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Changes in the concentration of reduced nitrogen in the air in Finland between 1990 and 2007

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The concentration of reduced nitrogen in the air declined in 1990–2007 at the Finnish background stations in the southern and central parts of the country. The annual mean values, the summer mean values and the winter mean values declined by 35%–60%. In 2007 the annual mean values were 0.1–0.4 μg N m^{-3}. The data from Utö and Oulanka were analysed in transport sectors. At both stations, the foreign contribution from the southern, southwestern and western sectors was dominant in the reduced nitrogen concentrations. In Utö, the ammonium concentration decreased in the air arriving from the sectors extending from the south to the west and in the air transported from the east. In Oulanka, the concentration did not decrease in any of the transport sectors. However, the exposure of reduced nitrogen from the west declined at both sites. In Utö, this was due to the decline in both the ammonium concentration and the frequency of western transport. In Oulanka, the reduced exposure was due to a shift of air transport from the west to the north.

Introduction

Reduced nitrogen, such as ammonia (NH_3) and ammonium (NH_4^+), is essential in cultivation. Due to the low nitrogen-use efficiency in agriculture, about half of the global industrial production of ammonia is estimated to be dispersed into the atmosphere (Erisman *et al*. 2007a). This excess reduced nitrogen can cascade through atmospheric, terrestrial, aquatic and marine pools causing several harmful effects (Erisman *et al*. 2007a, 2007b). Reduced nitrogen can play an important role in the control of the acidification of precipitation (Galloway 1995). Ecosystems can undergo acidification and eutrophication as a result of the deposition of reduced nitrogen (Erisman *et al*. 2003). Eutrophication is one of the critical risks for the Baltic Sea and according to a recent extensive assessment, the airborne load of reduced nitrogen on the sea surface and on the catchment area is one important factor causing eutrophication (HELCOM 2009). Excess deposition of reduced nitrogen can also threaten the biodiversity of ecosystems (Krupa 2003). In addition to the ammonia emissions and the atmospheric transport, the reduced nitrogen concentration is affected by the nonlinearity of the relationship between the emissions and depositions of the sulphur and nitrogen compounds (Fricke and Beilke 1992, Fowler *et al*. 2005). Thus, reduced nitrogen can have negative effect on people's health by stimulating the formation of secondary particulate matter in the atmosphere (e.g. Seinfeld and Pandis 1997, Erisman and Schaap 2004).

In parallel with the strong decline of sulphur emissions during the past decades, the

relative importance of the nitrogen compounds cycling through the atmosphere has increased. The annual ammonia emissions in Europe are of the same magnitude as those of nitrogen oxides, calculated as nitrogen (Vestreng 2003, *see* also http://www.ceip.at/emission-data-webdab). They are mainly the result of agricultural practices. In Europe, the ammonia emissions declined by about 20% between 1990 and 2005 (Aneja *et al*. 2008). Most of the reduction took place as a result of the economic situation in the agricultural sector, with only a small part of the reduction due to specific measures designed to reduce emissions (Erisman *et al*. 2003). The Protocol to Abate Acidification, Eutrophication and Ground-level Ozone (Gothenburg Protocol) under the framework Convention on Long-range Transboundary Air Pollution (CLRTAP) (UN/ECE 1999) established within the United Nations Economic Commission for Europe (UN/ECE) was enacted in May 2005. This multi-pollutant, multi-effect protocol sets targets for emission cuts by 2010. For the first time also the emissions of ammonia are included. Countries, whose sulphur and nitrogen emissions have the most severe health or environmental impact and whose emissions are the cheapest to reduce, will have to make the largest cuts. In parallel, the EU has developed polices limiting the total emissions of each Member State and setting legally binding limits. The National Emissions Ceiling Directive (NECD) is a key EU policy. Member States shall limit by the end of 2010 their annual national emissions of ammonia to amounts not greater than the given emission ceilings (EU 2001). In 2009, the European Commission plans to publish a proposal to revise the current NECD, including stricter ceilings for the year 2020 (EEA 2009). Investigation of the emissions, transport and concentrations of atmospheric ammonium is thus currently of considerable interest.

The ammonia emission surveys used in atmospheric chemistry and transport model calculations are assumed to contain a larger uncertainty than the emission surveys of other compounds related to acidification (Erisman *et al*. 2003, Syri *et al*. 2000). According to Syri *et al*. (2000), the agricultural ammonia emission inventories are based on statistics on e.g. fertilizer use, animal number, and the applied control measures. The average emission coefficients are based on limited measurements. The largest uncertainties are connected with the evaporation rates and the employed reduction techniques. Thus, observation-based estimation of the changes in the transport of ammonium gives additional information when considering how the reductions of the European emissions are reflected in the concentrations of atmospheric ammonium in Finland.

We report the concentrations and trends in atmospheric reduced-nitrogen at four Finnish background stations between circa 1990 and 2007. As the pollutant concentrations in all north European countries are strongly dependent on long-range transport, they are affected not only by the changes in European emission patterns but also by the changes in the atmospheric circulation in Fennoscandia. Therefore, changes in the transport routes of air masses are also examined and used to explain the behaviour of the concentrations at different stations.

Experimental

Stations and monitoring

Daily measurements of the concentration of the sum of gaseous (NH_3) and particulate (NH_4^+) ammonium in the air were started between 1989 and 1991 at four background air quality monitoring stations of the Finnish Meteorological Institute (Fig. 1). The stations operate under the EMEP (European Monitoring and Evaluation Programme) programme within the CLRTAP. The sampling time was extended to one week in 2003 at two of the stations, Ähtäri and Oulanka. The Ähtäri station was relocated 6 km northwards in 1997 due to increased traffic on the nearby road. Dates of the start and changes in the monitoring are presented in Table 1.

For the sampling, an open-face filter pack with an oxalic acid impregnated Whatman 40 filter was used. The sampling time was 24 hours or one week and the flow rate was about 15 l min^{-1}. The sample volumes were measured by a gas meter (Kimon PK III) checked at least twice a year against a similar reference gas meter. The sampling method is described in detail by Karls-

son *et al*. (2007). During the period 1989–1994, the filters were extracted with H_2O and analysed using the spectrophotometric indophenol blue method. Since 1995, ion chromatography has been used instead. Karlsson *et al*. (2007) studied the properties of the filter-pack method in monitoring of sulphur and nitrogen containing inorganic gases and particles. The combined expanded measurement uncertainty including sampling and analysis was found to be very near the analytical uncertainty when measuring higher atmospheric concentrations, being ± 4.5% for total ammonium concentrations > 0.8 μg m^{-3}. The detection limit was 0.05 μg N m^{-3} when calculated as three times the standard deviation of the field blanks. The sampling and analysis, as well as the quality control of the whole chain, was carried out using the best available practices according to the guidelines given in the EMEP Manual (EMEP 2001). The concentrations are given at temperature 20 °C and local pressure. The sampling method does not allow the separation of ammonia and ammonium and their amount is therefore expressed as a sum and quantified as nitrogen.

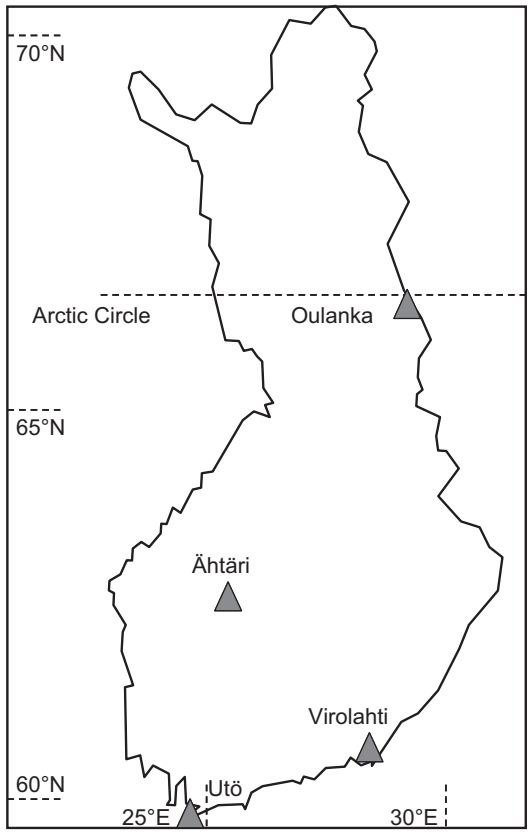

Fig. 1. Location of the sampling stations used in this study.

Trend calculation and sectoral analysis

The annual, as well as the summer and winter average concentrations of reduced nitrogen in the air at each station were calculated for the trend study. In analysing the differences between the seasons, the months from October to March were included in the winter season, and the months from April to September in the summer season. The trend calculations were performed with the MAKESENS software, which is developed at the Finnish Meteorological Institute (FMI) (Salmi *et al*. 2002) and used widely for the assessment of the EMEP data (e.g. Bartnichi and Lövblad

Table 1. Statistics of the daily concentration of reduced nitrogen (μg m^{-3}) in air and dating of the measurement activities at four Finnish background stations. All values are calculated for the entire monitoring period with daily measurements at each of the stations.

	Utö	Virolahti	Ähtäri	Oulanka
Maximum	4.70	9.25	7.92	1.97
95th percentile	1.52	2.14	1.24	0.59
Mean	0.49	0.79	0.45	0.17
Median	0.32	0.58	0.31	0.10
SD	0.50	0.67	0.40	0.21
Number of daily values	6494	6765	5411	4739
Start of measurements	3 July 1989	1 Feb. 1989	19 Jan. 1989	30 July 1990
Change to weekly sampling			28 Dec. 2003	21 Sep. 2003
Relocation of station			29 May 1997	

2004). The existence of a monotonic increasing or decreasing trend was tested with the nonparametric Mann-Kendall test as a two-tailed test (Gilbert 1987, Salmi *et al*. 2002). The Mann-Kendall test is suitable for data for which the trend is estimated to be monotonic and no seasonal or other cycles are present. For the estimate of the slope of the trend, nonparametric Sen's method was used (Gilbert 1987, Salmi *et al*. 2002). Sen's method uses a linear model to estimate the slope of the trend. The method calculates the slopes of all value pairs in the data and Sen's estimate is the median of these slopes. Missing values are allowed and Sen's method is not greatly affected by single data errors or outliers.

The exposure to reduced nitrogen from different transport sectors was studied with the help of backward trajectories. The daily concentrations measured at each station were classified into eight equal-size sectors using two-dimensional 925-hPa trajectories describing the transport route of the air mass for 96 hours backwards, four times per day, along modelled wind fields. The north sector extends from –22.5° to 22.5° and the next sectors were counted clockwise from it. The area around the arrival point extends from a radius of 150 km to a radius of 1500 km. The division of the trajectories in sectors was obtained from the EMEP/MSC-W (http://www.emep.int/index_data.html). The criterion for the allocation of the trajectories of a particular arrival day to a specific sector was that at least 50% of their given positions during transport were found within that sector. If the criterion was not fulfilled, the day was set as unclassified. About 30% of the days remained unclassified.

The frequencies of the transport from the different sectors and the sectoral mean concentrations were calculated. Further, the accumulated exposure from different sectors was calculated from the sector mean concentration and the number of days with transport from that sector. The software TRASEC developed by the Norwegian Institute for Air Research (NILU) and FMI for the use in EMEP Assessment (http://www.emep.int/assessment/) was applied. The exposure is given on hourly basis in mg h m^{-3}. The methods used in the monitoring and the sectoral analysis are described in detail by Ruoho-Airola *et al*. (2004).

Results and discussion

Seasonal variations and trends

The ammonium concentration was highest at the southeastern station Virolahti and lowest at the northeastern station Oulanka (Table 1). The medians of the daily values for the whole study period in Virolahti and Oulanka were 0.6 and 0.1 μg N m^{-3}, respectively. At the two other stations, Utö and Ähtäri, the median value was 0.3 μg N m^{-3} for both. The maximum daily ammonium concentrations were 2, 5, 8 and 9 μg N m^{-3} in Oulanka, Utö, Ähtäri and Virolahti, respectively, probably reflecting the influence of local agriculture in Ähtäri and Virolahti. The distribution of the daily values was skewed, with a large amount of very low concentrations and occasional elevated values (*see* Table 1). The median is 60%–80% of the annual mean value and the maximum value is 3–6 fold as compared with the 95th percentile.

The seasonal variation in the ammonium concentration had the same pattern at all four stations with low monthly mean concentrations during winter and elevated concentrations in March–April and in summer, especially July–August (Fig. 2). The highest monthly mean was about two times greater than the lowest.

At all sites, the concentration of reduced nitrogen declined in the first half of the 1990s (Fig. 3). A further decrease occurred at the three more southern stations, Virolahti, Utö and Ähtäri, even though also elevated concentrations occurred between 1996 and 2006. The lowest values of the time series at these stations were recorded in 2007. The highest annual mean values, 1.1 and 1.2 μg N m^{-3} were measured in Virolahti in 1989 and 1999, respectively. In Oulanka, the reduced nitrogen concentration fluctuated between 0.1 and 0.2 μg m^{-3}. At the end of the study period, the range of the annual mean was 0.1–0.4 μg N m^{-3} at all stations.

The temporal trends in the concentrations of atmospheric ammonia at the four stations were confirmed with the Mann-Kendall test (Table 2). The reduced-nitrogen concentration decreased in Utö, Virolahti and Ähtäri from 1989 (Utö 1990) to 2007. A statistically significant trend was found in the annual mean values and in the

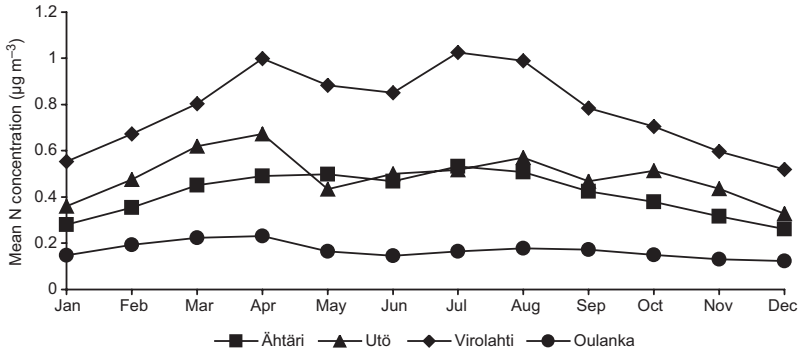

Fig. 2. Monthly mean concentration of reduced nitrogen in the air at the Finnish background stations from the start of the monitoring until 2007.

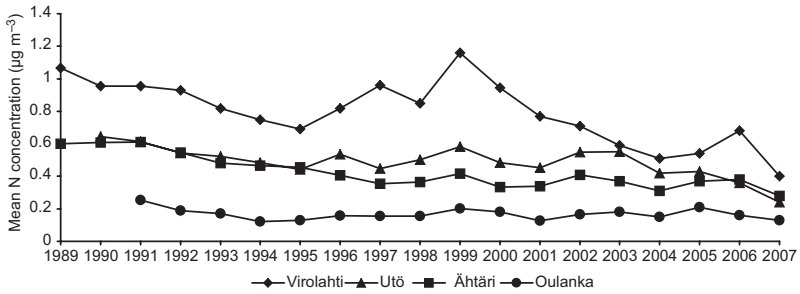

Fig. 3. Time series of the annual mean concentration of reduced nitrogen in air at the Finnish background stations.

summer and winter mean values. The change for the total period was –50% in Virolahti and Ähtäri and –35% in Utö. The decline was largest (–60%) in the summer period in Ähtäri. In contrast to the other stations, no statistically significant trend was obtained in the Oulanka data series.

According to the trend analysis carried out in Sweden, the concentration of reduced nitrogen in the air was slightly higher than in Finland. In 1987 and 1998, the highest annual mean concentrations in Sweden were 2.1 and 1.1 μg N m^{-3}, respectively (Kindbom *et al*. 2001). A decreasing trend was detected at all studied sites, with the total decline between –25% and –65%. It is notable that the northernmost site is located at the latitude 63°N, which is about the same as Ähtäri in our study.

Fagerli and Aas (2008) analysed the trends of nitrogen compounds in the air using the EMEP measurements and EMEP model results at the monitoring sites during 1990–2003. For reduced nitrogen, statistically significant downward trends were observed in Finland only in Ähtäri (–46%) and in Sweden at two stations (–25 and –40%). At two Norwegian sites, large positive trends (+89% and +217%) were detected, which were assumed to be due to local emissions. Elsewhere in Europe, in Denmark, the Czech Republic and Poland, a statistically significant decline was found at most sites.

Table 2. Trends (and their significances) in the concentration of reduced nitrogen in the air at background stations in Finland. All time series end in 2007. n = number of monitoring years. Q is the best estimate for the trend, Q_{min} and Q_{max} are the 95% confidence limits for the estimate of the trend. The unit is μg m^{-3} a^{-1}. Empty cell = no sign. trend.

Station	Start	n	p	Sen's slope estimate			Change (%) per period	Summer		Winter	
				Q	Q_{min95}	Q_{max95}		p	%	p	%
Virolahti	1989	19	< 0.01	–0.027	–0.041	–0.015	–50	< 0.01	–50	< 0.001	–45
Utö	1990	18	< 0.01	–0.013	–0.019	–0.005	–35	< 0.01	–35	< 0.01	–40
Ähtäri	1989	19	< 0.001	–0.016	–0.022	–0.010	–50	< 0.001	–60	< 0.01	–35
Oulanka	1991	17									

The decrease in reduced nitrogen in the air is reflected in the wet deposition of ammonium. In 1990–2000, the wet deposition decreased by 35%–40% in Utö and Ähtäri, while in Virolahti the large interannual variation prevented a monotonic trend (Ruoho-Airola *et al*. 2004). However, according to the EMEP monitoring data of the Finnish Meteorological Institute, the level of ammonium deposition halved in Virolahti between 1990 and 2007. In Oulanka, the level of the ammonium deposition has remained the same since 1990, and in Utö and Ähtäri since 2000.

In general, the pH value of rain water has increased over the last decades in consequence of the emission reduction of compounds related to the acidity of the precipitation. In 1993–2003, a negative trend of the H^+ concentration was found in whole Finland as well as at most stations in Europe (Kleemola and Forsius 2006). However, the effect of the decrease in the ammonium concentration is covered by the changes of the other regulators.

Changes in emissions and meteorological conditions

Several factors, including changes in the domestic emissions and the European emission pattern as well as the interannual variability of meteorological conditions have influenced the concentration of the reduced nitrogen. These changes are discussed below.

A minor decline in the ammonia emission in Finland was recorded between 1990 and 2006: from 38 to 36 kilotonnes NH_3 (Finnish Environment Institute 2008). However, an uncertainty of ±70% was estimated in the 2006 agriculture emissions (Finnish Environment Institute 2008). According to a recent detailed emission analysis, the highest emission areas in Finland in 2000 were situated in the southwest and along the northwestern coast of the Gulf of Bothnia where agriculture is intensive (Karvosenoja 2008). During the dominant western and southwestern air flows, large areas of the country are under the influence of domestic emissions.

However, the Finnish emissions are very low as compared with those of all the European countries which were estimated to be 6.5 million tonnes of NH_3 in 2000 (Vestreng 2003, Lövblad *et al*. 2004). Between 1990 and 2000, the largest relative decrease took place in the eastern European countries, where the emissions went down by nearly 50%. However, during that period the reduction in most European areas was only around 10% (Lövblad *et al*. 2004). In Norway, Sweden and in the southwestern Europe, even increases of the emissions between 1990 and 2003 have been reported (Fagerli and Aas 2008). According to the EMEP model estimates, the fraction of the domestic contribution to the total deposition of the reduced nitrogen in Finland was 30% in 2006 (Gauss *et al*. 2008). Along the southern and eastern borders, the transboundary contribution was 80%–90%, and in Utö and Oulanka it was higher than 90% (Gauss *et al*. 2008). The main contributors to the wet deposition of the reduced nitrogen in Utö and Ähtäri were Poland (15%–20% in 2003) and Germany (11%–15% in 2003) (Fagerli and Aas 2008).

A reduction in the precursor emissions of a particular pollutant may influence the transport distance of other pollutants (Fricke and Beilke 1992, Fowler *et al*. 2005). Thus, the concentration of reduced nitrogen is affected also by changes in the European sulphur emission pattern. The reduction in sulphur dioxide emissions are estimated to have decreased the reduced nitrogen in the air by 15%–25% in 1990–2003, because the formation of ammonium nitrate only partly compensated for the reduced formation of ammonium sulphate (Fagerli and Aas 2008). The largest effects were observed in areas with low ammonia emissions such as the northern parts of the Nordic countries.

The interannual variation of the contribution of air pollutants from one country to other due to the meteorological conditions is estimated to be up to 20% (van Loon *et al*. 2005). In addition, there has recently been a more long-term change in the air flow in Fennoscandia. The westerly wind flow from the North Atlantic was exceptionally strong after 1989 but decreased gradually during the 1990s, which enabled the increase of the southerly wind flow from 1990 to 2000. However, the westerly wind flow remained higher than the usual level in the earlier years (Tuomenvirta *et al*. 2000, Moberg *et al*. 2005).

Sectoral differences in Utö and Oulanka

According to our results, the concentrations of reduced nitrogen decreased in all locations but Oulanka. It is probable that the changes in the atmospheric circulation in Fennoscandia, in addition to the changes in the European emission patterns, have prominently affected the development of the concentrations. Therefore, we used the daily mean concentrations and atmospheric transport sectors to study the differences between the sites.

We assume that the reduced nitrogen in Utö and Oulanka is mainly long-range transported particulate ammonium and not notably affected by local ammonia emissions. This assumption is supported by the estimated high transboundary contribution to total deposition of ammonium in Utö and Oulanka obtained from the EMEP transport model calculations (Gauss *et al*. 2008). Besides, the Utö station is located on a rocky island, south of the Finnish coastline and the domestic emissions, and the Oulanka station is near the Arctic Circle. Both stations are far from agricultural activities and, consequently, the local ammonia emissions are very low (http://www.ceip.at/emission-data-webdab/). In addition, measurements of ammonium compounds with a denuder in Utö in March–June 1993 showed that the particulate ammonium covers on average 86% of the reduced nitrogen (Sorteberg *et al*. 1998). Further, in order to inspect the possibility of local ammonia emissions, we compared in equivalents the daily mean concentration of reduced nitrogen in the air with the sum of the sulphate and oxidised nitrogen. In the highest 5% of the daily mean reduced-nitrogen concentrations, we found 8% and 4% cases with any excess reduced nitrogen in Utö and Oulanka, respectively. Thus, we believe that the ammonium concentrations in Utö and Oulanka are strongly dependent on changes in the air transport routes to the locations.

Daily mean values for the atmospheric ammonium concentration were available for Utö for the years 1990–2007 and for Oulanka from 1991 until the end of the 2003 summer season. Daily sector values describing the transport route of the air masses to the location were derived from the EMEP programme (http://www.emep.int/index_data.html). They covered the study period from 1990 to the end of the year 2006 for both stations. The data enabled us to study the exposure to atmospheric ammonium from different transport sectors in Utö and Oulanka and the possible changes in the exposure pattern.

For each year, each summer season and each winter season the mean concentration of atmospheric ammonium arriving from different transport sectors as well as the frequency of the sectors and the ammonium exposure were calculated. The sectoral exposure means here the sum of daily loads arriving from a specific sector over the time period assessed.

At both stations, the ammonium concentrations were generally high in the air arriving from the sectors southeast (SE), south (S) and southwest (SW). The significance of the trends in the annual and seasonal mean concentrations from the different transport sectors was tested with the Mann-Kenndall test, with a 10% ($p < 0.10$) risk for no trend (Table 3). During 1990–2006, the ammonium concentration in Utö decreased in the air transported from the sectors east (E), south (S) and west (W). When the seasons were calculated separately, the concentration decreased also in the southwest (SW) sector, both in summer and in winter. On the other hand, in Oulanka, the concentration did not decrease in any of the transport sectors. A slight increase in the annual mean concentration was detected in the air arriving from the sector north (N), and in winter from the sector northwest (NW). However, the mean concentrations remained low as compared with those from the SE, S and SW sectors. Thus, the ammonium concentrations in some sectors with high concentrations and high influence on the average concentration (S, SW) decreased at Utö, while no decrease occurred in Oulanka. However, as there were fewer values in the Oulanka trend calculations, the possibility to find a significant trend was somewhat lower there.

Trajectories for both stations covered years to the end of 2006. The transport to both sites occurred most frequently from the sectors W, SW, NW and N. The frequency of the prevailing western air masses declined significantly in Utö in winter, while an increase was detected in the transport from the northern sectors. The same change is seen also on an annual basis (Table 3).

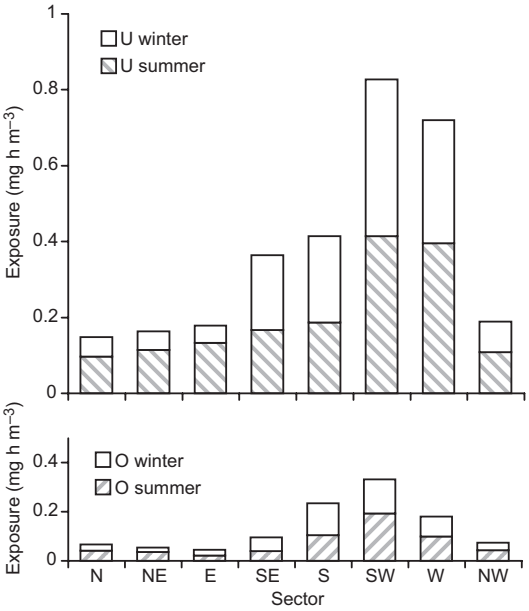

Fig. 4. Exposure to ammonium from different air transport sectors (N to NW). The values are averages accumulated during a summer or winter season for Utö (1990–2006) above, and Oulanka (1991–September 2003) below.

The SE transport, on average carrying elevated ammonium concentrations increased slightly in summer. In Oulanka, the transport from the NE sector increased during the winter periods.

At both stations Utö and Oulanka, the contributions from the S, SW and W sectors were dominant for the measured ammonium concentration (Fig. 4). During some years, also the SE exposure grew high. Both the concentration and the frequency from the west decreased in Utö. Consequently, the exposure of ammonium from the western sector declined significantly (Table 3). In spite of the significant increase of the northern exposure, its contribution to the annual concentration remained low.

In Oulanka, the exposure from the W sector decreased as well (Table 3). Exposure from the northern sectors (N, NW) increased in summer. In winter, a decrease was detected in the exposure from the SW sector. Because the fraction of the transport from the sectors SE–SW with elevated concentrations is usually larger in Oulanka than in Utö (Fig. 5), the decrease in the western transport has a lower influence on the mean concentration in Oulanka than in Utö. In addition, the distance to Oulanka from the large emission areas is longer, which causes larger episodicity in the annual load (Ruoho-Airola and Salmi 2001). Especially, the occassional transport from the SE with an elevated ammonium concentration most probably has counteracted the decrease in the western sectors in Oulanka, where the annual mean concentration did not decrease.

The atmospheric ammonium concentration in Finland is strongly affected by the changes in the ammonia emissions in large areas of Europe and also by changes in the air transport routes. The annual concentration decreased in Utö because the concentration in the air transported from sectors extending from south to west decreased due to emission reductions along the route. In

Table 3. Trends in the concentration of ammonium in the air, the frequency of transport and the exposure from different atmospheric transport sectors. Time periods: Utö: 1990–2006, Oulanka: concentration and exposure 1991–end of summer season 2003, frequency 1991–2006. AN = annual, SU = summer, WI = winter. Sectors from N = north to NW = northwest. Sign. of trends $p < 0.10$, – = decreasing trend, + = increasing trend, empty = no sign. trend.

	Utö									Oulanka								
	Concentr.			Frequency			Exposure			Concentr.			Frequency			Exposure		
Sector	AN	SU	WI	AN	SU	WI	AN	SU	WI	AN	SU	WI	AN	SU	WI	AN	SU	WI
N				+		+			+	+						+	+	
NE						+									+			
E	–																	
SE					+													
S	–	–	–															
SW		–	–															–
W	–	–		–		–	–		–							–	–	–
NW						–						+					+	

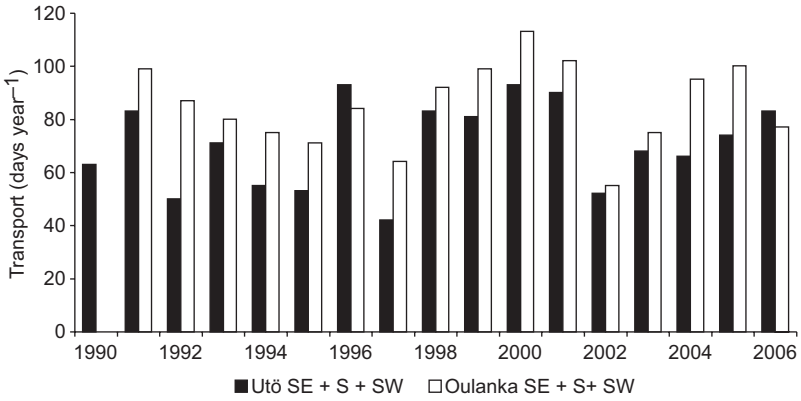

Fig. 5. Frequency of the transport of air from the sectors SE, S and SW to the stations.

addition, there was a shift in the frequency of air flow from the western sector to the more clean northern sector. In Oulanka, the decrease in the exposure from the west was not large enough to lower the annual concentration, mostly because of the high fraction of the transport from the sectors SE–SW with elevated concentrations.

Conclusions

We show that the reduced nitrogen concentration in the air decreased significantly at the background stations in southern and central Finland during 1990 and 2007, but no significant trend was detected at the northeastern site. The decline was largest (–60%) in summer at the site in the western central Finland. At the southernmost and northernmost stations, the reduced nitrogen in the air is assumed to consist mainly of ammonium particles transported from a long distance. Thus, the partition of daily values to different air transport sectors is relevant. At both stations the ammonium concentrations are generally high in the air arriving from southern sectors between southeast and southwest. In Utö, a significant decrease is shown in the reduced nitrogen concentration in the air transported from the east, southeast, south and southwest. In Oulanka, the concentration increased significantly in the sectors north and northwest but the change in the low concentrations from those sectors has only a minor influence on the changes in the mean values.

The changes in the atmospheric circulation in Fennoscandia have, in addition to the changes in the European emission patterns, an important effect on the atmospheric ammonium concentration in Finland. The westerly wind flow from the North Atlantic was exceptionally strong after 1989 but decreased gradually during the 1990s, which enabled the increase of the southerly and northerly wind flow. At the two stations studied, Utö and Oulanka, the air transport is most frequent from the sectors west, southwest, northwest and north. Occasionally, exposure from the southeast is also frequent. The emission reductions of nitrogen and sulphur compounds in these directions have the largest effect on the atmospheric concentration of reduced nitrogen in Finland.

The reduced nitrogen concentration is controlled by a complex combination of regulators. In addition to the ammonia emissions and the atmospheric transport, the ammonium concentration is affected by the nonlinearity of the relationship between the emissions and depositions of sulphur and nitrogen. The long-range transport of ammonium is strongly dependent on the atmospheric chemistry of sulphur and nitrogen. Thus, the difference in the trends of the ammonium concentration in different sectors and different stations is partly explained by the changes in the concentrations and emissions of the other air pollutants. As a consequence, there could also be changes in the scavenging of reduced nitrogen from the air.

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