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Nitrogen deposition along differently exposed slopes in the Bavarian Alps

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HIGHLIGHTS

- NO_2 and NH_3 air concentrations low
- Open field nitrogen deposition relatively independent of location
- Nitrogen throughfall deposition high at wide-exposed sites
- Extreme inversion frequency in Bavarian Alps in November 2011
- Different vertical distribution of air temperature along south- and north-facing slopes

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ABSTRACT

The Alps are affected by high nitrogen deposition, particularly in the fringe of the Northern and Southern Alps. In the framework of a two-year monitoring study performed in 2010 and 2011, we investigated the ammonia and nitrogen dioxide air concentration and ammonium and nitrate deposition at different altitudes between 700 and 1600 m a.s.l. in the Garmisch-Partenkirchen district in the Upper Bavaria region (Germany). Four-weekly measurements of deposition collected with bulk open field samplers and under-crown were performed in a profile perpendicular to the axis of the Loisach valley; measurements were conducted at eight sites. Whereas open field deposition ranged from 5 to 11 kg ha^{-1} a^{-1}, nitrogen throughfall has reached up to 21 kg ha^{-1} a^{-1}. Data from the valley and the slopes were compared with measurements performed on the platform of the Environmental Research Station Schneefernerhaus (Zugspitze) at an altitude of 2650 m a.s.l. For the rough estimation of the total yearly deposition rate of nitrogen, the canopy uptake model was applied. By regarding nitrogen uptake by the trees, total deposition can exceed the throughfall in all sites by up to 50%. Additionally, we estimated the total deposition from the sum of wet and dry deposition. On the one side, the wet deposition could be extrapolated from the open field deposition. On the other side, we used the inferential method to calculate the dry deposition on the basis of NH_3 and NO_2 air concentrations and their literature based deposition velocities. Since fixed deposition velocities are inappropriate particularly in complex orography, we tried to find correction factors based upon terrain characteristics and meteorological considerations. Temperature monitoring at the eight sites and wind measurements at two sites provided some evidence for the semi-empirical parameterization. Due to numerous imponderabilities, the results of the two methods were not consistent for all sites.

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1. Introduction

Nitrogen can be both a limiting nutrient and a pollutant in ecosystems (Fenn et al., 2009). The Alps, situated in the center of Europe, are dissected by high-traffic roads and surrounded by urban agglomerations and agricultural areas causing high emissions of air pollutants like nitrogen compounds. The highest impacts of such air pollutants on ecosystems are recorded in the northern and southern mountain fringes and in the Alpine foothills, particularly in Bavaria, Northern Switzerland and Lombardy (Lorenz et al., 2008; Kaiser, 2009). Despite a slightly negative trend (Rogora et al., 2006; Lorenz et al., 2008; Eickenscheidt and Brumme, 2012; ICP Forests, 2012), the input of both reduced and oxidized nitrogen into forest ecosystems in the alpine and subalpine areas is still well above the estimated critical loads (10–20 kg ha^{-1}) of nutrient nitrogen in many spruce forest sites (Jandl et al., 2012). The observed acceleration in tree growth rates is likely to be the result of fertilization by nitrogen, enhanced atmospheric CO_2 concentrations, and temperature increase (Eriksson and Johansson, 1993; Hunter and Schuck, 2002). However, as long as forest ecosystems in the Southern and Northern Limestone Alps are still deficient in nitrogen, high deposition rates increase the demand (Jandl et al., 2012). A further nitrogen deposition may consequently influence nutrient balance, soil acidification, and groundwater composition of forests (Aneja et al., 2001; MacDonald et al., 2002; Wuyts et al., 2011) and has an impact on the forest floor composition (Bernhardt-Römermann et al., 2006).

Nitrogenous gases such as nitrogen oxides and ammonia as well as ammonium and nitrate containing particles can be deposited as dry and wet deposition. Particularly on mountain ridges, where wind speed is higher than along the slopes, the wet deposition by rain and snow can be enhanced by occult deposition when fog droplets are trapped by foliage and branches of trees. Particularly spruce forests represent a much larger receptor than other vegetation types due to their higher aerodynamic roughness and their ability to capture fine particles. Forests at high elevations are likely to be wet for longer periods of time than vegetation in the valleys because of the greater cloud immersion (Lovett and Kinsman, 1990). Frequent fog, cloud events (Kalina et al., 2002), and rime (Burns, 2003) enhance the rates of atmospheric N deposition. The dry deposition of pollutants like ammonia increases with decreasing atmospheric stability (Phillips et al., 2004). Generally, the deposition rate in forests is largely influenced by orographic and meteorological characteristics such as altitude, orientation of the valley and exposure to the main wind directions, slope aspect, and slope steepness (Segal et al., 1988).

For the estimation of total nitrogen deposition, wet and dry deposition input data are needed. Compared with wet deposition, which is easily measurable by wet-only or bulk funnels, it is much more difficult to determine the dry deposition of gases and aerosols. Micrometeorological methods for measuring dry deposition can be performed only in homogenous terrain and require sophisticated and expensive instrumentation (Schmitt et al., 2005). Therefore, surface analysis methods such as throughfall measurement are much more prevalent in measuring campaigns. The estimation of dry deposition by under-crown measurements shows some uncertainties related to canopy interaction processes such as leaching and uptake as the throughfall flux of nitrogen can differ from the sum of wet and dry deposition (Draaijers and Erisman, 1995). Methods separating the contribution of dry deposition from canopy exchange processes, so-called canopy budget models, are based upon a work of Ulrich (1983) and have been developed and refined by several authors (Andersen and Hovmand, 1999; Devlaeminck et al., 2005; Thimonier et al., 2004; Schmitt et al., 2005; Adriaenssens et al., 2012).

With the inferential method, dry deposition can be estimated by multiplying the atmospheric gas concentration and the appropriate deposition velocity. Deposition velocities have been determined in many measuring campaigns by different methods, for a wide range of forest stands. In many studies, data were taken from the literature and adapted for the particular conditions (Schmitt et al., 2005).

The aim of this study, which was part of the project KLIMAGRAD, was to quantify the annual nitrogen deposition into forests along a measuring transect including different altitudes and aspects in the vicinity of Garmisch-Partenkirchen/Bavaria. We performed bulk and throughfall deposition measurements to depict the general situation and estimated the total deposition. Firstly, we used the canopy budget method, refined by Devlaeminck et al. (2005), in order to include the possible nitrogen uptake. Secondly, we calculated the total nitrogen input by the inferential method based on air concentration measurements and literature-based deposition velocities. As fixed deposition velocities are rather appropriate for flat terrain and homogenous forest stands and unlikely to represent the reality of a complex orography, we modified the deposition velocities on the basis of a semi-empirical parameterization. The installation of a basic meteorological instrumentation aimed to characterize the study sites.

2. Material and methods

2.1. Location and experimental sites

The study area was situated in the calcareous Alps near Garmisch-Partenkirchen in the Upper Bavaria region in Southern Bavaria (Germany). It consisted of eight sites along a transect perpendicular to the axis of the Loisach valley (Fig. 1). This region is characterized by an average annual precipitation amount of 1350 mm in the valley (719 m a.s.l.) and 2000 mm on top of the Zugspitze (2964 m a.s.l.); the mean annual air temperature (1961–1990) in Garmisch-Partenkirchen is 6.5 °C and on the Zugspitze −4.8 °C with a strongly variable monthly lapse rate between −0.35 °C/100 m in January and −0.61 °C/100 m in July (Kirchner et al., 2013). The prevailing wind directions on top of the mountains are SW and W. At lower altitudes, winds are influenced by topography; frequently mountain and valley breezes dominate the wind pattern.

The eight sites investigated exhibited different altitudes and expositions (Table 1). The sites were located in 80–150 year old Norway spruce forests [*Picea abies* Karst. (L.)] at 700, 1000, 1300, and 1600 m a.s.l on the northern and southern slopes of Mount Kreuzeck. On the opposite slope of the W–E oriented Loisach valley, two additional plots at 700 and 1000 m a.s.l on Mount Kramer were chosen. In the vicinity of each forested site, we selected an open field plot within a clearing. All plots are uninfluenced or scarcely influenced by traffic and agricultural emissions. Because of remoteness, lack of electric power, severe weather conditions, and widely fluctuating snow depths in winter, the logistic difficulties were enormous; some of the forest sites could only be reached by four-wheel drive vehicles or by foot in summer and ski mountaineering equipment in winter. Aside from the gradient study, simultaneous measurements were performed at the platform of the Environmental Research Station Schneefernerhaus/Zugspitze (UFS) in 2650 m a.s.l.

2.2. Measurement technique and analytical procedures

Deposition is calculated by multiplying the precipitation amount measured by the bulk samplers by the ion concentration in the sampled precipitation. Precipitation was sampled in open field and under crown by using bulk samplers (Ø = 200 mm) with a sampling interval of four weeks. During winter months, bulk samplers with greater funnel apertures (Ø = 40 mm) were used. In the framework of two years, we collected 26 open field bulk and 156 throughfall water samples at each site of the transect. Due to the rough terrain with steep slopes and limited accessibility, we exposed only six bulk samplers at each forest site and one in the open field, slightly in discordance with the recommendations for deposition measurements in forested areas (Starr et al., 2007). With heavy snow fall events, sampling intervals of

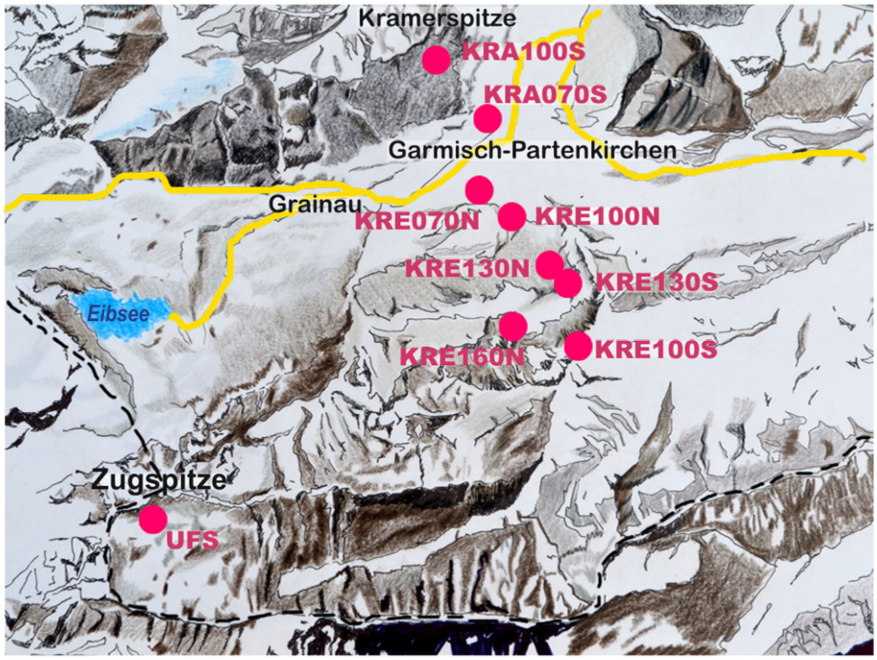

Fig. 1. Study sites in the district of Garmisch-Partenkirchen.

open field bulk samplers were reduced to two weeks; snow samples were stored at sub-zero temperatures and pooled before analysis. At the UFS, two parallel open field heated bulk samplers were used. The concentrations of NH_4^+, NO_3^-, SO_4^{2-}, PO_4^{3-} and Cl^- in bulk and throughfall were analyzed either by a segmented continuous flow analyzer or ion chromatography with conductivity detection, while the concentrations of Ca^{2+}, Mg^{2+}, K^+ and Na^+ were determined using the inductively coupled plasma atomic emission spectrometry (ICP–AES) at the analytical laboratory of the Helmholtz Zentrum München. The quality of the data records was checked by computing the ion balance equation.

Atmospheric air concentrations of NO_2 and NH_3 were monitored in the open field plots by using ventilated battery-powered samplers (flow rate: 2.7 $m^3\,d^{-1}$) which are particularly appropriate for remote areas (detection limit: 0.05 $\mu g\,m^{-3}$); due to the ventilation, small additional ammonium and nitrate aerosols and droplets (<3 μ) were included as well; The samplers were changed with the same frequency as the bulk samplers. They contained two different impregnated glass-fiber filter halves for the adsorption of reduced and oxidized nitrogen. After exposure, the filter halves were analyzed for NO_2 and NH_3 by ion chromatography and photometry, respectively. The air sampling rate results from the battery voltage before and after exposure (Kirchner et al., 1999; Kirchner et al., 2005). Two field blanks were stored at KRE160NF and at the platform of Schneefernerhaus. At UFS, parallel NH_3 measurements with an electric-powered pure gas sampling denuder system (Sutton et al., 1994) were performed by the German Environmental Protection Agency (UBA).

The measurement of air temperature and relative humidity was performed by using data loggers with integrated sensors (thermobuttons 23, www.proges.com/plug-and-track/index.html); data were recorded at 30-minute intervals. The sensors were protected by a commercial radiation shield (Friedrichs, Schenefeld, Germany) and exposed at a height of 2 m above ground level in each open field plot. Near KRE100SF and KRE160NF, additional wind measurements were started in 2011 or 2012 by the Technische Universität München. Lacking electric power and the restricted budget did not allow sophisticated meteorological measurements like fog detection along the profile.

2.3. Methods for estimation of total nitrogen deposition

The total nitrogen deposition (TD) consists of wet (WD) and dry (DD) deposition of all nitrogenous compounds (x) occurring in the atmosphere (Eq. (1)). Deposition rates are balanced by canopy uptake (CU) and throughfall (TF).

$$TD = DD_x + WD_x = CU_x + TF_x. \tag{1}$$

Wet deposition (WD) ideally should be measured by wet-only samplers which are open if precipitation occurs. Mostly because of the lack of electricity, as in our study, wet deposition is usually estimated

Table 1
Experimental sites consisting of open field (F) and spruce [*Picea abies* Karst. (L.)] forest (B) plots with geographic parameters.

Experimental site	Location	Latitude	Longitude	Altitude [m]	Exposition
KRE100S F/B	Kreuzeck: Reintal (slope)	47°27′04″N	11°06′14″O	1000	E
KRE130S F/B	Kreuzeck: Hausberg (crest)	47°27′59″N	11°05′42″O	1300	SW
KRE160N F/B	Kreuzeck: Kreuzalm (crest)	47°27′22″N	11°04′28″O	1600	N
KRE130N F/B	Kreuzeck: Hausberg (slope)	47°28′05″N	11°05′36″O	1300	N
KRE100N F/B	Kreuzeck: Tonialm (slope)	47°28′22″N	11°04′50″O	1000	N
KRE070N F/B	Garmisch-P. (valley)	47°28′50″N	11°04′25″O	700	N
KRA070S F/B	Garmisch-P. (valley–slope)	47°29′40″N	11°04′19″O	700	SE
KRA100S F/B	Kramer (slope)	47°29′57″N	11°02′56″O	1000	S
UFS	Schneefernerhaus (slope)	47°25′00″N	10°58′46″O	2650	S

from bulk deposition (BD) by adopting adjustment factors correcting for dry deposition. We applied average BD to WD conversion factors, given in the literature, which are about 0.89 for NH_4^+ and 0.83 for NO_3^- for example, but lower than 0.70 for Ca^{2+}, K^+, and Mg^{2+} (Draaijers et al., 1996; Gauger et al., 2000; Balestrini and Tagliaferri, 2001; Rogora et al., 2006).

2.3.1. Canopy budget method

To estimate dry deposition we performed similar calculations as recommended by Devlaeminck et al. (2005) based on the canopy budget method of Ulrich (1983). The canopy budget model is described in detail in de Vries et al. (2001). This method was developed to separate fluxes of the base cations (bc = Ca^{2+}, K^+, Mg^{2+}) due to dry deposition from the fraction caused by the internal leaf leaching. As these elements are assumed to have the same deposition velocity as Na^+ a dry deposition factor (DDF) can be calculated [$mol\,ha^{-1}\,a^{-1}$].

$$DDF = \frac{(TF-WD)_{Na}}{WD_{Na}}. \tag{2}$$

Dry deposition of the base cations (bc) is then calculated by multiplying the wet deposition with DDF:

$$DD_{bc} = WD_{bc}DDF. \tag{3}$$

Canopy leaching CL will be calculated in the following way:

$$CL_{bc} = NTW_{bc} - DD_{bc}. \tag{4}$$

The net throughfall water (NTW) is calculated by subtracting wet deposition from throughfall. Negative NTW values ($mol\ ha^{-1}\ a^{-1}$) indicate admission into canopy.

$$NTW = TF - WD. \tag{5}$$

Corresponding to de Vries et al. (2001) who assume that the leaching of base cations from the canopy equals the uptake of protons and ammonium in a proportion to their fluxes in wet deposition and throughfall, the canopy uptake of NH_4^+ can be calculated by the sum of the exchanged cations (Devlaeminck et al., 2005):

$$CU_{NH_4} = \frac{(WD+TF)_{NH_4}}{(WD+TF)_{NH_4} + (WD+TF)_H} CL_{K+Ca+Mg}. \tag{6}$$

Due to the further assumption that canopy exchange of SO_4^{2-}, Cl^- and Na^+ may be negligible and the uptake of NO_3^- being relatively small in comparison to NH_4^+ (Meesenburg et al., 2009) the dry deposition of these elements can be estimated by using NTW.

Using this method, the dry deposition rate can be indirectly calculated from Eq. (1). Dry deposition of reduced substances includes both more or less completely gaseous NH_3 and particulate NH_4^+.

$$DD_{NH_4} = TF - WD + CU. \tag{7}$$

All Eqs. (1)–(7) were calculated on an annual basis.

2.3.2. Inferential method

The second method for estimating dry deposition of nitrogen (Schmitt et al., 2005) relies on multiplying air concentrations (c_x) of gaseous components (x = NH_3, NO_2) by their deposition velocities (v_d) (8); the total deposition results from the sum of dry (DD) and wet deposition (WD) rates. The dry deposition can be calculated in the following way:

$$DD = v_{d(lit)}\ c_x. \tag{8}$$

We used averaged deposition velocities ($v_{d(lit)}$) obtained from the literature (Wyers et al., 1992; Erisman et al., 1993; Rihm, 1996; Zapletal, 1998; Ferm and Hultberg, 1999; Andersen et al., 1999; Horvath et al., 1998; Rattray and Sievering, 2001; Renard et al., 2004; Thimonier et al., 2004; Schmitt et al., 2005; Zimmermann et al., 2006). The selected values for spruce stands were 3.1 $cm\ s^{-1}$ for NH_3; the deposition velocities for particulate NH_4^+ were lower by a factor of 5–10 than v_d for ammonia (Schmitt et al., 2005). For NO_2 we selected 0.4 $cm\,s^{-1}$, similar to NO_3^-. According to Marner and Harrison (2004) and Zbieranowski and Aherne (2012), seasons have been taken into account by modifying $v_{d(lit)}$ by factors of 1.1 in spring, 1.2 in summer and no modification (1.0) in autumn. According to most studies reporting a lower deposition velocity in winter (Fischer-Riedmann, 1995), we assumed a factor of 0.8 in winter without considering the different length of the winter period at different altitudes as done by Schmitt et al. (2005).

The parameterization is based upon semi-empirical factors regarding slope inclination (k_{incl}), wind exposure (k_{wind}), frequency of inversions (k_{inv}) and upslope winds (k_{up}) (Benedikt et al., 2013). The factors have been determined rather subjectively on the basis of long experience in the area and have been partly confirmed by the concurrent meteorological measurements. Parameters as surface wetness time could not be observed (Lovett and Kinsman, 1990). Similarly, cloud water deposition, which plays an important role above 1000 m a.s.l., could not be taken into account explicitly for lack of measurements (Miller et al., 1993).

Slope inclination varies between 6% at the KRE070NB site and 87% at the KRA100SB site. The inclination of each slope allows a tentative parameterization of k_{inv} (inclination factor) between o.7 and 1.5. The parameterization of k_{wind} (wind exposure factor) is based upon the analysis of wind data registered by two measuring masts (6 m) at KRE100SF (mean velocity in 2011: 0.27 ms^{-1}) and KRE160NF (mean velocity in 2011: 2.47 ms^{-1}) and the extrapolation of raster data from the Bavarian wind map (http://www.stmwivt.bayern.de/fileadmin/Web-Dateien/Dokumente/energie-und-rohstoffe/Bayerischer_Windatlas.pdf). Caused by the lack of further meteorological data and the inaccuracy of such theoretical maps (spatial resolution: 50 × 50 m) the parameterization of k_{wind} is rather tentative. As the frequency of inversions which impede vertical motions decreases with elevation and south-faced slopes are less influenced by stable layers than north-faced slopes, k_{inv} (inversion factor) was parameterized between 0.8 (KRE070NB) and 1.0 (KRE130SB). Due to the lack of further meteorological towers, upslope wind effects could be hardly parameterized; north-faced slopes (KRE 100NB, KRE130NB, KRE160NB) may be less influenced by upslope winds than sun-exposed sites (KRA100SB, KRE130SB, KRA070SB).

So, the total deposition velocity is calculated by using the following formula and is based on factors which have been semi-empirically determined:

$$k_{tot} = k_{incl} * k_{wind} * k_{inv} * k_{up}, \tag{9}$$

$$v_{dcorr} = v_{dlit(saison)} * k_{tot}. \tag{10}$$

The semi-empirical deposition velocity correction factors for dry deposition (gas phase and partly aerosol deposition) are given in Table 2.

3. Results and discussion

3.1. Meteorology

Precipitation as the most significant mechanism of deposition shows an altitudinal increase from 1287 in the valley to 1537 mm per year on the highest monitoring plot in 2010 and 1277–1409 mm in 2011. The corresponding ranges of throughfall amounts were 758–1062 mm in 2010 and 787–1057 mm in 2011. Since there are many obstacles for reliably precipitation measurements in the mountains precipitation of the higher locations may be slightly underestimated; there are wind-

Table 2
Semi-empirical deposition velocity correction factors for the investigated forest sites based on terrain characteristics.

Site	Inclination (%) flat: − scarcely steep: + steep: ++ very steep: +++	k_{incl} 0.01 × inclination (%) + 0.6	Wind exposure extreme lee-side: −− lee-side: − windward: + crest: ++	k_{wind} 0.7 0.9 1.1 1.3	Frequency of inversions rare: − frequent: + very frequent: ++	k_{inv} 1.0 0.9 0.8	Upslope winds frequent: + very frequent: ++ extremely frequent: +++	k_{up} 1.1 1.2 1.3	k_{tot}
KRE100S B	(7) −	0.7	−−	0.7	+	0.9	+	1.1	0.5
KRE130S B	(34) +	0.9	++	1.3	−	1.0	+++	1.3	1.6
KRE160N B	(86) +++	1.5	++	1.3	−	1.0	+	1.1	2.1
KRE130N B	(34) +	0.9	+	1.1	−	1.0	+	1.1	1.1
KRE100N B	(71) ++	1.3	+	1.1	+	0.9	+	1.1	1.4
KRE070N B	(6) −	0.7	+	1.1	++	0.8	++	1.2	0.7
KRA070S B	(41) ++	1.0	−	0.9	+	0.9	++	1.2	1.0
KRA100S B	(87) +++	1.5	−	0.9	−	1.0	+++	1.3	1.7

induced errors particularly in winter. On top of the Zugspitze (2963 m a.s.l.), where daily maintenance of precipitation gauges is guaranteed, correspondent precipitation sums were 1953 mm in 2010 and 1793 mm in 2011. Due to the enhanced lee-side effect with snow drift from the crest, precipitation at UFS (2300 mm) was by 15% higher than on the top of Zugspitze.

During 1991–2000, averaged annual precipitation rates were 2087 mm at the Zugspitze and 1440 mm at the meteorological station in Garmisch-Partenkirchen (720 m a.s.l.) with a mean lapse rate of 29 mm/100 m; vertical increases of precipitation occur throughout the year, particularly correlated with cold fronts and northerly winds, whereas negative lapse rates are observed with south-westerly winds mostly during summer (Wastl, 2008).

The year 2011 was characterized by higher temperatures and more frequent inversions compared to 2010; relative air humidity was lower, whereas wind speed did not show huge differences between the study years (Schuster et al., 2013). The vertical distribution of temperature along the north-facing slope of Mount Kreuzeck (KRE070NF, KRE100NF, KRE130NF, KRE160NF) is shown in Fig. 2 on the basis of monthly means of four months representative of the four seasons. Annual means in 2010 and 2011 were 6.4 °C and 7.5 °C, respectively, at 700 m a.s.l. and 2.9 °C and 5.4 °C, respectively, at 1600 m a.s.l. with mean lapse rates of −0.39 °C/100 m and −0.25 °C/100 m. In winter, the mean monthly temperature increased up to 1300 m a.s.l., whereas temperatures mostly decreased during the rest of the year with the exception of November 2011. The frequency of inversions decreased with elevation during all months of 2010 and 2011. During the extremely dry and warm November 2011 (Schuster et al., 2013), inversions occurred between 700 and 1000 m a.s.l. in 82%, between 1000 and 1300 m a.s.l. in 58%, and between 1300 and 1600 m a.s.l. in 38% of the time. Additional lapse rate data which characterize the atmosphere between valley ground (Garmisch-Partenkirchen) and Zugspitze are given by Kirchner et al. (2013).

Due to the differences in solar radiation on slopes of different aspect, we expect that the north-facing slope of Mount Kreuzeck and the south-facing slope of Mount Kramer were characterized by different temperature regimes. Correspondingly, the vertical distribution of near-surface air temperatures on northern and southern slopes can differ (Tang and Fang, 2006; Moser et al., 2009). Schuster et al. (2013) experienced in the same region of Garmisch-Partenkirchen that mean inversion heights were about 220 m higher at southern than on northern slopes. On strongly stable days, increasing surface heating may cause rising or/and dissolution of inversion layers and the development of quite shallow upslope winds near the sun-exposed slope. Upslope winds, which are stronger (>1 ms^{-1}) than downslope winds, increase atmospheric mixing and may cause transport of pollutants from the valley ground. Upslope winds exhibit their maximum velocity approximately at the tree crown level (Orville, 1964).

Since anemometer measurements along the slopes could not be performed synchronously in 2010 and 2011, we tried to estimate vertical air movements by comparing the temperature difference between the sites of 1000 and 700 m a.s.l. along the two differently exposed slopes. For sunny days, we counted the hours in which temperature on the sun-exposed south-facing slope of Mount Kramer decreased with elevation, while still an inversion persisted on the heavily shaded north-facing slope of Mount Kreuzeck. Caused by the seasonality of inversions (Kirchner et al., 2013), the different incident solar radiation, and the frequency of sunny days in 2010 and 2011, we found a difference of 1–3 h in winter and less than 0.5 h in summer where we assume a high probability of upslope air flow (Fig. 3). On overcast days with or without precipitation, the vertical temperature distribution was similar along the two differently exposed slopes; similar results have been demonstrated by Segal et al. (1988).

3.2. Ventilated air sampler measurements

Annual means of NH_3 and NO_2 concentration measured by ventilated samplers are shown in Figs. 4 and 5. With the exception of the two sites in the valley of Garmisch-Partenkirchen, concentrations of both gases are low due to the long distance from possible sources of traffic, domestic wood burning, and agriculture. Concentrations are characterized by a seasonal cycle with maxima in winter for NO_2 and in summer for NH_3 which tend to decrease with increasing altitude. Due to the difference in upslope winds and a more intense venting of pollutants to upper layers (Segal et al., 1988) at Mount Kramer compared with Mount Kreuzeck particularly during the winter period, the ratios of NO_2 concentration monitored at KRA100SF to those of KRE100NF are 1.12 in winter and 0.89 in summer; the corresponding ratios for NH_3 are 0.92 in winter and 0.75 in summer. The main traffic sources (NO_2) are located rather in the center and in the northern parts of the valley, whereas the meadows with sporadic application of manure (NH_3) are located in the southern part of the valley ground (Löflund et al., 2002). The lowest plot on the north-facing slope of Mount Kreuzeck (KRE070N F/B) was influenced by some vehicle emissions during the Ski World Cup 2011. The situation at UFS with frequent 4-weekly concentrations <0.5 μg/m^3 seems to be rather decoupled from the situation in the valley. However, the seasonal dependence of NH_3 (or rather NH_x) concentrations measured at UFS during 2006–2011 is still noticeable with means of 0.19, 0.44, 0.51 and 0.29 μg/m^3 in spring, summer, autumn, and winter, respectively. Parallel NH_3 measurements performed by UBA resulted in concentrations of 0.05, 0.31, 0.27 and 0.07 μg/m^3 for the four seasons. The discrepancies between the two methods can be explained by the fact that our ventilated battery-powered samplers include particles and droplets <3 μ, whereas UBA uses an electric-powered pure gas sampling denuder system (Sutton et al., 1994). As small aerosols can hardly be deposited in bulk samplers, the use of battery-powered samplers is an advantage in comparison with diffusive passive samplers or denuder systems. Small N-aerosols may amount to 10–30% of the dry deposited gases which experienced Puxbaum and Gregori (1998), Ferm and Hultberg (1999) and Rattray and Sievering (2001) in the framework of several studies.

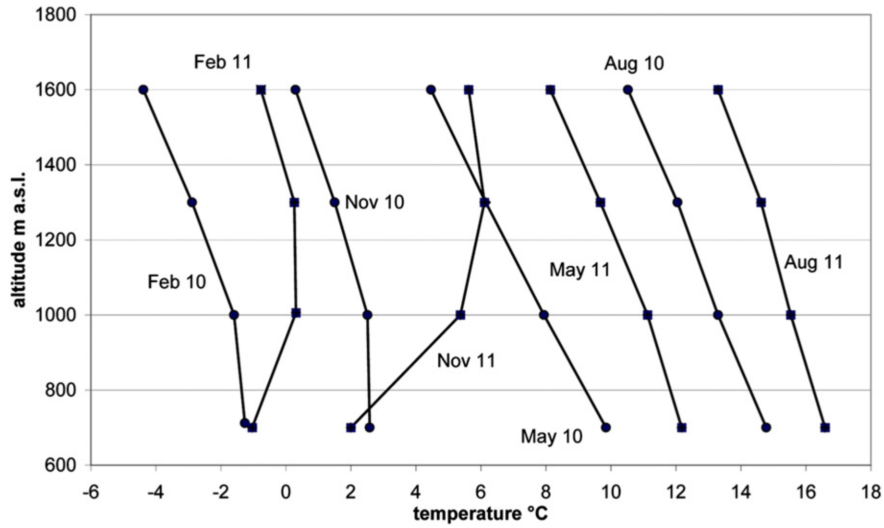

Fig. 2. Vertical distribution of air temperature along the north-facing slope of Mount Kreuzeck on the basis of four months representative of the four seasons.

3.3. Throughfall and open field deposition

Nitrogen throughfall and open field bulk deposition are shown in Figs. 6 and 7. Along the transect, the annual under-crown deposition of nitrate and ammonium shows great differences mainly due to meteorological circumstances. The fact that we installed a relatively low number of throughfall bulk samplers (six) in the forest sites may be of minor importance (Fenn et al., 2009).

A throughfall deposition rate of approximately 12 kg ha^{-1} a^{-1} is monitored in the Loisach valley (KRE070NB and KRA070SB). Due to the remoteness of the site, the nitrogen deposition is lowest at the Partnach valley plot KRE100SB, where only 6 kg ha^{-1} a^{-1} was deposited into the forest. The highest deposition rate of 18–21 kg ha^{-1} a^{-1} could be found at the steep north-facing slope KRE160NB near the crest of Mount Kreuzeck. Similar high deposition rates can be usually found in the vicinity of sources. For instance, throughfall deposition measurements perpendicular to a highway south of Munich resulted in 20.8 kg ha^{-1} a^{-1} at the forest edge near the road verge (50 m) and 12.5 ha^{-1} a^{-1} at a distance of 520 m from the road (Kirchner et al., 2005). Forests on steep and wind-exposed terrains may be characterized by similar deposition characteristics as forest edges which are particularly prone to filter out humidity and air pollution (Meyers et al., 1991; Kirchner et al., 2005).

Along the Garmisch-Partenkirchen transect, nitrate was deposited mainly in the winter half-year with a winter-to-summer deposition ratio of 1.1 (KRE0160NB)–1.6 (KRE130NB). The range of winter-to-summer deposition ratio of ammonium was 0.4 (KRE130SB)–0.9 (KRA070SB); ammonium deposition takes place predominantly during the summer half-year. The four-weekly NH_4–N and NO_3–N throughfall deposition rates are given in Figs. 8 and 9.

In the Southern Alps during 1994–1999, annual nitrogen loads were 13.6 and 13.1 kg ha^{-1} a^{-1} in the bulk input, and 15.0 and 18.0 kg ha^{-1} a^{-1} in throughfall deposition at Val Masino and Val Gerola (Lombardy/Italy), respectively (Balestrini and Tagliaferri, 2001). However, long-term monitoring of atmospheric deposition chemistry in Northern Italy showed a slight decrease of NO_3–N and a more widespread decline of NH_4–N in the subsequent years. Similar trends could be observed in other parts of the Alps; a significant decrease could be detected for ammonium only at Austrian sampling sites (Rogora et al., 2006).

The relation between NH_4–N and NO_3–N is quite balanced (0.9–1.1) with the exception of KRE160NB, where the ratio amounts to 1.3–1.5. Sulfur throughfall deposition ranged from 1.3 to 3.0 kg ha^{-1} a^{-1}

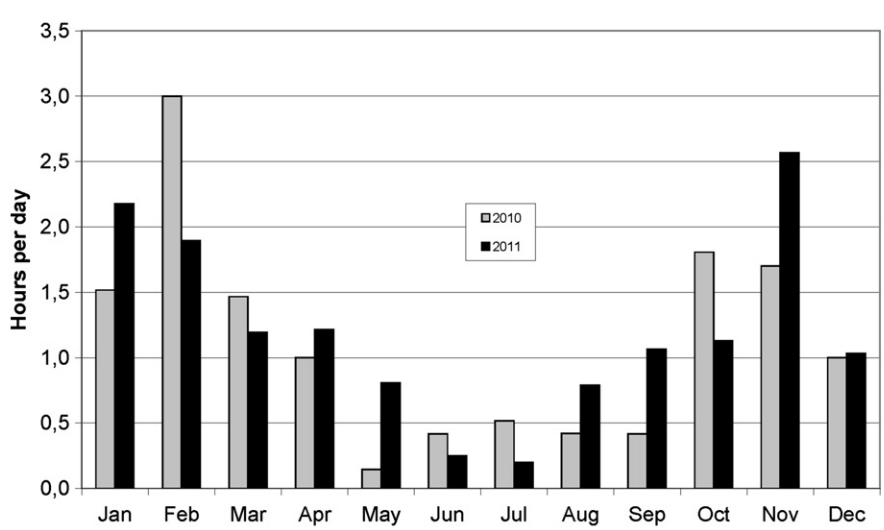

Fig. 3. Number of hours per month with temperature decrease along the south-facing slope of Mount Kramer (KRA070NF, KRA100NF) and contemporary temperature increase along the north-facing slope of Mount Kreuzeck (KRE070NF, KRE100NF) during sunny days in 2010 and 2011.

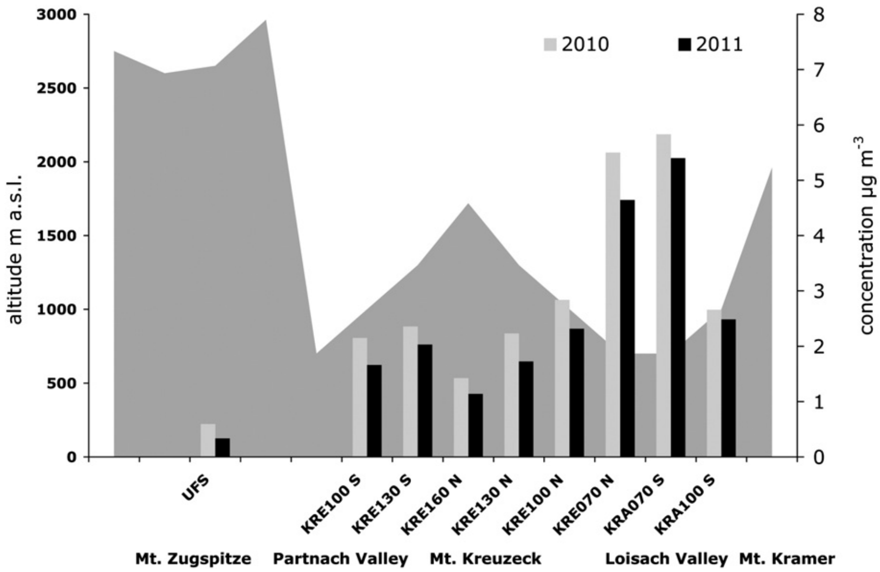

Fig. 4. Annual means of NO_2 air concentration in 2010 and 2011.

between 1000 and 1600 m a.s.l. in 2010, while deposition rates of 2.2 and 1.6 kg ha^{-1} a^{-1}, respectively, could be observed in 2011.

Open field deposition is mostly lower than throughfall deposition and characterized by less spatial variability. The range of open field bulk deposition is 6–8 kg ha^{-1} a^{-1} with the exception of the deposition monitored at KRE070NF in 2010; that year, NH_4–N input was twice as high as during 2011 and at the rest of the sites. Due to the high precipitation amount, the highest deposition rates were measured at UFS. However, our results confirm the findings of Lovett et al. (1999) who did not experience pronounced vertical trends in bulk deposition. Because of the relatively short distance of the study area to major urban sites (80 km) and agricultural areas (40 km) open field deposition of nitrogen in the Northern Alps is markedly higher than in Northern America, e.g. the Wyoming Mountains (Zeller et al., 2000). The mean ratios between NH_4–N and NO_3–N were 1.3 in 2010 and 1.5 in 2011. Similar to nitrogen input, deposition of SO_4^2 shows only small spatial variability (2010: 1.9–2.1 kg ha^{-1} a^{-1}; 2011: 1.3–1.6 kg ha^{-1} a^{-1}).

On one hand, it can be argued that the larger the difference between throughfall and open field deposition is, the more relevant the enrichment by dry and occult deposition within the forested sites becomes (Zimmermann and Wienhaus, 2004); this may be the case at the more elevated sites of our study area, particularly at KRE160NB. Deposition at high elevations can be markedly enhanced by fog and cloud precipitation and hoarfrost (Burns, 2003; McNeil et al., 2007). Sickles and Grimm (2003) stated for the total land area in the Eastern US beyond 600 m a.s.l. that clouds may account for 20–60% of the total wet ion deposition. A similar nearly exponential increase in ion deposition could be observed due to steep gradients in wind speed and cloud immersion frequency along an elevational transect at Whiteface Mountain, NY, U.S.A. (Miller et al., 1993). On the other hand, the corresponding ratios between NH_4–N and NO_3–N related to the throughfall deposition rates were rather balanced (0.9 in 2010 and 1.1 in 2011); this may be due to preferential uptake of ammonium N by the crowns.

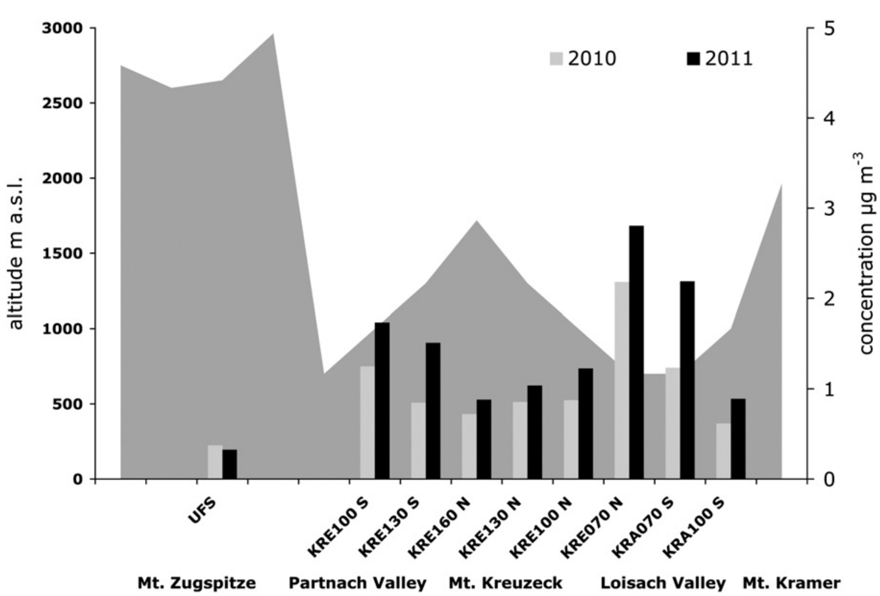

Fig. 5. Annual means of NH_3 air concentration in 2010 and 2011.

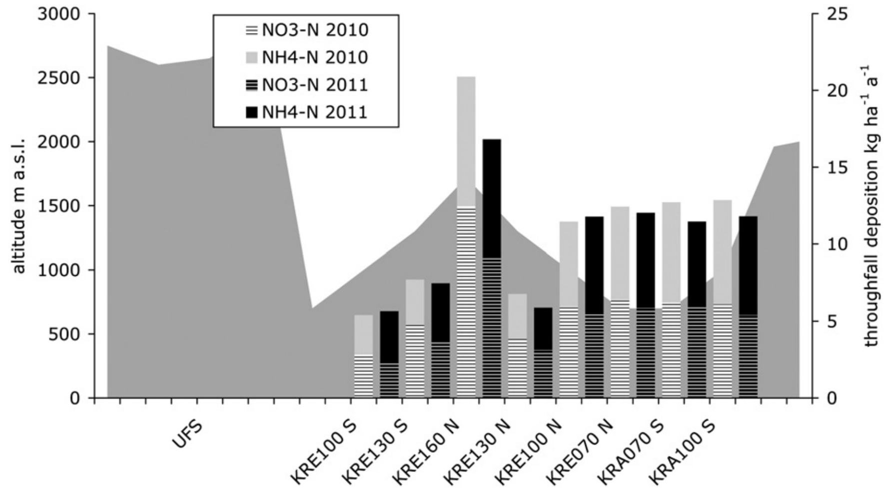

Fig. 6. NO_3–N and NH_4–N throughfall deposition in 2010 and 2011.

The assessment of the organic fraction in the N-deposition was not routinely foreseen in our monitoring study. As determined by Balestrini and Tagliaferri (2001) for Italian forest sites in the Alps, the contribution of the organic fraction amounted to 17% and 40%, respectively. These findings are congruent with the data from other studies (Carillo et al., 2002; Cornell, 2011; Zhang et al., 2012). Spangenberg and Kölling (2004) estimated for different spruce stands in Bavaria that the organic N-ratio in total throughfall is in the range of 13–18%; therefore, we assume a missing organic nitrogen contribution of about 15% for our study sites. Neglecting the possible uptake by the crowns, we suppose that the deposition of nitrogen into the forest sites may still be well above the estimated critical loads (CL: 10–20 N kg ha^{-1} a^{-1}) at some plots of our study area.

3.4. Comparison of the canopy budget and the inferential methods

The results based upon the two different methods for estimating the total deposition are depicted in Figs. 10 and 11. Both methods, the canopy budget approach and the adjusted inferential method, consider the total nitrogen input. Therefore, the resulting deposition is higher than the throughfall deposition which may be assumed to determine the lower limit of real deposition in forests.

According to the canopy balance method, the calculated total nitrogen deposition is in the range of 11–30 kg ha^{-1} a^{-1}, i.e. mostly higher on average by a factor 2.1 (with a range of 0.7–4.1) compared with the under-crown deposition. However, Meesenburg et al. (2009) stated for beech ecosystems that the estimation of total N deposition by the canopy budget model of Ulrich (1983) probably still underestimates the real input rates. In any case, an exceedance of critical loads is likely to occur at some parts of the sites of our study area.

By employing this method, the canopy uptake (CU) of nitrogen was estimated to average values of 7.5 and 7.7 kg ha^{-1} a^{-1} for 2010 and 2011, respectively. CU seems to be relatively high, speculatively due to the location of the forests stands in the calcareous Alps. The range of CU is high with a minimum of 3 kg ha^{-1} a^{-1} for KRE100SB, where throughfall deposition was low, and a maximum for KRE130SB (14 kg ha^{-1} a^{-1}), where under crown deposition was high. The marked CU differences between the two years, which are not well understood, consequently result in corresponding variations of the total deposition between 2010 and 2011. Estimations performed in the Fichtelgebirge (Northern Bavaria) by throughfall analysis indicate that uptake by trees from wet-deposited nitrogen as NH_4^+ and NO_3^- might be in the range of 1–10 kg ha^{-1} a^{-1}(Eilers et al., 1992). Sievering et al. (2000) found a canopy N uptake to be in the range of 1–5 kg ha^{-1} during the growing season for eastern US conifer sites. Schmitt et al. (2005)

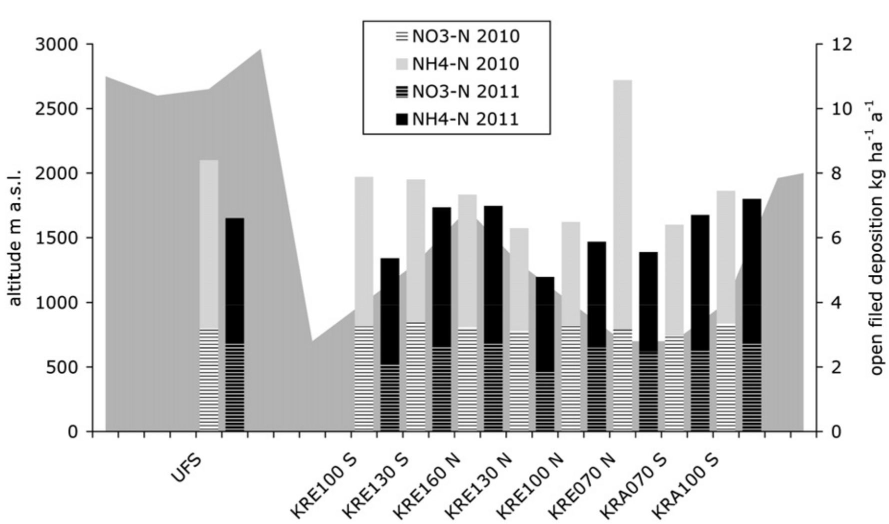

Fig. 7. NO_3–N and NH_4–N open field bulk deposition in 2010 and 2011.

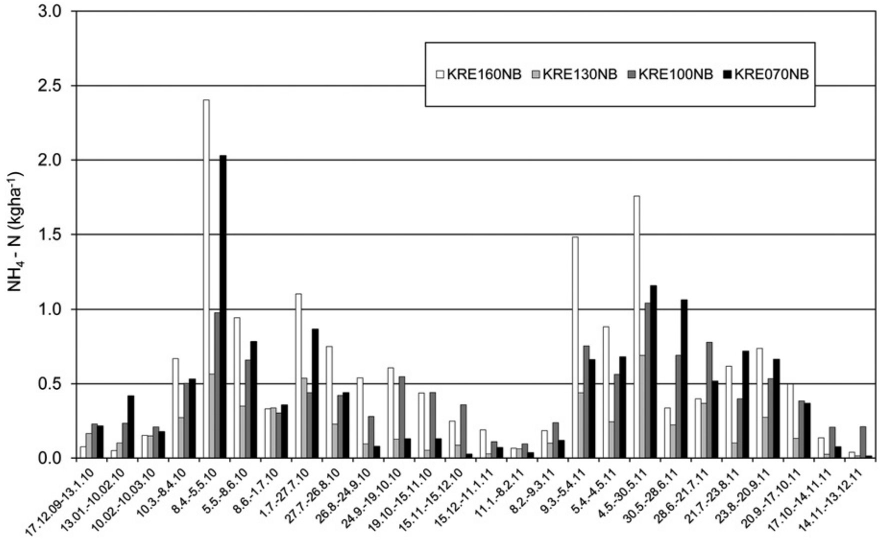

Fig. 8. Four-weekly sums of NH_4–N throughfall deposition along the Kreuzeck profile.

calculated a nitrogen canopy uptake of 1.3–5.5 kg ha^{-1} a^{-1} based upon measurements in ten forest stands in Switzerland. Sievering et al. (2007) calculated approximately 2.5 kg ha^{-1} a^{-1} during the growing season for a spruce forest in Colorado (USA). Canopy N uptake for spruce is markedly higher in the growing season than in the dormant season (Adriaenssens et al., 2012). In relation to the discrepancy between our CU values and the literature based N uptake, our results of total deposition must be treated with caution given the considerable uncertainty concerning the deposition monitoring, the model inputs, and the parameterization.

Despite the aim to select comparable forest stands in the study area, slight differences in stand characteristics could not be avoided. Taking into account the measurement of the meteorological parameters, uncertainties in rain and snow gauging at the elevated sites, the relatively small number of precipitation or bulk samplers, and too long bulk sampling intervals, despite the shortened exposure time in winter, are reasons for the discrepancies. These factors may have an impact on throughfall and total deposition.

Additionally, the canopy budget approach is based upon parameterizations and assumptions, which may lead to some uncertainties in the outputs (Meesenburg et al., 2009). This method separates fluxes of the base cations due to dry deposition from the fraction caused by the internal leaf leaching. Thus, these elements are assumed to have the same deposition velocity as Na^+. An uncertainty may be related to the practice, that the ion concentrations were set to be 0.5 × detection limit (DL), if concentrations were below DL.

The inferential method yielded total nitrogen inputs in the range of 12–24 kg ha^{-1} a^{-1} in 2010 and 12–29 kg ha^{-1} a^{-1} in 2011. Lowest N depositions were calculated for the sampling site in the remote Partnach valley (KRE100FB). Total deposition was estimated to be highest in the rather urbanized Loisach valley and some elevated sampling site. Differences of the total deposition between 2010 and 2011 are evident; KRE130SB and KRA070SB exhibited the largest discrepancies between the two years. Therefore, the interpretability is reduced.

Although the inferential method for estimating dry deposition in forest stands needs gas concentration data within the canopy, we measured the gases at 2 m above the ground in small forest clearings. Therefore, we assume that the real concentrations were underestimated to some extent. This was shown by Gadsdon and Power (2009) for a

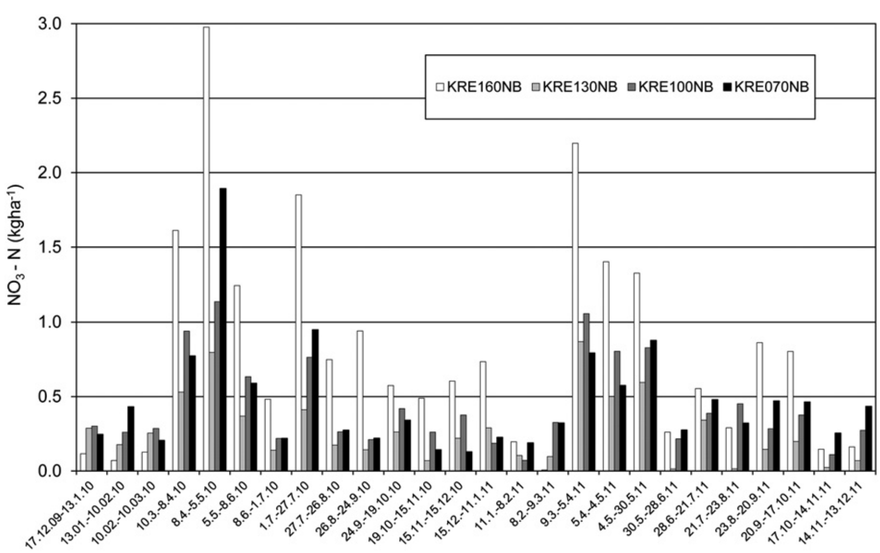

Fig. 9. Four-weekly sums of NO_3–N throughfall deposition along the Kreuzeck profile.

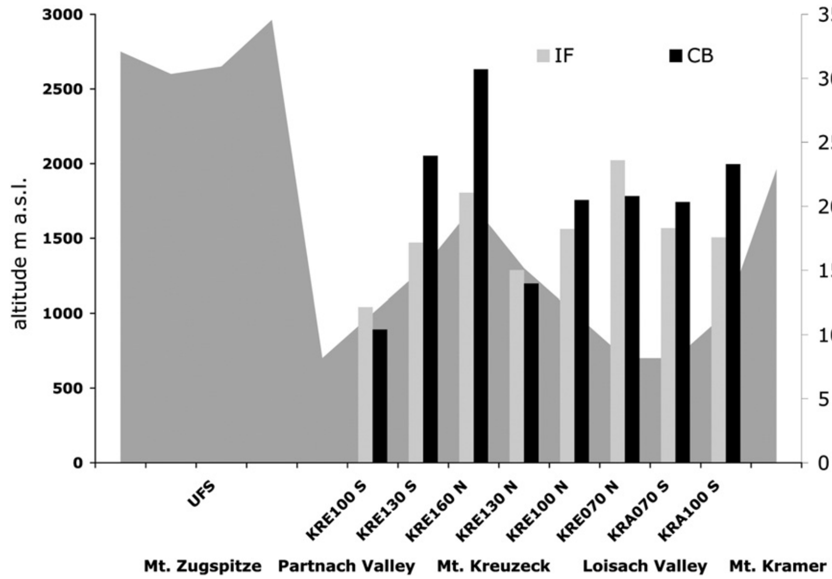

Fig. 10. Total deposition estimated with canopy budget TD_{CB} and inferential method TD_{IF} for 2010 ($r^2 = 0.52^*$; $p < 0.05$).

forest in South England, where within-canopy concentrations of NO_2 and NH_3 were up to 25% and 140% higher, respectively, than at ground level. Additionally, passive samplers measure only average concentrations and obscure diurnal and other temporal trends (Fenn et al., 2009); thus, strongly varying NO_2 and NH_3 concentrations within the sampling periods could not be taken into account.

As the assumption of an invariant v_d leads to unrealistic results, we introduced a semi-empirical parameterization based on slope steepness and interacting meteorological parameters such as wind exposure and inversion frequency. The parameterization of literature-based fixed deposition velocities is based upon the assumption of horizontal wind velocity and upslope wind effects which can only be roughly estimated; in our study, sophisticated meteorological measurements at eight forested sites and at eight adjacent open field plots could not be performed. The assumption of correction factors, which are mostly independent of the weather conditions, day–time, and seasons, may not satisfactorily reflect the reality. Wet deposition was estimated from the bulk deposition rate. The assumption of a constant ratio between both rates entails further implications.

Nevertheless, the results obtained by the two methods are rather comparable at most of the sites. Remaining discrepancies can be found particularly if results from KRE160NB are compared. The coefficients of determination r^2 between the total annual deposition rates estimated on the basis of the two different models are relatively low (0.52 in 2010 and 0.62 in 2011), but statistically significant ($p < 0.05$). Due to the numerous uncertainties, the discrepancies are not surprising.

Similar discrepancies between the two methods, performed with a partly different parameterization, have been found by Schmitt et al. (2005) for Swiss long-term forest sites. Several additional factors accounting for these differences have been identified. The uncertainties about the role of the occult deposition and the assumption of an annually averaged deposition velocity have been recognized by Schmitt et al. (2005) as potential sources of errors. In contrast to the parameterization of v_d performed by Schmitt et al. (2005), our assumptions did not

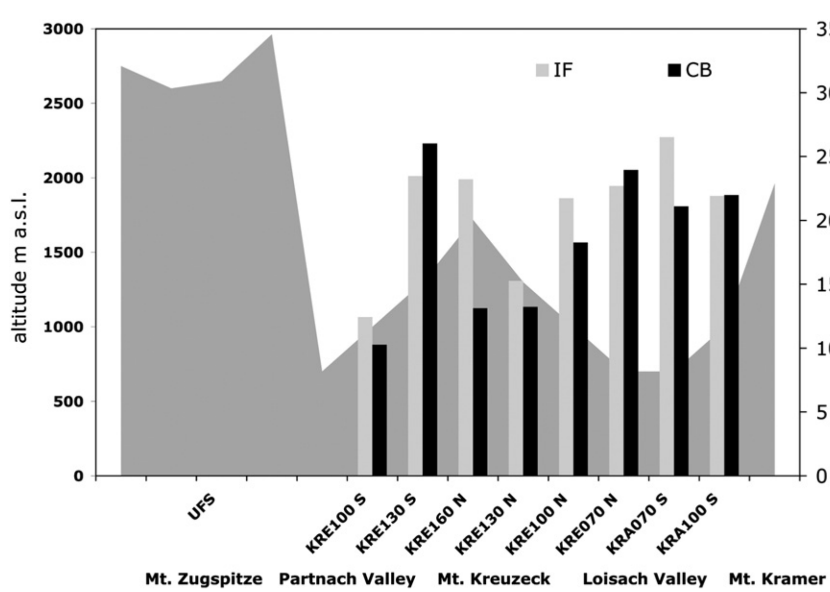

Fig. 11. Total deposition estimated with canopy budget TD_{CB} and inferential method TD_{IF} for 2011 ($r^2 = 0.64^*$; $p < 0.05$).

consider the fact that the duration of winter periods at slopes may have an impact upon the deposition velocity; the observation that throughfall nitrogen deposition was highest at KRE160NB led to the impression that wind exposure may dominate the effect of lower deposition velocities in winter (Schmitt et al., 2005).

4. Conclusion

In the framework of a two-year monitoring study performed in the Alps near Garmisch-Partenkirchen/Bavaria, we measured different components of nitrogen deposition in forests sites. Passive sampler measurements of NO_2 and NH_3 showed that most sites along the transect are scarcely affected by local emissions from traffic and agricultural activities in the valley. Nevertheless, our throughfall measurements indicate that high-elevated spruce stands receive a marked deposition rate of nitrate and ammonium up to 21 kg ha^{-1} a^{-1} due to long range transport. The high atmospheric input of nitrogen is caused by mechanisms, which are not well recorded by open field bulk sampler measurements. Apparently, deposition of chemical compounds in forests is severely influenced by the topography, as altitude or slope steepness, and wind as important factors determining both precipitation and deposition chemistry. We assume that the extra input may be caused by cloud droplets combined with high wind velocities. However, our data suggest a nonlinear pattern of throughfall deposition of nitrogen versus elevation.

By the calculation of the total nitrogen deposition, the possible uptake by the trees has been estimated. Our calculations, which based upon the canopy budget model, yielded total inputs of up to 30 kg N ha^{-1} a^{-1} particularly at the 1600 m a.s.l. site near the crest and 12 kg N ha^{-1} a^{-1} in lee side valleys. Alternatively, we estimated the total deposition which can be separated into the sums of wet and dry deposition, using the inferential method. This method requires a comprehensive data set and is based on concentrations of gases and aerosols and their corresponding deposition velocities. Because they can be hardly measured for different sites, plant stands, and meteorological situations, we used mean concentrations of NH_3, NO_2, and aerosols, measured by ventilated battery-powered samplers and literature-based deposition velocities. Based partly upon simple semi-empirical factors regarding slope inclination and wind exposure, a parameterization of the mean deposition velocity for each site was made. Both the canopy budget and the inferential methods yielded satisfying results; however considerable discrepancies remained for some sites.

Therefore, throughfall deposition characterizes best the minimum input of nitrogen and other elements in forests; under-crown deposition monitoring is the most reliable method for quantifying the deposition in forests and still has the most advantages when differences between differently exposed sites are investigated. If models should be adopted to quantify the total deposition in forests, different concepts of parameterization are conceivable to consider the dependency of the deposition velocity on different surface conditions and meteorological parameters. Major attention should be directed towards the effects of canopy wetting. In future research programs, it should be investigated how the predicted climate change with enhanced temperatures throughout the year and the increased precipitation in winter will affect the nitrogen deposition in the Alps. Furthermore, studies at mountain sites above the timberline would be of major importance in order to investigate the impact of nitrogen on grass and alpine vegetation.

Acknowledgments

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