was very high. During the winter half year, when RF was quite frequent, fog interception was even the dominant pathway for nitrogen, whereas precipitation was more important with regard to base cations and acidity. Mean daily nitrogen input by fog was 1.9 times greater than wet deposition between 21 September 2001 and 10 April 2002. On an annual basis its contribution was still an additional 62%, despite the near absence of RF during the summer half year. Occult deposition contributes roughly 16.5% to the total N-deposition of 26.1 kg ha^{-1} yr^{-1}, which exceeds the critical load of the forest ecosystem at Lägeren. This is a clear indication that the emission reduction measures taken so far have not yet been sufficient to solve the eutrophication problem of forest ecosystems in Switzerland.

Acknowledgements

This work was funded by the Swiss National Science Foundation, grants 20-63484.00 and 21-55554.98. W. E. was also supported by a Hans Sigrist Fellowship from the University of Bern. We are grateful to M. Oetliker, S. Schmid, and J. Schenk for their help and support during the field experiment, H. Wernli for the chemical analysis. In addition, many thanks also to B. Buchmann and T. Bruggisser from the Swiss Federal Laboratory for Materials Testing and Research (EMPA) for making the measurement tower available for us and to F. Joos from the Institute of Climate and Environmental Physics of the University of Bern for providing the original data of the 1986/1987 field campaign. E. Thalmann is acknowledged for English proof-reading.

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