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Total number of authors:

11

Published in:

Environmental Pollution

Link to article, DOI:

[10.1016/j.envpol.2012.12.006](https://doi.org/10.1016/j.envpol.2012.12.006)

Publication date:

2013

Document Version

Publisher's PDF, also known as Version of record

[Link back to DTU Orbit](#)

Citation (APA):

Karlsson, P. E., Ferm, M., Pihl Karlsson, G., Hellsten, S., Tømmervik, H., Hole, L. R., Akselsson, C., Ruoho-Airola, T., Aas, W., Mikkelsen, T. N., & Nihlgård, B. (2013). Biomass burning in eastern Europe during spring 2006 caused high deposition of ammonium in northern Fennoscandia. *Environmental Pollution*, 176, 71-79. <https://doi.org/10.1016/j.envpol.2012.12.006>

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Biomass burning in eastern Europe during spring 2006 caused high deposition of ammonium in northern Fennoscandia

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ARTICLE INFO

Article history:

Received 4 July 2012

Received in revised form

12 December 2012

Accepted 14 December 2012

Keywords:

Ammonium deposition

Biomass burning

Vegetation damage

Northern latitudes

Eastern Europe

ABSTRACT

High air concentrations of ammonium were detected at low and high altitude sites in Sweden, Finland and Norway during the spring 2006, coinciding with polluted air from biomass burning in eastern Europe passing over central and northern Fennoscandia. Unusually high values for throughfall deposition of ammonium were detected at one low altitude site and several high altitude sites in north Sweden. The occurrence of the high ammonium in throughfall differed between the summer months 2006, most likely related to the timing of precipitation events. The ammonia dry deposition may have contributed to unusual visible injuries on the tree vegetation in northern Fennoscandia that occurred during 2006, in combination with high ozone concentrations. It is concluded that long-range transport of ammonium from large-scale biomass burning may contribute substantially to the nitrogen load at northern latitudes.

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1. Introduction

Nitrogen deposition may increase in high latitude regions due to climate change (Hole and Enghardt, 2008) as well as due to increased shipping activities in the Arctic (Corbett et al., 2010). Furthermore, the Arctic and sub-Arctic ecosystems are experiencing considerable changes in the climate, both during summer and winter (Bokhorst et al., 2009; Karlsen et al., 2009). Nitrogen deposition has been shown to influence forest, heathland and bog ecosystems even on remote parts of Europe (Bobbink and Hettelingh, 2011; Dise et al., 2001; Phoenix et al., 2012). Northern ecosystems are especially vulnerable to nitrogen deposition, as bryophytes and lichens are important parts of these ecosystems and as bryophytes and lichens take the major part of their nutrition directly from the air (Cornelissen et al., 2001; Gordon et al.,

2001; Bates, 2002). Furthermore, nitrogen may increase primary productivity of the ecosystems and above a certain level it is common that a few competitive species are able to exploit the available N and to dominate other species by shading and lead to changed biodiversity (Dise et al., 2001; Stevens et al., 2011; Phoenix et al., 2012). An effect threshold for annual nitrogen deposition to arctic and alpine ecosystems has been suggested to be between 5 and 15 kg N/ha (Bobbink et al., 2010; Bobbink and Hettelingh, 2011).

There has been an increase in NO_x and NH_y deposition at northern latitudes since 1860 (Galloway et al., 2004; Hole et al., 2009; Vestreng et al., 2009; Ruoho-Airola et al., 2012) as a result of industrialization and intensified farming. After the 1990s there has been a decline in the observed concentrations of nitrogen components in air and precipitation over Europe as a result of emission reduction measures such as the UN-ECE Gothenburg protocol (Tørseth et al., 2012). Future increased precipitation may however again increase deposition of sulphur and nitrogen compounds in northern regions (Hole and Enghardt, 2008). The current

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deposition of NO_x and NH_3 in northern Norway and northern Sweden has been estimated to below 2 kg N/ha (Aas et al., 2008; Pihl Karlsson et al., 2011).

The alkaline NH_3 reacts readily with acidic substances in the atmosphere to form ammonium salt particles, such as $(\text{NH}_4)_2\text{SO}_4$, NH_4NO_3 and NH_4Cl , that occur predominantly in the fine particle fraction (Hertel et al., 2011). $(\text{NH}_4)_2\text{SO}_4$ can under certain meteorological conditions be transported over long distances (Brosset et al., 1975; Irwin and Williams, 1988; Krupa, 2003). In the absence of sulphate, NH_3 will mainly react with HNO_3 and form NH_4NO_3 . NO_3 containing particles in general have higher deposition velocities as compared to SO_4 containing particles (Erisman et al., 1997).

Emissions from the large-scale biomass burning represent an important component of the hemispheric air pollution (Yurganov et al., 2004; Simmonds et al., 2005). There are however not many studies of linking pollution of reduced nitrogen with biomass burning. Agricultural activities, including livestock productivities and fertilizing, are by far the most important source of ammonia, but biomass burning may also be a significant source (Krupa, 2003; Lamarque et al., 2010; Hecobian et al., 2011). During the last decade there have been several large-scale biomass burning events in eastern Europe. Over the past decade, improved remote sensing has permitted a more accurate assessment of the annual fires in Russian boreal forest, revealing that Russia in general has the largest area burned among boreal forests (Goldammer, 2010). Comparative fire statistics for total vegetated area and forest area burned in the Russian Federation in the period 2000–2007 are presented in Fig. 1, based on agency reports and remote sensing. The number of forest fires in Russia has increased from an annual average of 17 800 fires during the period 1986–1990 to 21 700, 30 000 and 29 300 fires for during the periods 1991–1995, 1996–2000 and 2001–2005, respectively (Goldammer, 2010). For the year 2006, Goldammer (2010) reports 36 000, 1306 and 3800 fires in Russia, Belarus and Ukraine, respectively. Increased fire activity due to climate change is anticipated in the circumboreal forest (Flannigan et al., 2009). There are large inter annual variability of the magnitude and importance of biomass burning (van der Werf et al., 2006), for Russia this is illustrated in Fig. 1 where 2003 and 2006 seem to be important years.

Transport of air pollution from biomass burning depends on the weather situation during the period of burning as well as the plume height of the fire, i.e. the amount of heat in the system. In the beginning of May 2006, there was an unusual episode with snow at Svalbard coloured black (Stohl et al., 2007). There were also record high concentrations of ozone and most other pollutants detected on Svalbard, as well as on Iceland. This highly polluted air originated from biomass burning in eastern Europe and it

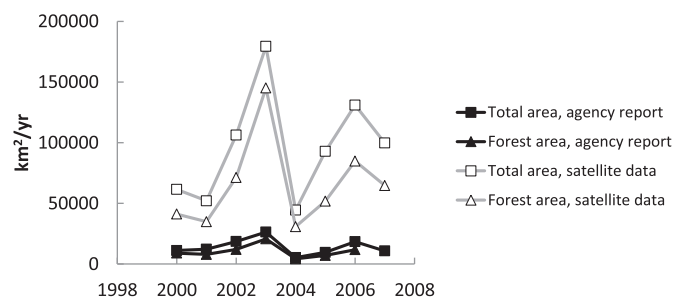


Fig. 1. Comparative fire statistics for total vegetated area and forest area burned in the Russian Federation during the period 2000–2007, based on agency reports and remote sensing. Source: Goldammer, 2010.

was estimated that approximately 2 M ha burnt during May and June 2006 (Stohl et al., 2007). An anti-cyclonic weather system over Russia transported this polluted air towards west and north, ending up from Scotland to Finland (Anttila et al., 2008; Whitham and Manning, 2007). Significant concentrations of several pollutants, among them ammonium, were detected in the polluted snow on Svalbard (Stohl et al., 2007). High amounts of ammonium were also detected in the PM_{10} particles sampled in Virolahti, south-eastern Finland close to the border of Russia, during May 2006 (Anttila et al., 2008).

Trajectory analysis (Fig. 2) showed that some of these distinct, polluted air masses were transported across central and northern Fennoscandia, in particular the county of Jämtland in central Sweden. In connection with these pollution episodes during 2006, some unusual vegetation damage was detected in northern Fennoscandia (Manninen et al., 2009).

The aims of this study were to analyse occurrence of high amounts of ammonia/ammonium in the air and deposition at northern latitudes in Fennoscandia, to examine links to large scale biomass burning events and to analyse the possible role for the occurrence of visible damage on the vegetation.

2. Methods

2.1. Sites

In this study, results were used from sites within different monitoring networks in Finland, Sweden and Norway (Fig. 3). Air concentration values were used from the EMEP (European Monitoring and Evaluation Program, www.emep.int) sites Virolahti and Utö in Finland, Breckälven and Esrange, in Sweden and Tustervatn in Norway. Trajectory analysis was also used for the Norwegian EMEP site Karasjok. In addition, air concentrations were measured at high altitudes in the Swedish mountain region in a special project. These sites were Katterjäck, Tjärnberg, Skorvfjället and Prästbodarna. Data for throughfall- and bulk-deposition was used from several sites in Sweden operated by the Swedish Throughfall Monitoring Network (SWETHRO, Pihl Karlsson et al., 2011), both at low (Nymyran, Sör-Digertjärn, Storulvsjön) and high (Branten, Sänfjället, Hundshögen, Fiskåfjället) altitudes. Information on bulk- and throughfall-deposition data was also used from the Norwegian EMEP site Tustervatn.

2.2. Air concentration sampling and analysis

The air concentration samples were measured at all sites in Finland, Sweden and Norway according to the EMEP Manual (EMEP, 1996). Measurements in Finland have been described in detail by Karlsson et al. (2007). At the high altitude sites in Sweden, sampling was made with the same air flow rate as the EMEP sites (15 l/min) intermittently 5 min every 30 min. The amount of black smoke collected on the first filter at the Swedish site Breckälven was determined using an optical, reflectance method (ISO 9835:1993). The authors recognize that “black smoke” is a poorly defined group of substances and that there are large uncertainties in the quantification. However, in this article the data are mainly used as a tracer for large air pollution episodes.

2.3. Deposition sampling and analysis

Samples from the bulk- and throughfall-deposition measurements within the SWETHRO network in Sweden were collected and analysed as described by Pihl Karlsson et al. (2011). The throughfall measurements at Swedish high altitude sites Katterjäck (515 m a.s.l.), Tjärnberg (500 m a.s.l.), Skorvfjället (808 m a.s.l.) and Prästbodarna (680 m a.s.l.), were similar to those described above for the SWETHRO network, with some differences. The density of trees is low at the elevational tree line. A random positioning of throughfall collectors would therefore mainly result in precipitation collection. Therefore the five throughfall collectors were positioned directly under single Norway spruce trees. Even though the absolute values for throughfall might be somewhat uncertain due to this arrangement, the relative values between years should be reliable.

2.4. Meteorological information

Meteorological information for the different sampling sites in Sweden was provided by the weather service (“LuftWeb”) provided by the Swedish Meteorological and Hydrological Institute (SMHI). Estimates can be obtained for each position in Sweden based on interpolated weather data.

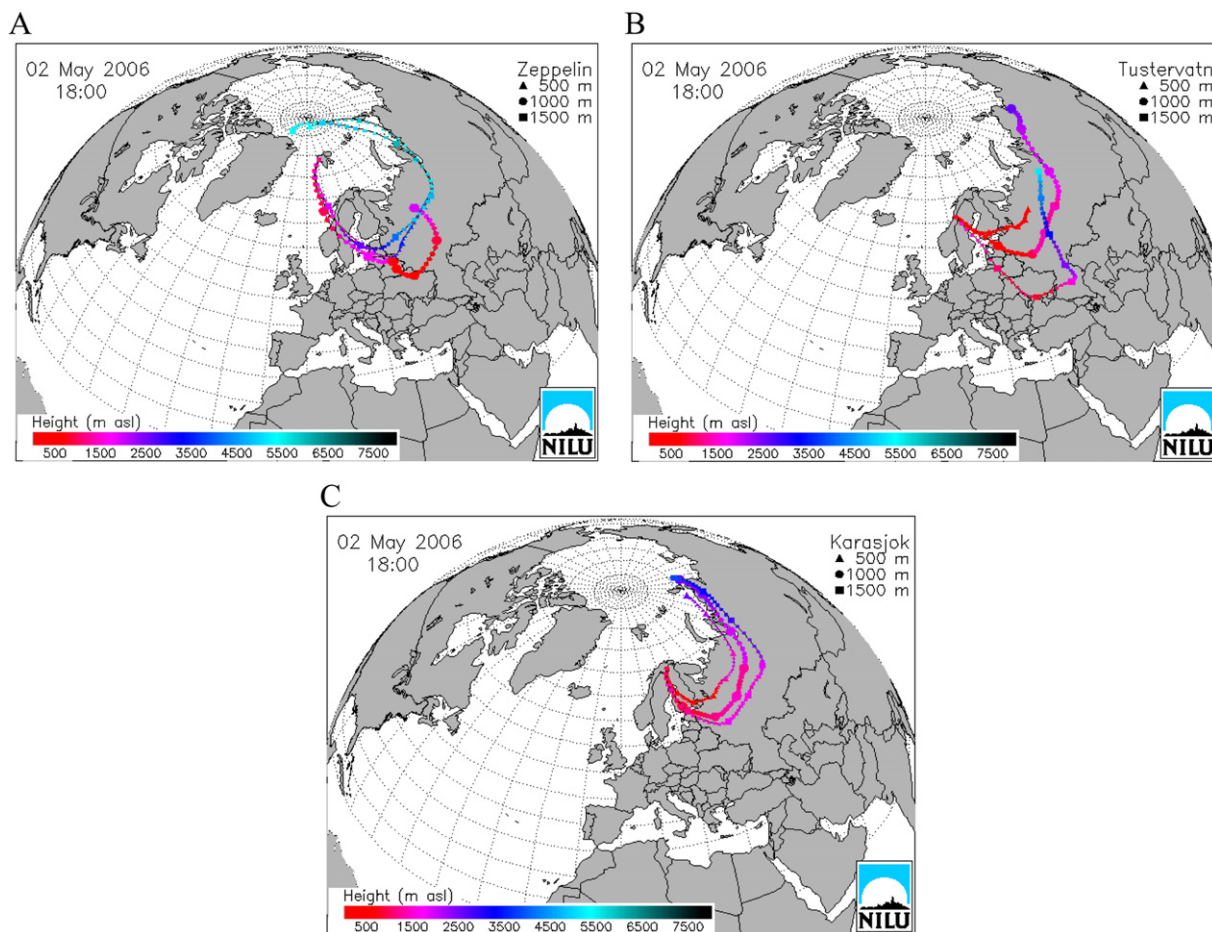


Fig. 2. Trajectory analysis for air masses arriving in the afternoon on May 2nd 2006 at three different sites; Zeppelin on Svalbard (A), Tustervatn in central Norway (B) and Karasjok in northern Norway (C). Trajectories calculated using the Flextra model were derived from NILU (<http://www.nilu.no/projects/ccc/trajectories/>). Triangles show trajectories for air arriving at the destination at between 0 and 500 m above ground, circles 500–1000 m above ground and squares at 1000–1500 m above ground. The colour shows height above ground during air mass transport. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

3. Results and discussion

3.1. Air concentrations of pollutants over Finland, Sweden and Norway

Concentrations of black smoke can be used as an indicator for polluted air originating from biomass burning (Hyvärinen et al., 2011). Daily mean air concentrations of black smoke during 2006 are shown in Fig. 4 for the EMEP monitoring site Bredkälén, positioned in the county of Jämtland, Sweden. There was a clear event around the beginning of May, where the concentrations of black smoke started to rise above the baseline level around April 26 and maximum daily mean concentration, $12 \mu\text{g}/\text{m}^3$, was measured on May 2. The concentrations returned to baseline levels around May 13.

The daily mean air concentrations of $\text{NH}_3 + \text{NH}_4^+$ (gaseous + particle, hereafter referred to as total NH_x , however for Tustervatn only particle concentrations of NH_4^+ were included, since there is a well-documented problem with local emissions of NH_3 at this site) and SO_4^{2-} during 2006 are shown in Fig. 5 for Utö, Bredkälén and Tustervatn. All three sites are positioned so that the polluted air masses from eastern Europe during the beginning of May (Fig. 2) passed over. High air concentrations of NH_x and SO_4^{2-} were detected at all three sites during the period 25 April–13 May 2006. However, there were high concentrations measured also

during other parts of 2006. At Utö, the highest daily air concentration of total NH_x for the year 2006 was measured on May 3rd. The high concentrations of total NH_x at Utö were accompanied by relatively high concentrations of SO_4^{2-} . Also at Bredkälén, the highest daily air concentration of total NH_x for the year 2006 was measured on May 3rd, and also here this was accompanied by high concentrations of SO_4^{2-} . Finally, at Tustervatn there was a distinct episode for particulate NH_4^+ and SO_4^{2-} , as well as for other pollutants measured at the site in the period from end of April to 10 May 2006, with the highest peak at 2nd May. The ozone concentrations during this period were among the highest ever observed at Tustervatn. What makes the episode special was the record high concentration of potassium, $0.27 \mu\text{g}/\text{m}^3$, measured during this period (data not shown), which confirms that the source of air pollution was from biomass burning (Chaiyo et al., 2011).

At the high altitude sites in northern Sweden, air concentrations of pollutants were measured on a monthly basis (Fig. 6). Even though the air concentrations for May 2006 were not always record high, there were high values also in particular during 2003, the values for May 2006 were always among the highest during the period of measurements. Also high sulphur, potassium and calcium concentrations were detected. The concentrations of calcium were record high at all sites during May 2006. High concentrations of calcium are also a typical component of particulate emissions from forest fires (Chaiyo et al., 2011).



Fig. 3. Map showing the positions of the different monitoring sites.

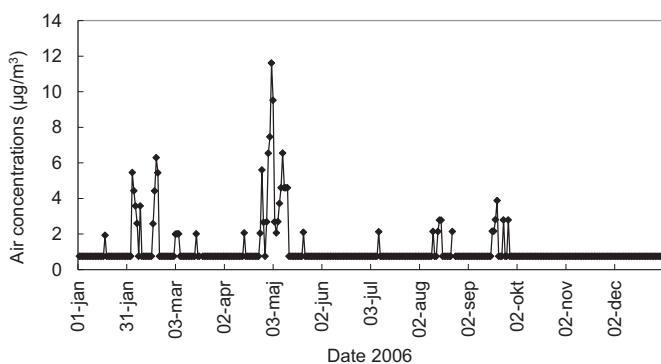


Fig. 4. Daily mean air concentrations of black smoke during 2006 for the EMEP monitoring site Breckålen, positioned in the middle of the county of Jämtland, Sweden.

During the period April–May 2006, there were also unusually high ozone concentrations detected at the monitoring sites Tustervatn and Esrange (Fig. 7). There were several episode days with hourly ozone concentrations ranging 75–81 ppb between April 26th, and May 10th. There was, however, also one more ozone episode at these sites later during 2006, with ozone concentrations at Tustervatn on June 12th up to 87 ppb. There were also high ozone concentrations on July 7th. The ozone concentrations were not so high at Breckålen during these periods, with a maximum of 55 ppb (data not shown). Thus, the polluted air masses passing over northern Fennoscandia in the spring and summer of 2006 also contained volatile organic compounds and nitrogen oxides leading to a high ozone formation under the prevailing sunny weather conditions during this time.

Overall, the results may be interpreted as that there were several events with polluted air masses passing over central and

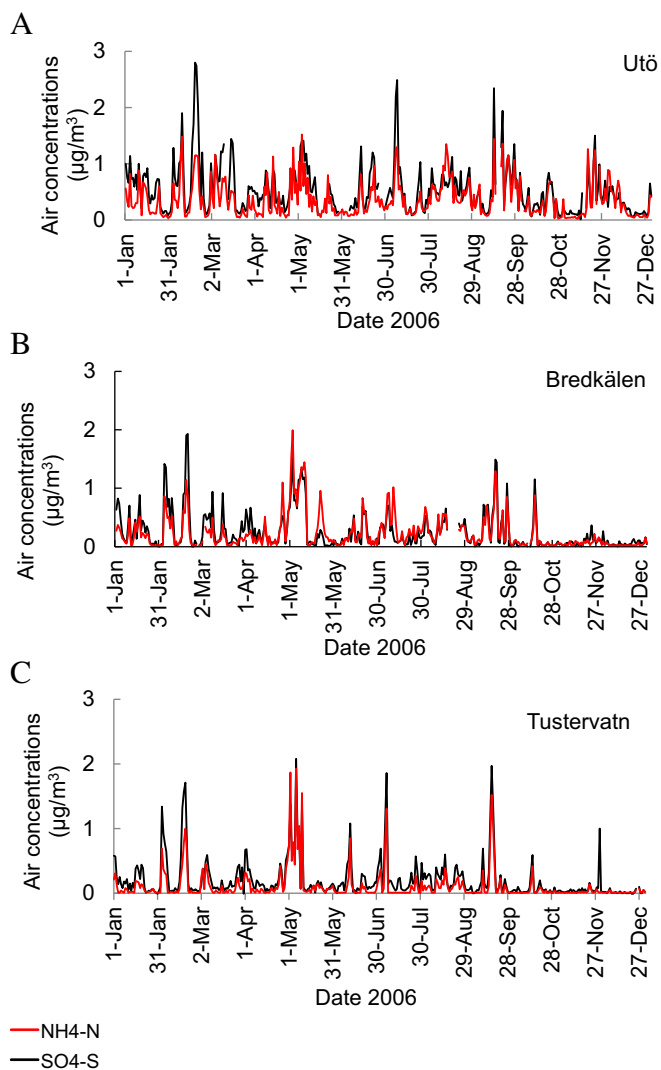


Fig. 5. Daily mean air concentrations of $\text{NH}_3 + \text{NH}_4$ (gaseous + particles, at Tustervatn only particles) and SO_4^{2-} during 2006 at three different sites, Utö in Finland (A), Bredkälen in Sweden (B) and Tustervatn in Norway (C).

northern Fennoscandia at high altitudes during the spring and summer 2006, moving in slightly different directions. One well-documented event in particular, during the end of April and beginning of May, resulted in high air concentrations of NH_3/NH_4 detected also at low altitude monitoring sites along the trajectory pathway. That the polluted air masses in May originated from biomass burning in eastern Europe was confirmed by trajectory analysis (Fig. 2), but also by measurements of air concentrations of levoglucosan at Helsinki and Svalbard (Stohl et al., 2007; Saarikoski et al., 2008). However, the air plume was transported over other source areas and it is possible that some of the enhanced reduced nitrogen also originated from other sources that the plume took up on its way.

3.2. Deposition

An unusually high monthly value for ammonium deposition in throughfall was detected at Nymyran in June 2006, a site with a 75-year-old Norway spruce forest at an altitude of 300 m a.s.l in the county of Jämtland, Sweden (Fig. 8). The sample volume was relatively normal but the NH_4^+ concentration in the sample was high. The sample was carefully checked for contamination, but there was

no sign of bird droppings such as e.g. high pH or high phosphorous concentrations. Analysis of Kjeldahl-N showed that the sample content of organic N was relatively low.

Throughfall measurements at two nearby Swedish sites, Sör-Digertjärn under Scots pine and Storulvsjön under Norway spruce, and at the nearby Norwegian site Tustervatn under Norway spruce (Fig. 3), did not show elevated NH_4^+ or Kjeldahl-N deposition during 2006 (data not shown).

The sampling of throughfall deposition is, of course, heavily dependent on precipitation amounts. Therefore we did a careful analysis of the daily precipitation events at the site Nymyran (Fig. 9). The analysis was based on interpolated data on daily precipitation for the exact position of the Nymyran site, provided by the Swedish Meteorological and Hydrological Institute (SMHI). There were only small amounts of precipitation during the period between the pollution event on 2–3 May 2006 and the end of the May sampling period for throughfall at Nymyran (Fig. 9). Between May 1st and the turn of the sampling period on May 22nd 2006, the accumulated precipitation was only 15 mm and the daily maximum precipitation 5 mm. In contrast, during the nine days between 22nd May and 31st May 2006, the accumulated precipitation was 49 mm. Hence, this analysis shows that the high amounts of NH_4^+ found in the June 2006 throughfall sample from Nymyran might well have included dry deposition from the highly polluted air masses that passed over the county of Jämtland in the beginning of May 2006. There was little precipitation during the remaining part of the May sampling period but substantial precipitation to wash out the NH_4^+ to the throughfall samplers during the June sampling period.

Monthly ammonium deposition in throughfall is shown in Fig. 10 for three sites at high altitude (600–800 m a.s.l) in the county of Jämtland. All three sites show high monthly ammonia deposition during summer 2006, but at different months, also as compared to Nymyran. The relative ammonia deposition peak value for Hundshögen was not as high compared to the other sites. No Kjeldahl-nitrogen analyses are available for these sites. Elevated NH_4^+ deposition in throughfall at high altitudes was also evident at four other sites outside the county of Jämtland (data not shown), one site south of Jämtland (Branten) and three sites at different distances north of Jämtland (Skorvfjället, Tjärnberg and Katterjåkk, Fig. 3). At these sites the NH_4^+ peak during 2006 occurred in June or July. However, the peaks at these sites were not as pronounced during 2006, as compared to peaks during other years. Similar to what was described for Nymyran, the appearance of NH_4^+ in the throughfall samples at different months at different sites most likely depended on the precipitation pattern at the respective site (data not shown).

There were no measurements of bulk deposition at the site Nymyran during 2006. However, there were bulk deposition measurements available from the nearby site Storulvsjön (Fig. 3). There were no pronounced peaks in the monthly, bulk NH_4^+ deposition during 2006 at Storulvsjön (data not shown).

To summarize, several monthly pronounced NH_4^+ throughfall deposition events were detected during the summer of 2006, both at low and high altitudes, in particular in the county of Jämtland, but also in other parts of northern Sweden. These events occurred during different months, probably related to the timing of precipitation events. It is likely that the deposition to the forests occurred as dry deposition and then was washed to the throughfall sampling collectors when there was enough precipitation. The dry deposition at Nymyran most likely occurred during the pollution episode in the beginning of May 2006, originating from biomass burning in eastern Europe, but for the other sites the deposition might be related to other pollution episodes during the summer 2006.

It is difficult to quantify the NH_4^+ deposition to the forests in northern Sweden, since a large fraction of the nitrogen deposition

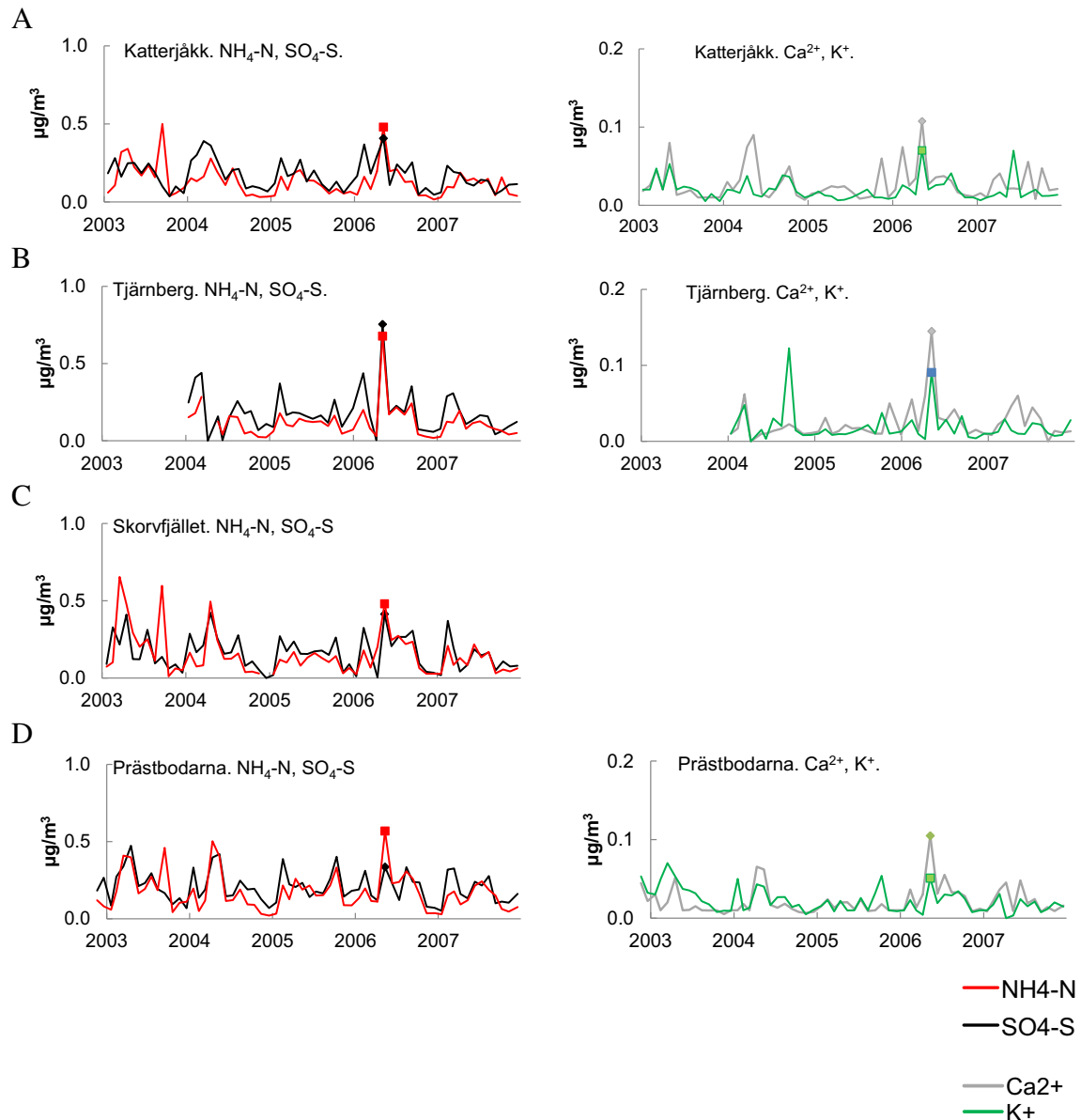


Fig. 6. Air concentrations measured at high altitudes in northern Sweden, 2003–2007. Monthly mean air concentrations of $\text{NH}_3/\text{NH}_4^+$ (gaseous + particle), SO_4^{2-} , K^+ and Ca^{2+} . Nitrogen compounds are expressed as $\mu\text{gN}/\text{m}^3$. A, Katterjåkk, 515 m a.s.l.; B, Tjärnberg, 500 m a.s.l.; C, Skorvfjället, 808 m a.s.l.; D, Prästbodarna, 680 m a.s.l. Values for May 2006 are indicated with a symbol. Air concentrations of Ca^{2+} and K^+ were not available for Skorvfjället.

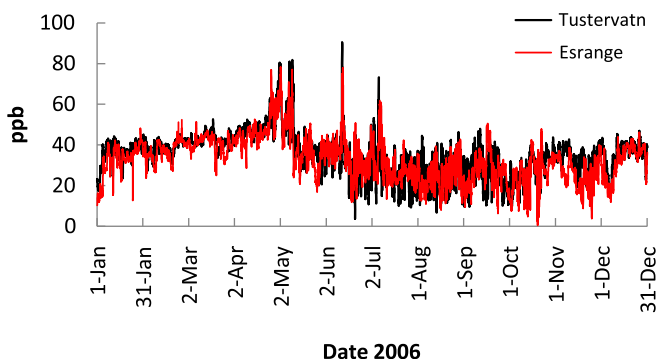


Fig. 7. Hourly ozone concentrations measured during 2006 at the Swedish sites Bredkälen and Esrange, the Finnish site Utö and the Norwegian site Tustervatn.

generally is taken up directly to the canopies and not included in the throughfall measurements (Parker, 1983; Ferm, 1993; Adriaenssens et al., 2012). The fraction of nitrogen taken up directly to the canopy is particularly large in low deposition regions, such as northern Sweden. During five years, 1997–2001, there were parallel measurements of bulk- and throughfall-deposition measurements at Nymyran. On average, the annual throughfall deposition measured was only 20% of the annual bulk deposition (data not shown). Normally, the dry deposition of nitrogen compounds at these high latitudes is small, due to the long distance from major emission sources at continental Europe, so the total deposition to the forests and to the open field should in principle be similar (mediated by wet deposition). Hence, at normal low levels of deposition approximately 80% of the nitrogen deposition to the canopy at Nymyran would have been taken up directly by the canopy, not reaching the throughfall collectors. However, in this unusual case with high ammonium deposition, it can be assumed that the

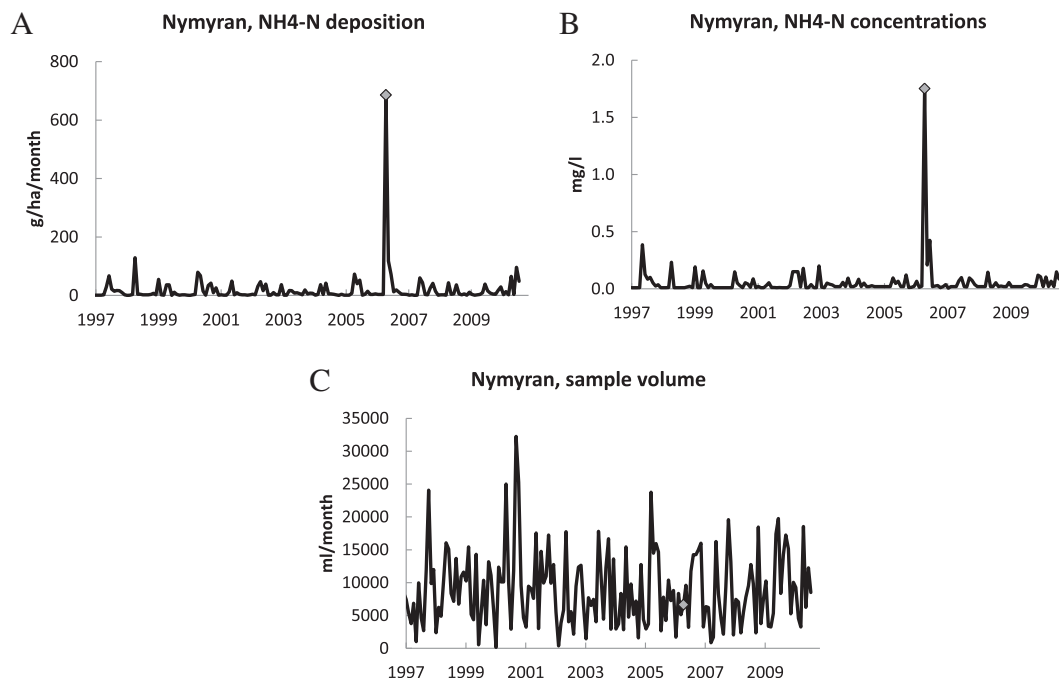


Fig. 8. Monthly ammonium deposition in throughfall at the low altitude site Nymyran, 75-year-old Norway spruce forest at an altitude of 300 m a.s.l. Data are shown for the calculated deposition (upper), the sample ammonium concentration (middle) and the sample volume (lower). A symbol is used for the value for June 2006.

fraction taken up directly by the canopy would be less. In any case, it seems likely to assume that the deposition of total NH_4^+ at Nymyran during May and June 2006 at least exceeded 1 kg N/ha. Since the normal, annual deposition of inorganic N in north Sweden is below 2 kg N/ha (Pihl Karlsson et al., 2011), this NH_4^+ deposition

event is of considerable importance. It has been shown that nitrogen wet deposition at Svalbard is strongly depending on a few episodic “strong” events (Kühnel et al., 2011) and a similar conclusion was made also for sulphur deposition at remote sites in Finland (Ruoho-Airola and Salmi, 2001).

Ammonia is generally emitted in a highly water-soluble form (e.g. Ferm, 1998). It will consequently to a large extent be dry deposited close to the source. Alternatively ammonia can form salt particles with available sulphuric acid droplets and gaseous nitric acid shortly after being emitted. These particles are in a size range that has a relatively low deposition rate to the ground. They are mainly scavenged out by clouds and sooner or later deposited to the ground as precipitation. Hence, in order for long-range transport of NH_x to occur, there must be high concentration of acidic species available and cloud-free weather (Ferm and Hellsten, 2012). ERA interim reanalysis data from the European Centre for Medium-Range Weather Forecasts (ecmwf.int) showed that a large anti cyclone was present above southeast Europe before and during the transport event in early May 2006. A very moderate large scale precipitation belt over Poland and the Baltic Sea (1–2 mm per day) was probably not enough to scavenge the particles generated from the fires during the rapid air mass transport towards northwest.

During the spring and summer 2006 unusual leaf visible injuries were observed on deciduous trees like birch (*Betula pubescens*) and rowan (*Sorbus aucuparia*) in northern Fennoscandia. Manninen et al. (2009) presumed these injuries to be effects of high O_3 concentrations. Similar injuries to these species, like what was observed in 2006, have not been observed during other periods (Tömmervik, personal communication). It is possible that the combined exposure to high ammonium deposition may exacerbate the impacts of other components like O_3 on vegetation, resulting in these unusual vegetation symptoms. Occasional frost nights during the spring of 2006 (annual reports: www.met.no; www.smhi.se) may also have reinforced the sensitivity and hence the injury (Manninen et al., 2009). Hence, there is a possibility that the dry deposition of ammonium might have contributed to these injuries, together with high O_3 concentrations.

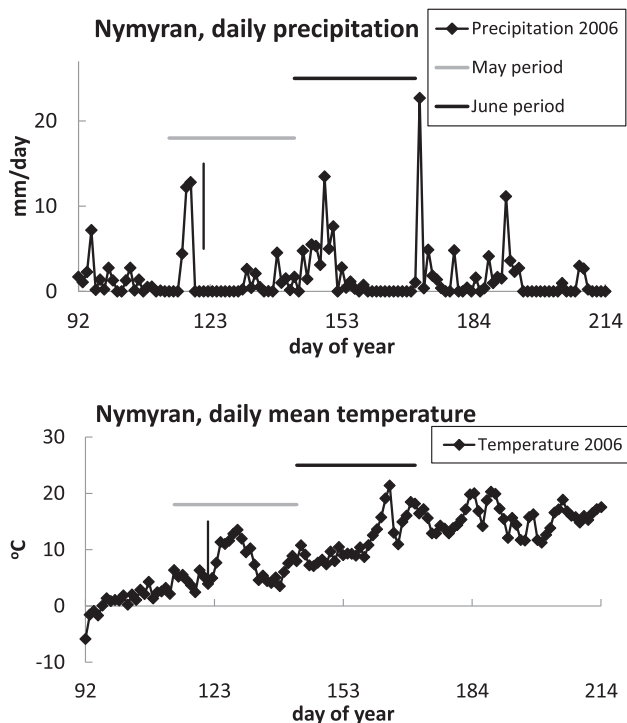


Fig. 9. Daily precipitation and daily mean temperatures at the site Nymyran for the period 1 April–31 July, 2006. The horizontal grey line indicates the sampling period for throughfall at Nymyran for May 2006 and the thick black horizontal bar the corresponding period June 2006. The thinner, vertical, black line indicates the date 1 May 2006.

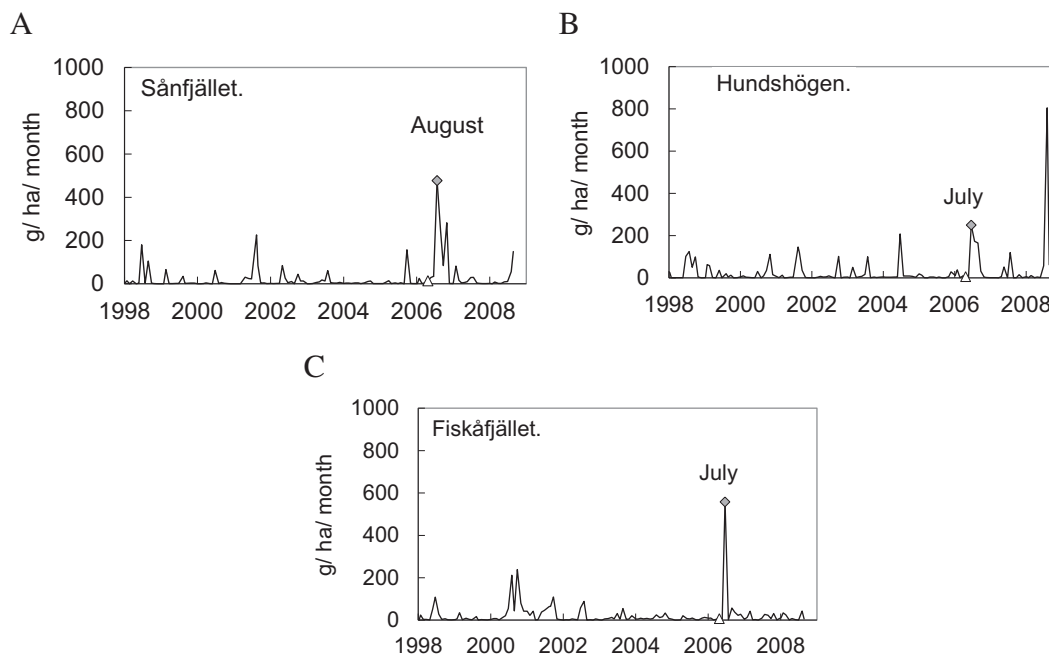


Fig. 10. Monthly ammonium deposition in throughfall at the high altitude sites in the county of Jämtland (600–800 m a.s.l.) Fiskåfjället, Hundshögen, Sånfjället. Throughfall deposition measurements are made with a slightly different methodology, as compared with low altitude measurements. Peak values are shown as a grey diamond together with an indication of the month. Values for May 2006 are shown as an open triangle.

4. Conclusions

Unusually high air concentrations of gaseous and particulate ammonia/ammonium were detected at high altitudes in central and northern Fennoscandia during May 2006, coinciding with a well described event with highly polluted air originating from large scale biomass burning in eastern Europe, passing over central Fennoscandia. High air concentrations of ammonia/ammonium and sulphate were also detected at low altitude areas over which this polluted air mass passed; at Utö, a small island between Finland and Sweden, at Breckkälen in the county of Jämtland, Sweden and at Tustervatn, a Norwegian site just north for Jämtland. At Tustervatn high potassium levels confirmed that the sources of air pollution was from biomass burning. At Breckkälen, also high air concentrations of black smoke were detected. Unusually high values for throughfall deposition of ammonium were detected during the summer 2006 at one low altitude site and several high altitude sites in the county of Jämtland. The occurrence of the high ammonia in throughfall in Jämtland varied between different summer months most likely related to precipitation events. The high ammonia air concentrations and dry deposition might have contributed to unusual visible injuries on the tree vegetation in northern Scandinavia occurred during 2006.

Long-range transport of polluted air from large scale biomass burning might have an important role for nitrogen deposition in northern Fennoscandia in a future climate change perspective.

Acknowledgements

The preparation of this article was financed by the Nordic Council of Minister (NMR). The deposition measurements in Sweden were operated by the Swedish Throughfall Monitoring Network (SWETHRO). The air pollution concentration and deposition measurements at high altitudes in the Swedish mountain region were financed by the Swedish Environmental Protection Agency. NILU is acknowledged for providing the FLEXTRA trajectories (www.nilu.no/trajectories) used in this study.

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