Acid deposition trends at the Finnish Integrated Monitoring catchments in relation to emission reductions

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The effects of transboundary air pollutants on deposition levels and precipitation concentrations have been monitored within the UN/ECE Integrated Monitoring Programme since 1988 at four Finnish background stations. A monotonic decreasing trend was detected for the concentration and deposition of sulphate, nitrate, ammonium and potassium at the southern stations. Further to the north, fewer compounds showed a significant reduction. For sulphur, model calculations with the operational deposition model DAIQUIRI also indicate a clear decrease in deposition. At the southern stations this reflected rather well the reduction of sulphur emissions in Finland and elsewhere in Europe. In the northern part of the country, where the distance to the major emission sources is longer and transport is more affected by meteorological parameters, wind direction especially, the annual amounts of sulphur deposition varied more independently of the changes in emission levels. The proportion of domestic oxidised nitrogen and total ammonium deposition decreased on moving northwards. The downward trend for nitrogen compounds throughout the country seems to be due to international emission reductions.

Introduction

The long-range transport of acidifying components has been a severe environmental problem in Finland and the other Nordic countries for decades. In other parts of Europe the damage caused by acidifying compounds to natural ecosystems has likewise given rise to concern. The UN-ECE Convention on Long-range Transboundary Air

Pollution (LRTAP) resulted in a protocol to reduce sulphur emissions by 1993 to 30% of the emissions in 1980, being signed by 23 European countries in 1985. A new protocol to further reduce sulphur emissions in Europe followed in 1994. The protocol to control emissions of nitrogen oxides and their transboundary fluxes signed in 1988 requested that nitrogen oxides emissions in 1994 should not exceed those in 1987. In Fin-

land a further reduction in the deposition of acidifying compounds would still be needed, because the critical loads of acidity are still being exceeded in many parts of the country (Posch *et al.* 1997).

The Integrated Monitoring (IM) Programme is one of five International Co-operative Programmes currently being carried out under the Convention on Long-Range Transboundary Air Pollution. The aim of the programme is to monitor the effects of transboundary air pollution on natural ecosystems. The results of the IM Programme are being used to determine the amount of acid deposition entering the terrestrial ecosystem. The effects of the international agreements on emission reductions can be followed within the IM Programme, and a recovery of the ecosystems should be reflected in the monitoring results.

The aims of this study are to evaluate the deposition of acidifying components in Finland on the basis of precipitation measurements made at the IM stations and model calculations, as well as to estimate changes in the load in relation to emission reductions.

Emissions

Emissions of acidifying components in Europe and Finland

The emissions of sulphur, nitrogen oxides and ammonia in the individual countries are given in the reports of the EMEP (Co-operative programme for monitoring and evaluation of the long range transmission of air pollutants in Europe) project under the LRTAP Convention. The present emission values are based on national emission reports and, in some cases, on estimates made within the framework of the EMEP project (Berge 1997a). The EMEP emission data base is currently probably the most complete and reliable emission survey covering the whole of Europe.

Table 1 shows the emissions of sulphur, nitrogen oxides and ammonia in Europe for the year 1987, the first year of IM monitoring, and for the year 1995. The emissions are given separately for Finland, and for the main source areas affecting acidifying deposition in Finland. There has been a strong decrease in the emissions of sulphur, and a less marked decrease in the emissions of nitrogen. These emission changes are in accordance with the emission reduction plans of the LRTAP Convention.

Emissions used in the model calculations

For the calculations with the DAIQUIRI (Deposition, AIr QUality and Integrated Regional Information) deposition model, detailed spatially located annual Finnish emissions were available from CORINAIR and other national inventories for the years 1990 and 1995 (Melanen and Ekqvist 1997). For other years, the 1990 emissions were scaled to the corresponding official emission figures. However, this does not take into account possible annual changes in emission distributions. The annual Finnish emissions of sulphur, nitrogen oxides and

Table 1. Emissions of sulphur and nitrogen in Finland and elsewhere in Europe in 1987 and 1995. The relative contribution (%) from the different emission source areas to deposition in Finland in 1995 are given in parentheses for oxidised sulphur, oxidised nitrogen and reduced nitrogen, respectively. Emission levels 1 000 tonnes per year.

Source area	1987			1995		
	SO_2	NO_x (as NO_2)	NH_3	SO_2	NO_x (as NO_2)	NH3
Finland	328	288	35	96 (11)	259 (15)	31 (34)
Nordic countries[1]	552	997	199	279 (4)	834 (17)	200 (7)
Russia, Baltic States	6 234	3 007	1 440	3 224 (23)	2 141 (8)	862 (12)
Eastern-central Europe[2]	8 263	2 808	876	4 365 (10)	1 876 (7)	668 (5)
Germany[3]	7 347	3 177	837	2 995 (8)	2 210 (10)	622 (5)
Western Europe[4]	5 863	5 241	1 376	3 762 (5)	4 844 (13)	1 247 (4)

[1] Sweden, Denmark, Norway. [2] Poland, Czech Republic, Hungary, Slovakia. [3] including former GDR. [4] France, U.K., Benelux countries.

ammonia during 1987–1995, as used in the model calculations, are listed in Table 2.

In this study the transport of sulphur compounds originating from Estonia, the Leningrad area, Karelia and the Kola Peninsula was also estimated with the mesoscale transport matrices of DAIQUIRI, since there are several large sulphur emission sources located so close to the IM stations that the 150 km grid of the EMEP/MSC-W (Meteorological Synthesizing Centre-West) model might induce significant resolution errors. Spatially detailed sulphur emissions for these regions were obtained from official Russian and Estonian emission statistics. For Estonian sulphur emissions, the results of a detailed study carried out by the Finnish Meteorological Institute were also applied (Kivivasara 1994). For nitrogen compounds, EMEP/MSC-W transport matrices were applied to all emissions outside Finland. The emission values used are reported in Barrett and Berge (1996).

Trends in acidifying deposition in Finland

Observed trends

Deposition chemistry has been monitored at four background stations of the Integrated Monitoring Programme: Kotinen, Hietajärvi, Pesosjärvi and Vuoskojärvi. The stations are located in areas well removed from major anthropogenic local sources, and are situated in different parts of Finland (Fig. 1).

Fig. 1. Location of IM stations in Finland.

Table 2. Emissions of sulphur, nitrogen oxides and ammonia in Finland and sulphur emissions in the neighbouring Russian regions and in Estonia during 1987–1995 as used in the model calculations. *) 1993 level, **) 1994 level, ***) 1990 level assumed. Fin = Finland, Len = Leningrad area, Kar = Karelia, Kol = Kola Peninsula, and Est = Estonia.

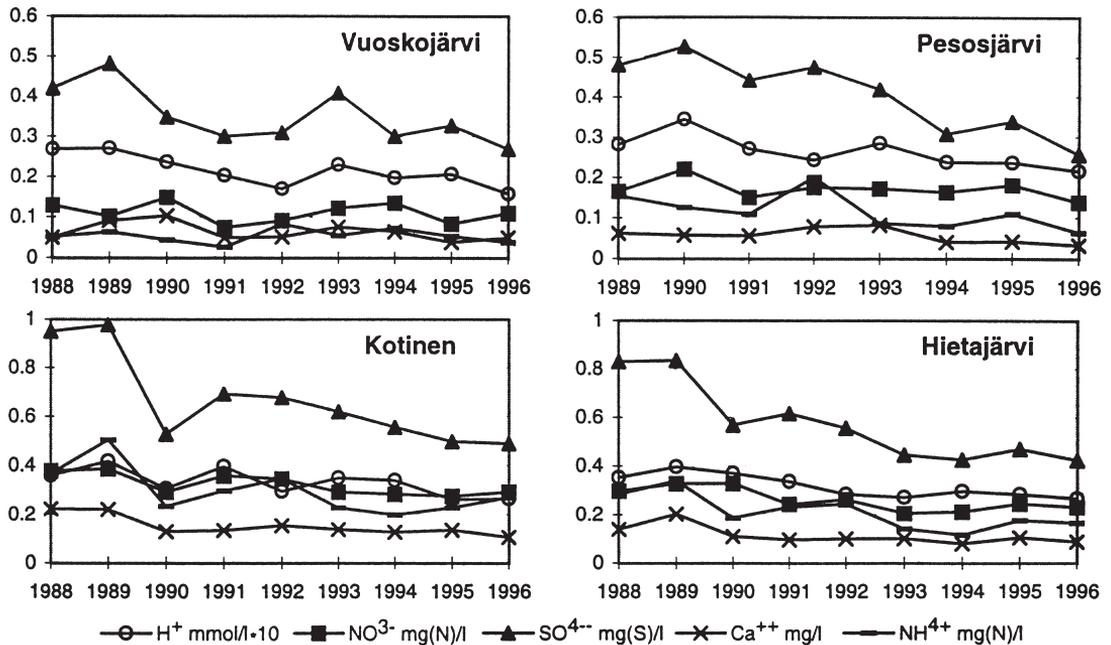| Year	Fin kt SO_2	Fin kt NO_2	Fin kt NH_3	Len kt SO_2	Kar kt SO_2	Kol kt SO_2	Est kt SO_2
1987	328	288	45	330	172	658	296***)
1988	302	293	44	285	167	598	296***)
1989	244	301	42	259	158	574	296***)
1990	260	300	41	281	173	601	296
1991	194	291	41	285	159	552	286
1992	141	282	41	250	147	517	179
1993	123	280	41	217	139	460	145
1994	116	283	41	217*)	134	456	141
1995	96	257	34	217*)	134**)	456**)	110

Fig. 2. Annual mean values of precipitation-weighted ion concentrations at the Finnish IM stations.

The representativeness of the deposition measurements at the IM stations has been tested by comparing the three-year results with corresponding results obtained in other parts of the country (Ruoho-Airola 1995). The annual mean deposition of sulphate, calcium, ammonium, nitrate and chloride were comparable with other regional values. Hydrogen ion deposition at the Hietajärvi station was somewhat higher than that at the other eastern stations. As no trend for potassium was found throughout the country, its representativeness could not be tested in the same way as for the other components.

Weekly bulk deposition measurements were started in 1987 at the IM stations, except for Pesosjärvi where the measurements were started in 1988. The weekly samples were combined after visual checking to give monthly samples and analysed in the laboratory of the Finnish Meteorological Institute. High quality assurance principles were followed in the sampling protocol and chemical analysis procedures. The ECE/EMEP Data Quality Objectives (EMEP 1996), i.e. a maximum of 15%–25% uncertainty for the combined sampling and chemical analysis of major ion deposition measurements, were mostly met with the help of three parallel sampling devices (Ruoho-Airola and

Leinonen 1997). A detailed description of the programme is given in the relevant manuals (Environment Data Centre 1989, 1993).

Precipitation-weighted annual mean values for acidifying components at the stations are given in the Appendix and the time series of sulphate, nitrate, ammonium, calcium and hydrogen ion concentrations in Fig. 2. There was a reduction in the concentrations of hydrogen ion and sulphate at all stations during the monitoring period. The sulphur concentration decreased by 45%–50% at Kotinen, Hietajärvi and Pesosjärvi, and by 35% at the northern station at Vuoskojärvi. The largest absolute decrease in the sulphate concentration occurred in the first years of the monitoring period. The exceptionally dry summer in 1990 resulted in the minimum mean concentration at Kotinen in 1990. There is usually a sulphate concentration peak during the summertime and this, together with the relatively high precipitation during the same period, has a strong effect on the annual mean. In the two southern catchments there was also a slight decline in nitrate, ammonium and calcium concentrations.

The non-parametric Seasonal Kendall slope estimator is a recommended and widely used method for the time-series analysis of environ-

mental data characterised by skewness, seasonality and/or serial dependence (Gilbert 1987, Hirsch *et al.* 1982, Driscoll *et al.* 1995). This method is based entirely on ranks within the time series, and is intended to identify and quantify significant monotonic trends. The trends for acidifying compounds (Table 3) were calculated from the monthly concentration and deposition values. There was a significant decrease in both the concentration and deposition of all acidifying compounds — sulphur, nitrate, ammonium and hydrogen ion — at Kotinen in southern Finland. The similar decreasing trend in the concentration and deposition of base cations reduces the positive effects of this change in the acidifying load. At Hietajärvi the trends for acidifying compounds were similar to those at Kotinen, but the magnitude of the decline was smaller, except for the hydrogen ion concentration. There was no significant change in hydrogen ion deposition. Further to the north, fewer compounds show significant reductions; at Vuoskojärvi there was a reduction only in the sulphur, hydrogen ion and calcium concentrations and potassium and calcium depositions. None of the stations showed a significant trend in the amount of precipitation.

Several studies cover trend analysis of the data for European background air pollution stations. The steep decline in sulphur dioxide concentration measured widely in Central and Northern Europe did not necessarily lead to a significant decrease in the concentration of sulphate in precipitation. The non-parametric Mann-Kendall method for trends in the sulphur components was used by the EMEP Chemical Co-ordinating Centre (EMEP/CCC) on the data monitored in the EMEP programme. They found a negative trend of 0.04–0.08 mg (S) l^{-1} per year for sulphate in precipitation at the Finnish southern stations for the time period 1986–1993 (Schaug *et al.* 1996). Their result is in good agreement with the outcome of this paper. A significant downward trend for sulphate was obvious at sixteen of the 28 European stations tested. Fricke and Beilke (1993) reported changes in the concentration and deposition of sulphur and nitrogen compounds in rural areas of central Europe during the period 1980 to 1992. In their study both wet and dry deposition fluxes were examined. The reduction in total sulphur deposition at three German stations was between 33%–57%; wet deposition had decreased by 17%–38%, and dry deposition by over 50%. In contrast to sulphur, nitrogen dioxide as well as nitrate and ammonium in precipitation had de-

Table 3. Significant monotonic trends calculated by the Seasonal Kendall method from 1988 to 1996 (1989–1996 at Hietajärvi) in precipitation concentration (C) and deposition (D). Slope units are mg $l^{-1}a^{-1}$ (C) and mg m^{-2} month^{-1} (D), except H$^+$ µmol $l^{-1}a^{-1}$ (C) and µmol m^{-2}month^{-1} (D).

Compound		Kotinen	Hietajärvi	Pesosjärvi	Vuoskojärvi
SO$_4$-S	C	−0.059***	−0.046***	−0.033**	−0.018*
	D	−4.6***	−2.6***	−1.7**	n.s.
NO$_3$-N	C	−0.011*	−0.015**	n.s.	n.s.
	D	−1.2***	−0.64*	n.s.	n.s.
NH$_4$-N	C	−0.035***	−0.020***	−0.009*	n.s.
	D	−2.2***	−0.98**	−0.40*	n.s.
H$^+$	C	−0.62**	−1.6**	−0.97*	−1.2*
	D	−110***	n.s.	−74*	n.s.
Na	C	n.s.	−0.003*	n.s.	n.s.
	D	−0.28**	−0.17**	−0.14**	n.s.
K	C	−0.003**	−0.003**	n.s.	n.s.
	D	−0.39**	−0.15**	n.s.	−0.029*
Ca	C	−0.018***	n.s.	−0.007*	−0.006*
	D	−1.4***	−0.48*	−0.19**	−0.10*
Mg	C	−0.002*	n.s.	n.s.	n.s.
	D	−0.14***	n.s.	n.s.	n.s.
rain	D	n.s.	n.s.	n.s.	n.s.

Significance levels: *= $p < 0.05$, **= $p < 0.01$, ***= $p < 0.001$, n.s. = not significant
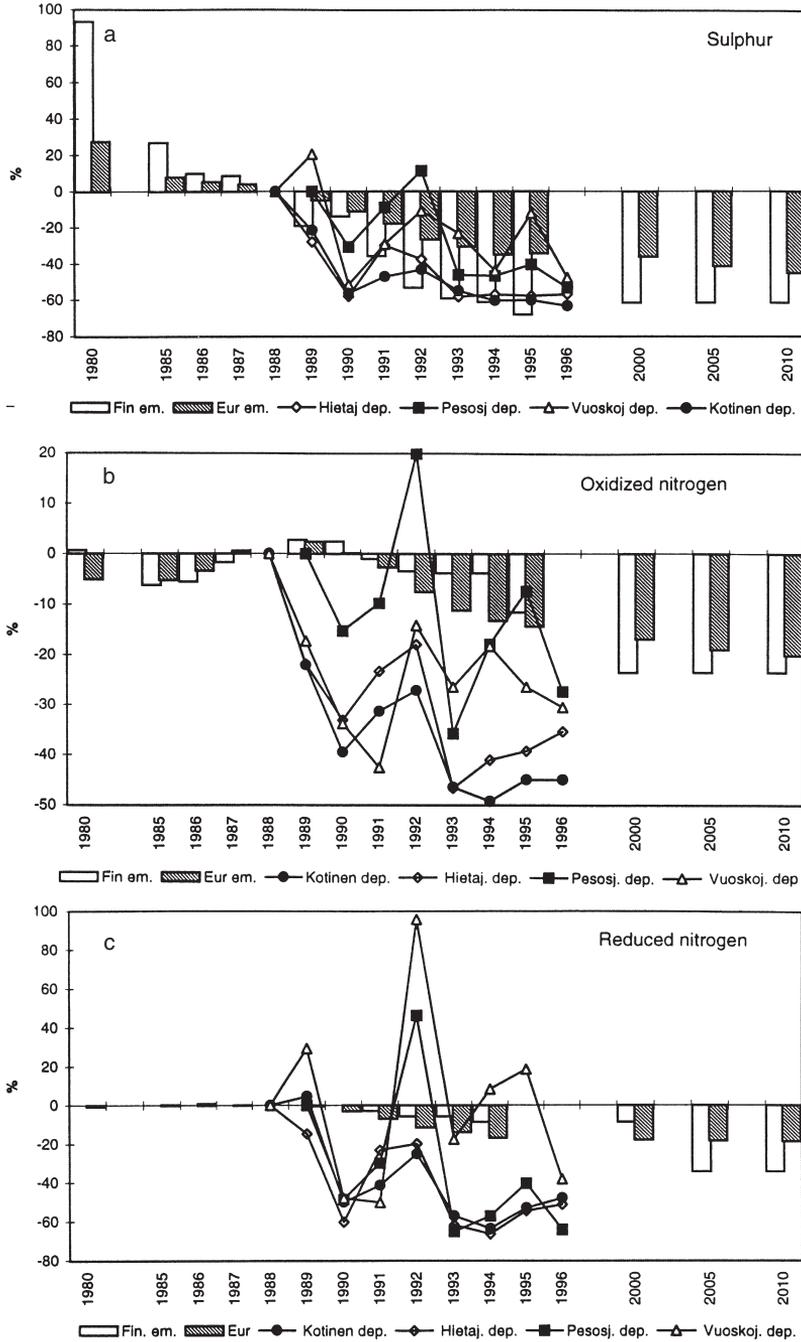
Fig. 3. Relative change compared to the 1988 data (1989 at Pesosjärvi) in the emission and deposition of (a) sulphur, (b) oxidised nitrogen and (c) reduced nitrogen. The emissions and scenarios are taken from Berge (1997a).

creased only slightly. A weak downward trend was observed in the annual sulphate and nitrate concentrations in precipitation at two further German stations and most of the nine other EMEP stations, while some stations even showed a slight increase in wet deposition. The western Meteoro-

logical Synthesizing Centre of the EMEP programme has compared the trends of modelled and observed sulphur (SO$_2$ and SO$_4$) concentrations across the whole Europe (Barret and Berge 1996). According to these comparisons the trends follow each other very well. However, the observed

decrease in sulphur dioxide concentrations between 1985–1994 is somewhat faster than the decrease in the modelled concentration values.

When the changes in the annual bulk deposition of sulphur and nitrogen compounds at the Finnish IM stations are compared with the corresponding emission reductions, the spatial differences between the stations again increase. In Fig. 3a–c the relative emission (Berge 1997a) and deposition values are given in per cent of the 1988 level (1989 at Hietajärvi). The reduction in sulphur bulk deposition at the two southern stations — Kotinen and Hietajärvi — very well reflects the decline of sulphur emissions (Table 1) in Finland and Europe, whereas in the northern part of the country the decrease in sulphur deposition fluctuates more independently of the emission levels (Fig. 3a). One possible explanation for this is that the northern parts of Finland are more influenced by European emissions; the distance to the northern stations is longer and annual variations in meteorology, e.g. wind direction and distribution, have a strong effect on the long-distance transport of pollutants.

Similar comparisons for the deposition of nitrogen compounds are presented in Fig. 3b and c. The decrease in bulk deposition of nitrate and ammonium does not match the corresponding change in emissions (Table 1) in any part of the country. The decline in emissions of 10%–20% is far less than the reduction in bulk deposition, which appears to settle at about 30%–50% of the base value. A possible change in the ratio between the dry and wet deposition of nitrogen compounds might explain part of the difference. While trend calculations in this paper are based only on bulk deposition measurements, the proportion of dry deposition and the changes in it remained unexplained. Air chemistry measurements have recently been included in the monitoring programme at the IM stations, and this will permit more precise deposition calculations in the future. Insufficient base cation emission data were available for a comparison with changes in deposition.

Mesoscale emission patterns — site analysis

All the Finnish IM stations are located in nature conservation areas with no nearby pollution sources. However, especially at Kotinen and Hietajärvi, there are sources within a distance of some ten to one hundred kilometres. These sources may have an influence on the observed pollution concentrations and deposition trends. The possible influence of these sources is analysed here, but is restricted to sulphur and nitrogen oxides. The emissions of ammonia in Finland are concentrated in agricultural areas, i.e. in the south-western parts of the country and in the Ostrobothnia region in western Finland. None of the IM stations are located in these areas, and the ammonia emissions close to the stations are very low.

Kotinen

On the mesoscale distance, here defined as distances ranging from tens of kilometres to about one hundred, there are large air pollution emission sources to the north-west of Kotinen. The paper and pulp industry town of Valkeakoski is located 50 km from Kotinen (for emissions *see* Table 4). Tampere, and the neighbouring industrial town of Nokia, are located at a distance of 70 km in the same direction. The town of Lahti is located at a distance of 40 km to the south-east. To the south of Kotinen is the Helsinki Metropolitan area and the Kilpilahti oil industries. The

Table 4. Emissions (t/a) of SO_2 and NO_x in 1995 from major sources at mesoscale distances from the Kotinen IM station. Met. area = Metropolitan area.

Source area	Lahti	Valkeakoski	Tampere	Nokia	Helsinki Met. area	Kilpilahti
Distance km	40	50	70	80	100	100
SO_2 emission	950	600	1 000	500	8 600	5 000
NO_x emission (as NO_2)	3 200	1 200	4 025	1 300	25 000	4 000

distance to both these source areas is approximately 100 km.

The influence of traffic emissions along a road (approx. 1 300 vehicles per day as the yearly average, and 250 vehicles per hour at the maximum) located 1.9 km away, is marginal (maximum of 7.5 µg m^{-3} for NO$_x$ in air, and a corresponding average of 0.2 µg m^{-3}, estimated using the Finnish CAR model, Härkönen *et al.* 1995) compared with the contribution from emission sources at mesoscale and longer distances.

Hietajärvi

There are no nearby emission sources at Hietajärvi. The closest emission sources of any significance are the pulp mills at Uimaharju (SO$_2$ emissions 110 t and NO$_x$ emissions 910 t in 1995) located 36 km to the south-west. The medium-sized town of Joensuu (SO$_2$ emissions 1 250 t and NO$_x$ emissions 1 600 t in 1995) is located in the same direction at a distance of 80 km. The Russian border is located 20 km to the north-east of the station, and there are no known emission sources at mesoscale distances on the Russian side.

Pesosjärvi

This station is located in the Oulanka National Park, i.e. in an area which, together with north-western Lapland, are the cleanest regions in Finland. The nearest urban areas are Kuusamo 40 km to the south-west, and Kemijärvi 80 km to the north-west. Both are small towns, with no air pollution emitting industries of any significance. There is no road traffic in the immediate vicinity of the IM station. As far as is known there are no pollution sources on the Russian side of the border, which is located less than ten kilometres to the east of the station.

Vuoskojärvi

The station is located in the northernmost corner of Finnish Lapland. There is a main road (170 cars per day on the average, and 25 cars per hour at the maximum) 3.5 km to the east of the station.

There are no air pollution emission sources, apart from the rather small amount of traffic, in the vicinity of the station. However, there are very high emissions, e.g. 130 000 t a^{-1} for sulphur in 1994 (Reimann *et al.* 1997), from the Russian metallurgical complex at Nikel, located no more than 120 km to the east. With easterly winds the air pollution plumes temporarily pass over the Vuoskojärvi station, resulting in high air pollution concentrations and corresponding high dry deposition. However, air concentration measurements by which this fact could be confirmed for Vuoskojärvi, are available only from the last year.

Observed trends versus the modelling results

Model description

In order to evaluate the extent to which long-term changes in deposition at the IM stations are caused by domestic emission reductions and the extent to which they are due to reduction measures abroad, deposition in the IM catchments was also estimated by the operational deposition model DAIQUIRI. DAIQUIRI has been developed at the Finnish Environmental Institute in collaboration with the Finnish Meteorological Institute (Syri *et al.* 1998), and its construction enables determination of the contributions of individual emission sources or regions to total deposition.

DAIQUIRI employs a transfer matrix concept, in which annual transport has been calculated using more detailed models and the results expressed as two-dimensional matrices, each matrix cell describing the annual amount of deposition caused by a unit source located in the centre-point of the matrix. These matrices are then used to depict the deposition fields caused by all the individual emission sources, and the total deposition is obtained as the sum of all the deposition fields caused by individual emission sources within the calculation domain and the long-range transported deposition.

The mesoscale sulphur transport matrices represent ten year-averaged weather conditions, and separate matrix sets have been generated for four climatological regions in Finland. The sulphur transport matrix scheme is described in more de-

tail in Johansson *et al.* (1990) and the nitrogen transfer matrix system in Syri *et al.* (1998). For nitrogen compounds, average transport in 1990 and 1993 was used, since transport matrices for other years are not yet available. The transport matrices of sulphur and nitrogen have a resolution of about 14 km and 30 km, respectively. The annual sulphur, nitrogen oxides and ammonia transport matrices of the EMEP/MSC-W ROOT150 model have been linked to DAIQUIRI in order to obtain estimates of the deposition derived from outside the DAIQUIRI calculation region (Barrett and Berge 1996). In this study, the annual deposition values were calculated using the corresponding year's EMEP transport matrices. Neither emission data nor transport matrices for 1996 were available, so the model study extends only up until 1995. The European emissions of 1994 also had to be used for 1995.

Comparison of DAIQUIRI results with the measured deposition values

Annual deposition values based on measurements and on model runs were compared in order to evaluate their mutual correspondence, and thus to obtain an estimate of possible uncertainties in the results. The annual measured and modelled deposition of sulphur, nitrogen oxides and ammonia at the IM stations have been plotted in Fig. 4a–c. The modelled deposition values have been divided into the proportion originating from Finland and that from other areas. The modelled sulphur deposition originating from the Leningrad region, Republic of Karelia, the Murmansk region and Estonia (marked as Rus+Est), as calculated from the mesoscale transport matrices, is shown separately from deposition derived from other parts of Europe. In Fig. 4a–c the inattributable origin, as calculated with the EMEP model, has been included in the proportion for the rest of Europe. At the northernmost station, Vuoskojärvi, the deposition loads derived from the Murmansk area and other parts of Europe are dominant, and the modelled proportion of domestic sources in deposition is only a few percent, and is barely visible in the figures.

In general, the modelled deposition values are higher than the measured ones. This is in accordance with the fact that the modelled values represent total deposition, whereas the measured bulk deposition contains only a fraction of the dry deposition. The proportion of dry fraction in total deposition tends to increase northwards in Finland due to the lower annual precipitation. This can also be seen in Fig. 4a–c as an increase in the difference between the modelled and measured deposition values on moving northwards. The Kotinen station, on the other hand, is located only about 100 km from the main NO_x emitter regions in Finland, and within this range dry deposition is the dominating NO_x deposition mode in the mesoscale transfer matrices. This effect is clearly evident in Fig. 4b, where the measured NO_x deposition at Kotinen is relatively smaller in relation to the modelled total deposition at Hietajärvi, which is located several hundred kilometres from any major NO_x emitter region.

For ammonium, the correlation between the measured and modelled annual deposition values was the poorest. Ammonium is deposited more locally than other acidifying compounds, and the deposition of ammonium is therefore more dependent on local changes in emissions and weather conditions. This may explain the large inter-annual variation observed in measured deposition not captured with DAIQUIRI. Yearly changes in the emission distribution were not available for 1987–1989 or for 1991–1994.

In an ideal situation, the annual modelled deposition at a specific location would be higher than the measured deposition by the amount of dry deposition not included in the measurements. The variation in the difference between the measurements and model results indicates the co-impact of different error components. First of all, the modelled deposition estimates in this study are not intended to accurately describe the annual deposition received at the catchments, but rather the average long-term changes due to emission reductions. This is because of the lack of annual transport data for Finland and incomplete emission inventories for all the years of the study period. Therefore, the modelled deposition values in this study only take into account annual meteorological changes in the deposition derived from abroad, as estimated from the EMEP matrices, and the domestic proportion of modelled deposition does not include the effect of annual meteorological changes or changes in the emission distribution. Secondly, measurements are often subject to local, small-scale conditions, such as wind
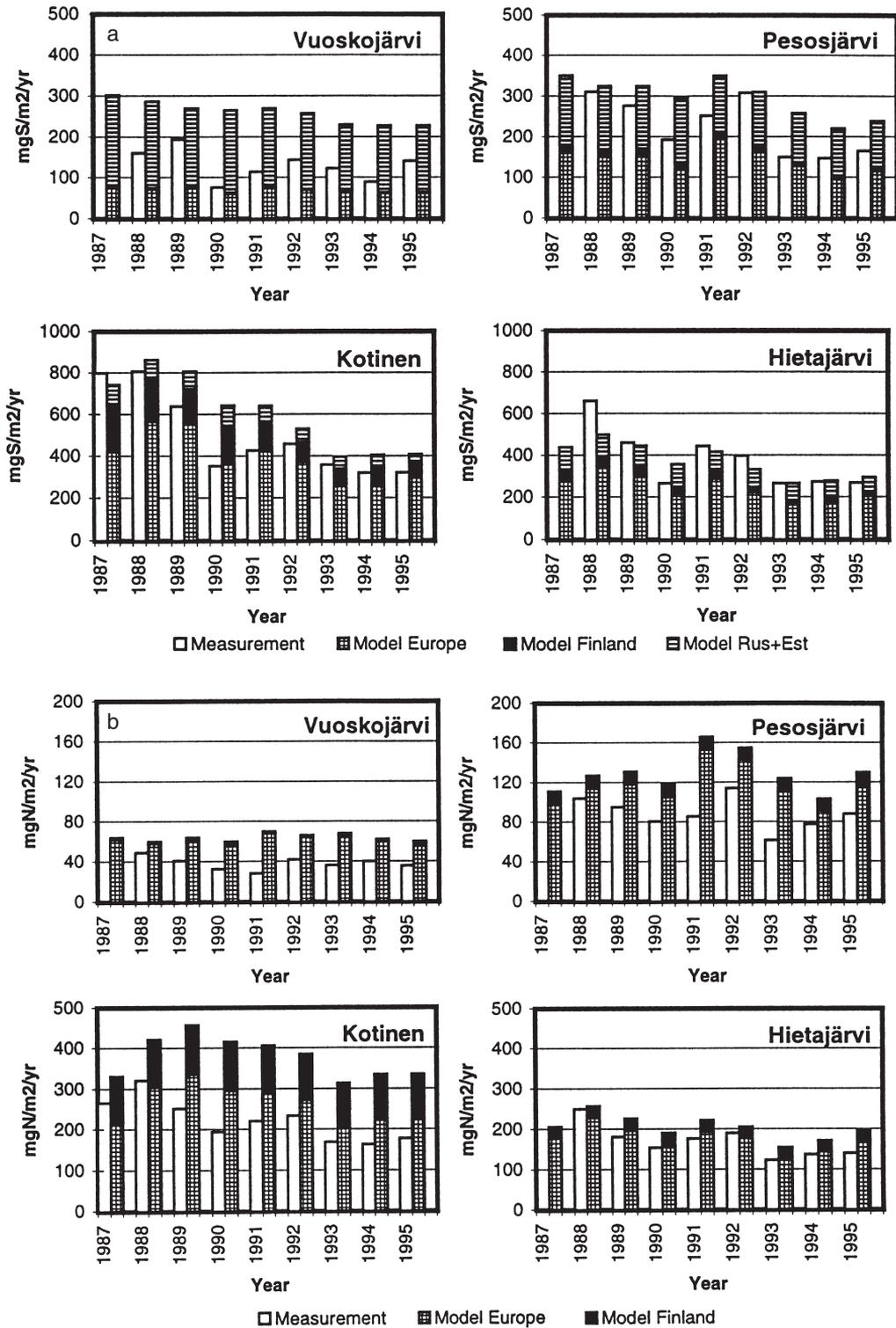
Fig. 4a and b. The modelled and measured annual deposition of (a) sulphate and (b) nitrate at the monitoring stations during 1987–1995. Model stands for modelled contribution from emission in the respective area.
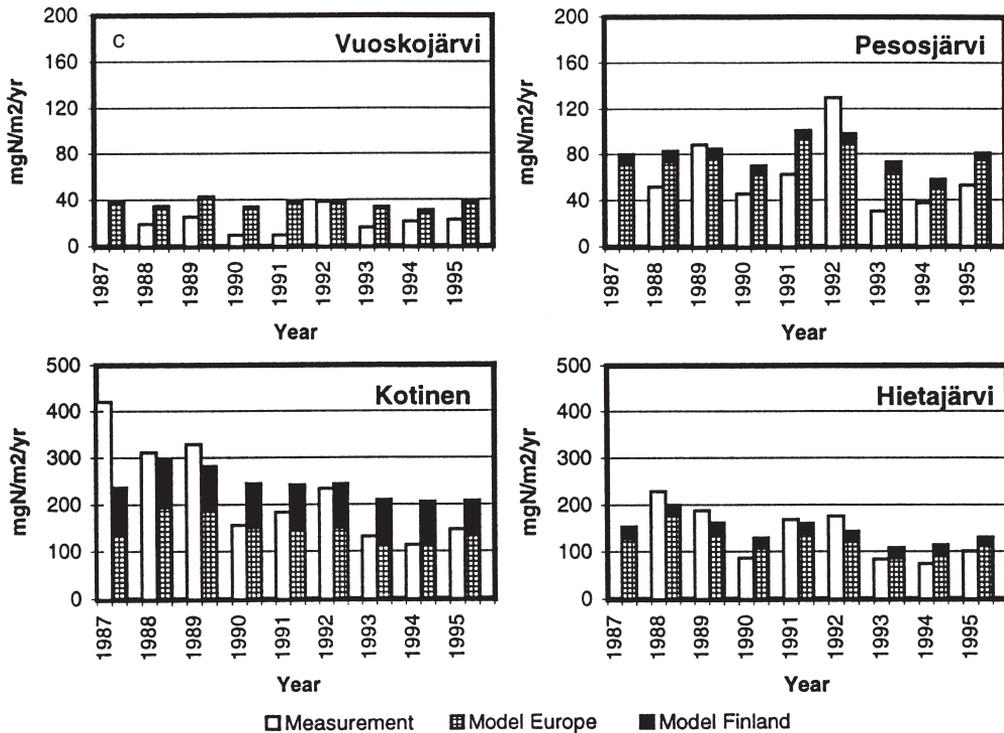
Fig. 4c. The modelled and measured annual deposition of ammonium at the monitoring stations during 1987–1995. Model stands for modelled contribution from emission in the respective area.

fields, nearby emission sources or local patterns of annual rainfall, which cannot be captured with models of this resolution and time scale. Thirdly, possible measurement errors also contribute to the total deviation observed.

Model calculations of the deposition origin at the northernmost station have more uncertainty than calculations at the southern stations. This is due to several reasons. Firstly, transport of pollution from the large sulphur emission sources in the Kola peninsula is an episodic phenomenon, since the prevailing winds are in the opposite direction. Therefore, annual total deposition received at the station may differ considerably from year to year. Since dry deposition is dominating in northern Lapland (Laurila and Tuovinen 1991), the bulk deposition measurements are not sufficient for estimating the total annual deposition received. Secondly, the deposition levels are so low that the inattributable deposition in the EMEP transfer matrices used becomes significant. In northernmost Finland it is of the order of ten per cent of the total calculated deposition.

Deposition at Kotinen and Hietajärvi in 1988

can be used to demonstrate errors caused by local meteorological conditions not captured by DAIQUIRI. The measured deposition values at Kotinen and Hietajärvi in 1988 were considerably higher compared to the modelled values than in other years. This may be partly explained by the higher precipitation in 1988 than the average precipitation for 1985–1995, which was used in the DAIQUIRI model. The amount of precipitation is also specific for the station in relation to its surroundings. This results in a site-specific deposition value at the station, which cannot be reproduced by the DAIQUIRI model. Table 5 shows the annual precipitation at the IM stations for the period 1988–1995 compared to the average precipitation in the surroundings.

Origin of acidifying deposition during 1987–1995

For sulphur, both the measurements and the model results indicate a clear decreasing trend in deposition. In northernmost Finland, domestic emissions

seem to have little influence on the deposition level, whereas in southern Finland the dramatic decline in Finnish emissions (from 328 to 96 kt SO_2 in 1987–95) has significantly contributed to the drop in the total deposition level. At Kotinen, the share of deposition of domestic origin, as estimated with DAIQUIRI, has dropped from 230 mg (S) m^{-2} a^{-1} to 70 mg (S) m^{-2} in year. During the same period deposition derived from sources outside Finland has dropped from 510 mg (S) m^{-2} in year to 340 mg (S) m^{-2} in year. At the northern stations Pesosjärvi and Vuoskojärvi, on the other hand, model calculations indicate that more than 90% of the sulphur deposition originates from outside Finland. At Vuoskojärvi, the decrease in deposition is partly due to the decline in sulphur emissions in the Kola Peninsula. During the period studied, the Kola Peninsula SO_2 emissions have declined from approximately 700 000 t in 1987 to about 350 000 t in 1994 in the Murmansk oblast (Kulmala 1987, Reiman *et al.* 1997). However, the decrease in deposition levels is, even at Vuoskojärvi, partly due to the overall decline in sulphur emissions. According to the EMEP MSC-W model calculations (Berge 1997b), the average contribution of the Kola Peninsula emissions to sulphur deposition in Finnish Lapland is between 55 and 80 percent. At Pesosjärvi, the main sources of sulphur deposition are emissions from the Murmansk region and the Kostamus metallurgical complex in Karelia, located less than 200 km

to the south-east of the station. The contribution of the rest of Europe to the total sulphur load is similar to that of the neighbouring Russian sources, and the modelled share of domestic origin has decreased from 20 mg (S) m^{-2} in year to only 7 mg (S) m^{-2} in year.

Emissions of nitrogen oxides both in Finland and abroad have not decreased as dramatically as sulphur emissions. Therefore, only a slightly decreasing trend in NO_x deposition was observed at the IM stations. This is mainly due to international emission reductions, because NO_x deposition in Finland is dominated by imports from abroad. At Kotinen the NO_x deposition of domestic origin is in the range of 30%, and its share out of total deposition decreases on moving northwards: at Vuoskojärvi, the modelled oxidised nitrogen deposition of domestic origin is only a couple of percent. Total European NO_x emissions have decreased by 13% during 1987–1994 (Barrett and Berge 1996). Finnish emissions have decreased by 10% from 1987 to 1995 (288 kt NO_2 to 259 kt NO_2), which is also reflected in the modelled deposition of domestic origin at the IM sites.

There have been no major reductions in ammonia emissions in Finland or elsewhere in Europe during 1987–1995. Deposition of domestic origin and that derived from abroad have, according to DAIQUIRI, been reduced somewhat during the study period; total modelled deposition at the IM sites has decreased by 10% to 20%. As is the case for oxidised nitrogen, the proportion of

Table 5. Annual precipitation (mm) at the IM stations during 1988–1995 and the means of precipitation amounts, (Mean surr.), at precipitation stations within 50 km distance from the respective IM station. The number in parentheses indicate the number of stations in the surrounding area taken into account. Std. dev. surr is the standard deviation of the precipitation.

	1988	1989	1990	1991	1992	1993	1994	1995
Kotinen	849	652	667	617	676	587	574	645
Mean surr. (7)	734 (6)	591 (6)	656	614	669	560	594	643
Std. dev. surr. (7)	76 (6)	43 (6)	35	32	46	37	33	56
Hietajärvi	770	551	466	722	717	593	641	570
Mean surr. (5)	839 (4)	623 (4)	600 (4)	739	741	650	743	663
Std. dev. surr. (5)	60 (4)	48 (4)	70 (4)	74	45	56	67	75
Pesosjärvi		575	365	571	647	356	476	487
Mean surr. (4)	612	614	460	654	779	526	542	569
Std. dev. surr. (4)	8	19	33	24	33	46	40	30
Vuoskojärvi	381	402	219	378	462	301	299	431
Mean surr. (2)	409	522 (1)	295 (1)	448	592	406	353	544

ammonia of domestic origin out of total ammonia deposition decreases northwards, accounting for about one third of total deposition at Kotinen, and a only a few percent at Vuoskojärvi.

Conclusions

The site analysis of the four IM stations shows that their location enables reliable monitoring of background deposition levels, i.e. that possible changes at the stations are not due to changes in local emissions. Within mesoscale distance (tens of kilometres to one hundred) to Kotinen there are rather high air pollution emissions on the Finnish scale. However, Kotinen is representative of clean areas in southern Finland. The very large emissions from Kola Peninsula in Russia are located at a distance of about 120 km from Vuoskojärvi station. Thus, this station is sharply affected by these Russian emissions when the wind is from the east.

There are large regional variations in the deposition of acidifying compounds in Finland, the highest concentrations and deposition values having been measured at the IM stations in the southern part of the country. At Kotinen, about 100 km inland from the Gulf of Bothnia, the concentration and deposition of almost all the compounds measured in precipitation show a decreasing trend. The results from Kotinen partly fulfil the expectations of the IM programme in following the transport of acidifying components; the changes in sulphur concentration and bulk deposition seem to reflect rather well the changes in sulphur emissions in Finland and elsewhere in Europe. The monitoring results for nitrogen compounds in precipitation at Kotinen do not reflect the changes in emission levels as clearly as those for sulphur.

The results for Hietajärvi are very similar to those for Kotinen, but the trend in the concentration and deposition of acidifying compounds is weaker. This is understandable, because the effect of meteorology, wind direction especially, increases with increasing distance to the heavy emission sources in Finland and elsewhere in Europe: looking from the station, the sector in which the pollution sources are located is narrower. Despite this, however, Hietajärvi is still a good station to follow the effects of emission reductions.

The results from the two stations further to the north show that, despite the low initial deposition values and strong decrease in the emissions of acidifying components, there are hardly any signs of a declining trend, and critical loads are still being exceeded.

As the trend calculations in this paper were based on only bulk deposition measurements, a possible change in the ratio between the dry and wet deposition of the compounds might explain part of the different behaviour of the emissions and deposition.

The use of the DAIQUIRI operational model in this study has provided complementary information about the extent to which emission reduction measures in Finland on the one hand, and those elsewhere in Europe on the other hand, have caused the observed long-term decreasing trends in acidifying deposition at the Finnish IM sites. Due to the use of averaged long-term transport data in the deposition model and partly uncertain emission data, the calculated estimates of the origin of annual deposition may have large deviations from the actual total deposition, as well as from the measurements. The calculated long-term deposition changes, however, indicate similar trends as the measurements.

According to both the measurements and model calculations, there has been a considerable reduction in sulphur deposition, especially at the stations in southern and central Finland. In the south, domestic emission reductions have significantly contributed to a reduction in total deposition. The model calculations indicate that sulphur deposition in the northernmost parts of Finland would be strongly influenced by imports from Russia, and that the long-term decrease in deposition would to a large extent be caused by the decline in emissions from these areas.

The model calculations indicate only a slight decrease in the deposition of nitrogen compounds during the study period. At Kotinen and Hietajärvi, deposition of domestic origin is marked, whereas the contribution of Finnish origin in modelled nitrogen deposition is, especially at the Vuoskojärvi station, only a couple of percent. During the study period there were no major reductions in Finnish emissions of nitrogen compounds. The decreasing trend in nitrogen deposition seems to be mainly caused by international emission reductions.

A further reduction in the deposition of acidifying compounds is needed for Finland, because the critical loads of acidity are still being exceeded in many parts of the country (Posch *et al.* 1997).

This analysis suggests that domestic measures have only a limited reduction potential, especially in northern Finland. On the other hand, most of the areas where the critical loads are exceeded are situated in southern Finland, where both the total deposition loads of all acidifying compounds and the domestic shares in total deposition are at their highest. In this region, properly allocated further domestic reduction measures might be a cost-effective means of preventing the exceedance of critical loads.

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Appendix. Precipitation-weighted annual mean concentrations from 1988 to 1996 at the Finnish IM stations.

	1988	1989	1990	1991	1992	1993	1994	1995	1996
Kotinen									
H^+ µmol/l	36	42	31	40	30	35	34	26	27
Cl^- mg/l	0.23	0.29	0.24	0.27	0.24	0.21	0.23	0.21	0.18
NO_3^- mg(N)/l	0.38	0.39	0.29	0.36	0.35	0.29	0.28	0.28	0.29
SO_4^{--} mg(S)/l	0.95	0.98	0.53	0.69	0.68	0.62	0.56	0.50	0.49
Mg^{++} mg/l	0.03	0.04	0.03	0.03	0.04	0.03	0.03	0.03	0.02
Ca^{++} mg/l	0.22	0.22	0.13	0.13	0.15	0.14	0.13	0.14	0.11
NH_4^+ mg(N)/l	0.37	0.50	0.23	0.30	0.35	0.23	0.20	0.23	0.27
Na^+ mg/l	0.12	0.15	0.19	0.16	0.16	0.15	0.14	0.13	0.10
K^+ mg/l	0.08	0.12	0.15	0.08	0.07	0.06	0.05	0.08	0.10
Rain mm	849	652	667	617	676	587	574	645	606
Hietajärvi									
H^+ µmol/l	35	40	37	34	29	27	30	29	27
Cl^- mg/l	0.16	0.23	0.24	0.16	0.13	0.14	0.12	0.16	0.20
NO_3^- mg(N)/l	0.30	0.33	0.33	0.24	0.26	0.21	0.21	0.25	0.23
SO_4^{--} mg(S)/l	0.83	0.84	0.57	0.62	0.56	0.45	0.43	0.47	0.42
Mg^{++} mg/l	0.02	0.03	0.02	0.02	0.02	0.02	0.02	0.02	0.02
Ca^{++} mg/l	0.14	0.20	0.11	0.10	0.10	0.10	0.08	0.10	0.09
NH_4^+ mg(N)/l	0.29	0.34	0.19	0.23	0.25	0.14	0.12	0.18	0.17
Na^+ mg/l	0.10	0.12	0.14	0.11	0.10	0.11	0.09	0.11	0.08
K^+ mg/l	0.06	0.08	0.07	0.05	0.05	0.05	0.04	0.05	0.05
Rain mm.	770	551	466	722	717	593	641	570	646
Pesosjärvi									
H^+ µmol/l		28	35	27	24	29	24	24	22
Cl^- mg/l		0.15	0.17	0.16	0.12	0.21	0.14	0.18	0.15
NO_3^- mg(N)/l		0.17	0.22	0.15	0.18	0.17	0.16	0.18	0.14
SO_4^{--} mg(S)/l		0.48	0.53	0.44	0.48	0.42	0.31	0.34	0.26
Mg^{++} mg/l		0.02	0.02	0.02	0.02	0.03	0.02	0.02	0.01
Ca^{++} mg/l		0.06	0.06	0.06	0.08	0.08	0.04	0.04	0.03
NH_4^+ mg(N)/l		0.15	0.13	0.11	0.20	0.09	0.08	0.11	0.06
Na^+ mg/l		0.08	0.13	0.10	0.09	0.14	0.08	0.11	0.06
K^+ mg/l		0.04	0.05	0.04	0.03	0.03	0.03	0.04	0.03
Rain mm.		575	365	571	647	356	476	487	502
Vuoskojärvi									
H^+ µmol/l	27	27	24	20	17	23	20	21	16
Cl^- mg/l	0.48	0.71	1.05	0.51	0.67	1.31	0.63	0.66	1.21
NO_3^- mg(N)/l	0.13	0.10	0.15	0.07	0.09	0.12	0.13	0.08	0.11
SO_4^{--} mg(S)/l	0.42	0.48	0.35	0.30	0.31	0.41	0.30	0.33	0.27
Mg^{++} mg/l	0.03	0.05	0.08	0.04	0.05	0.10	0.05	0.05	0.08
Ca^{++} mg/l	0.05	0.09	0.10	0.05	0.05	0.08	0.06	0.04	0.05
NH_4^+ mg(N)/l	0.05	0.06	0.04	0.03	0.08	0.05	0.07	0.05	0.04
Na^+ mg/l	0.29	0.40	0.63	0.31	0.41	0.81	0.38	0.37	0.67
K^+ mg/l	0.05	0.04	0.07	0.04	0.04	0.05	0.04	0.04	0.06
Rain mm.	381	402	219	378	462	301	299	431	314