

**Estonian Environmental Research Centre**



## **Assessment of the Estonian EMEP data**

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## 1. Emissions

**Local.** The Estonian Air Emissions database has been maintained since 1976. At the beginning, the database included data only for 5 largest Estonian towns: Tallinn, Tartu, Pärnu, Narva and Kohtla-Järve. Later on some industrial counties were added and from 1990 the inventory has been covering the whole Estonian territory. At present, some data are being recalculated for 1980 and 1985.

The main branch of industry in Estonia is energy production. Approximately 89% of energy in Estonia is produced by combustion of fossil fuels. The remaining 11% comes from biomass.

The energy sector, as well as chemical industry, is based on oil shale. In 1999, 92% of electricity was generated from oil shale, a fuel specific for Estonia, characterized by a high ash content (45-50%), moderate sulphur content (1.4-1.8%) and low net caloric value. Oil shale mining and combustion, accounting for about 81% of the total harmful emissions of Estonia has put a severe load on the environment.

In Estonia, SO<sub>2</sub> emissions have declined by ~67 % from their peak in the 1980s to the 2000 (Fig. 1a). A rapid decrease took place in the early 90's. This was caused by the reshaping of local economy, resulting in a gradual decline in the oil shale amounts used annually by the thermal power plants.

The emissions of nitrogen oxides have declined by ~40 % compared to the 90's. In the middle of the 90's NO<sub>x</sub> emissions slightly increased in Estonia (Fig. 1b), whereas the total number of motor vehicles has increased about twice. The average age of cars in Estonia - 13 years - is higher than in EU countries, which is also reflected in higher emission rates of cars. At the same time, the share of public transport in the passenger traffic volume decreased more than 2 times between 1990 and 1999.

The main source of ammonia emission in Estonia is agriculture: manure management and fertilizer consumption. The emission of ammonia has reduced by 68% since 1980 due to the decrease in livestock numbers (especially dairy cows and cattle as the most significant sources of NH<sub>3</sub> emission) and reduced use of mineral fertilizers.

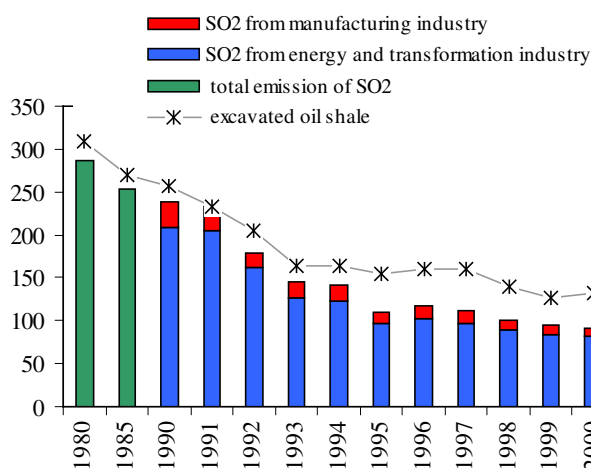


Figure 1a. Emissions of SO<sub>2</sub> (kilotons, left scale) and supply of oil shale (megatons, right scale) in Estonia.

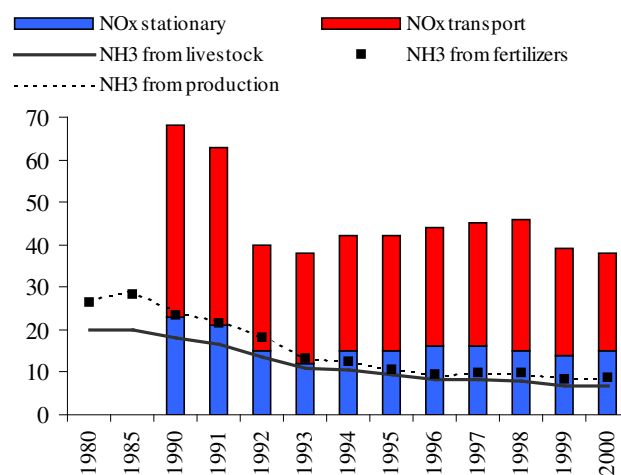


Figure 1b. Emissions of NO<sub>x</sub> and NH<sub>3</sub> (kilotons) in Estonia.

**Transboundary fluxes.** The EMEP database of source-receptor matrices for 1996 and 1998, and the paper EMEP/ MSC-W (2000) shows that polluted air was mostly imported to Estonia from Western and Central Europe and neighboring countries, considerable part of pollution originates in international shipping. Most countries have reduced their emissions of SO<sub>2</sub> and NO<sub>x</sub> during the 90s. Thus if patterns of transport of air masses were throughout the study period similar to those in 1996 and 1998, the import of pollution to Estonia has reduced during the considered period.

## 2. Monitoring stations and data quality

In Estonia, the first systematic measurements in background stations begun on the islands lying in western Estonia (Sõrve and Vilsandi stations) in the 80s and soon after in the Lahemaa National Park (Fig. 2). At the beginning (1985-1991), Estonian stations were operated by the Hydrometeorological Institute using measuring equipment and methods of the former USSR. Samples were analyzed in different laboratories and unfortunately differences in test results raise doubts concerning reliability of data. There are also many gaps in data series.

After gaining independence in 1991, environmental monitoring underwent reorganization. This also explains the existence of gaps in data for the period 1992-1993. The collection of data on the state of the environment has been budgetary since 1994. The Estonian Environmental Research Center (EERC) has been operating the monitoring stations from the same year. The CLRTAP and its first protocols were signed by Estonia as late as in 2000 and the EMEP financing protocol in 2001.

The longest available data series exist for Lahemaa (SU09/EE09) station (Fig. 2). The station at Sõrve (SU02) operated shortly until 1991. The station located on the island of Vilsandi (SU11/EE11) provided data occasionally in 1989-1990. Systematic data submission has taken place since 1994 after installation of continuous analyzers.

Improvements have been made in data quality for precipitation components, which reach now the level A and B for most of compounds (EMEP, 2003). EERC was accredited internationally for the precipitation analysis in 1998 and for air quality measurements in 2000.

For statistical analysis air quality data series with more than 55% data coverage round the year were used.

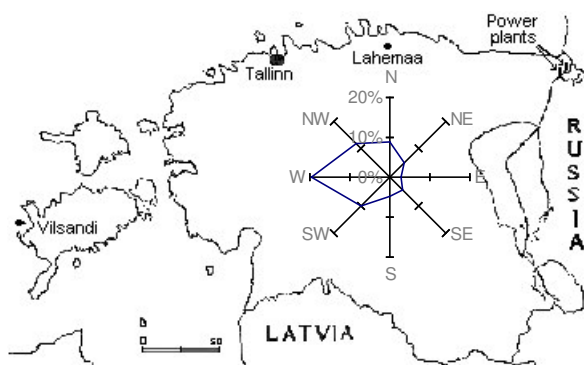


Figure 2. Location of stations and the main polluters in the area. Average frequency (%) of air mass transport sectors at Estonian EMEP stations is given.

Table 1. Measurement program.

Medium	Measurement frequency	Components
Air	24 h	SO <sub>2</sub> , NO <sub>2</sub> , SO <sub>4</sub> (EE09)
	1 h	SO <sub>2</sub> , NO <sub>2</sub> (EE11)
	1 h	O <sub>3</sub>
Precipitation	Weekly	SO <sub>4</sub> <sup>2-</sup> , NO <sub>3</sub> <sup>-</sup> , Cl <sup>-</sup> , NH <sub>4</sub> <sup>+</sup> , base cations pH, conductivity
	Monthly	As, Cd, Cu, Hg, Pb, Zn

### 3. Air Quality

#### 3.1. Sulphur dioxide

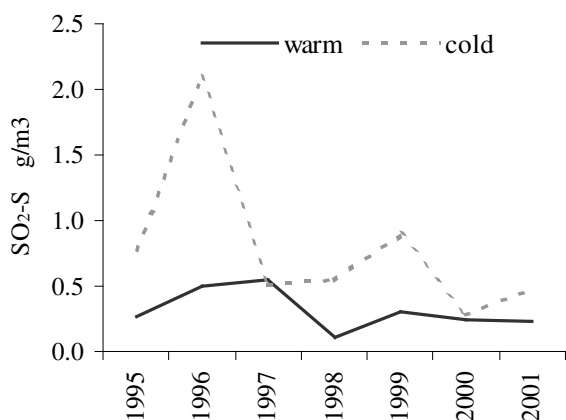


Figure 3. Mean annual concentrations of SO<sub>2</sub>-S at Vilsandi in the cold and warm period 1995-2001.

The average SO<sub>2</sub>-S concentration during last three years (1999-2001) has been 0.4 μg/m<sup>3</sup> at Vilsandi and 0.7 μg/m<sup>3</sup> at Lahemaa station. The winter concentrations are generally higher than summer concentrations (Fig. 3). There is no statistically significant downward trend in air SO<sub>2</sub> concentrations during the assessment period. Nevertheless episodes with high concentrations were measured more frequently at the beginning of measuring period. It could be suggested that a rapid decline in the sulfur dioxide concentration in Estonia has occurred during the first half of 90s (Fig. 1a) before the beginning of measurements.

The origin of the elevated SO<sub>2</sub>-S concentrations were found by using the daily air mass trajectories obtained via the EMEP Lagrangian model (EMEP/MSC-W, 2003) using the sector template prepared by Finnish Meteorological Institute for the EMEP Assessment work (EMEP, 2003).

The direction of air masses with higher SO<sub>2</sub> concentrations are related to the location of measurement stations. At the western border of Estonia (Vilsandi station) the higher concentrations are measured from southern and southwestern air mass transport directions (Fig. 4a). At Lahemaa high concentrations are observable from sectors between NE and SE (Fig. 4b) where big power plants are located. The low concentration air masses have arrived from north direction in both stations. The most frequent transport sectors at Estonian stations were western with average concentration of SO<sub>2</sub>-S 0.2 μg/m<sup>3</sup> (Vilsandi) and 0.4 μg/m<sup>3</sup> (Lahemaa).

It should be noted, that for 30 % of days the air mass transport sector was undetermined. Those days were randomly distributed throughout the year.

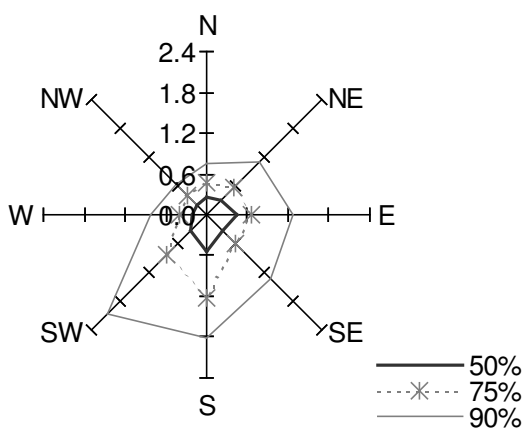


Figure 4a. SO<sub>2</sub> concentrations (μg S/m<sup>3</sup>) in air masses of different origin at the Vilsandi station 1994-2001. Measured by UV-fluorescence.

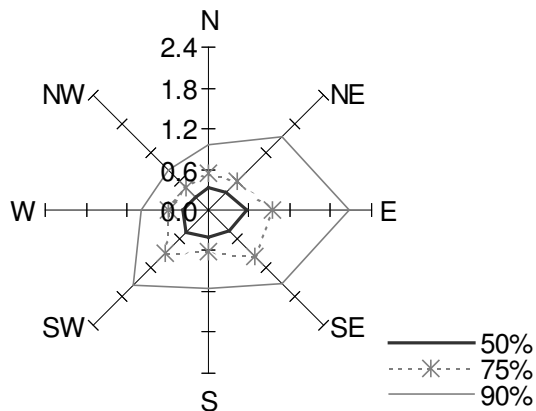


Figure 4b. SO<sub>2</sub> concentrations (μg S/m<sup>3</sup>) in air masses of different origin at Lahemaa station 1994-2001. Measured by filters, analyzed by ion chromatography.

### 3.2. Nitrogen dioxide

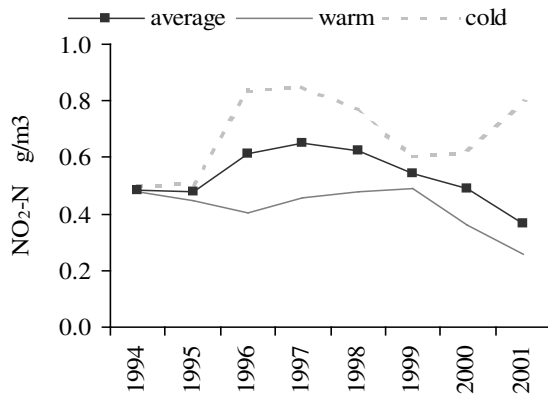


Figure 5. Mean annual concentrations of NO<sub>2</sub>-N at Lahemaa in the cold and warm period 1995-2001.

The average NO<sub>2</sub>-N concentration during last three years (1999-2001) has been 0.96 µg/m<sup>3</sup> at Vilsandi and 0.47 µg/m<sup>3</sup> at Lahemaa station. Since the beginning of measurements the yearly concentrations have not fluctuated much (Fig 5). Interestingly, concentrations of NO<sub>2</sub>-N at Vilsandi are about twice as high as at Lahemaa which is opposite to SO<sub>2</sub>-S. The highest concentrations were measured in 1997 in occurrence of air mass transport from S and SW sectors. Relatively higher NO<sub>2</sub> concentrations from same direction were registered in Latvian station Rucava too.

The concentration rose of NO<sub>2</sub> is almost round at Lahemaa station (Fig 6b) indicating probably local emission sources. The highest 10 % of NO<sub>2</sub> concentrations were measured from eastern direction and the lowest from near-by NE direction. At the western border of Estonia (Vilsandi station) all concentration roses are clearly stretched out in SW direction (Fig 6a). The concentrations from W and S directions are also elevated and all together almost 35% of time air masses from these directions are prevailing. Nevertheless decreasing tendency for average NO<sub>2</sub> concentrations from SW may be assumed. The relatively lower nitrogen dioxide concentrations were measured when the air masses originated from NE and E directions.

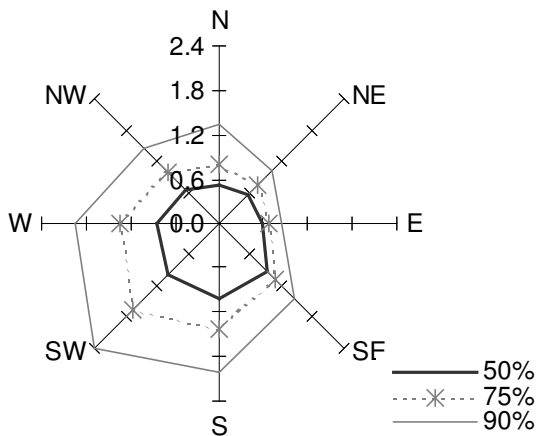


Figure 6a. NO<sub>2</sub> concentrations (µg N/m<sup>3</sup>) in air masses of different origin at Vilsandi station 1994-2001. NO<sub>2</sub> have measured by chemiluminescence analyzer.

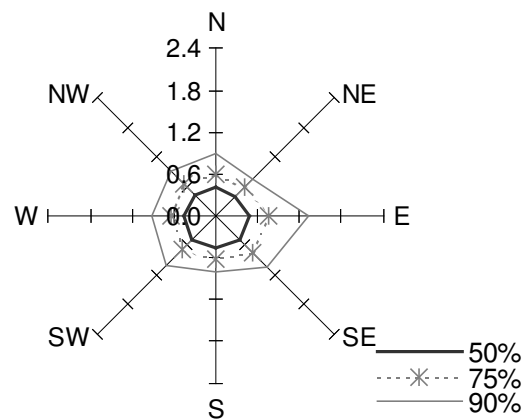


Figure 6b. NO<sub>2</sub> concentrations (µg N/m<sup>3</sup>) in air masses of different origin at Lahemaa station 1994-2001. NO<sub>2</sub> have measured by absorbing tubes

### 3.3. Ground level ozone

The annual average concentrations of ozone are significantly higher near the seaside at Vilsandi station (67-79  $\mu\text{g}/\text{m}^3$ ) and remain below the 60  $\mu\text{g}/\text{m}^3$  at Lahemaa station. In the latter case diurnal and seasonal cycles of ozone concentrations are more pronounced but the period with elevated concentrations during the year is shorter. The mean concentration roses of ozone in both stations are almost rounded (Fig. 7). There have been no significant concentration trends during 1995-2001 (Fig. 8).

The five year average value of the AOT40 index for forests (April-September) has been below the critical level of 10 000 ppb h. The critical level for crops (3000 ppb h for May-July) have been exceeded at both stations in last 5 years.

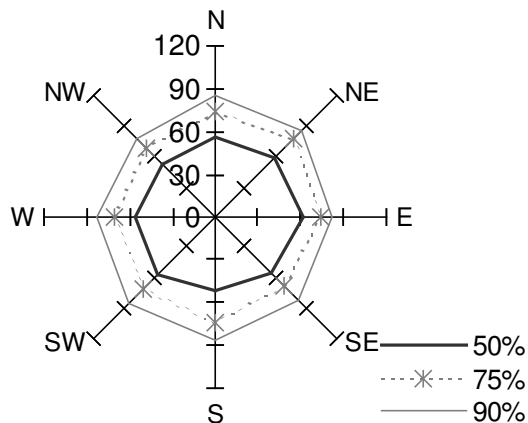


Figure 7. Distribution of daily ozone concentrations ( $\mu\text{g}/\text{m}^3$ ) during 1995-2001 at Lahemaa station

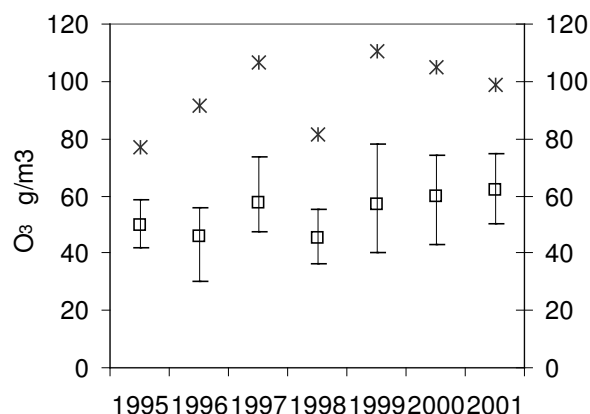


Figure 8. Annual quartiles (lines), medians (boxes) and 99-percentiles (crosses) of ozone ( $\mu\text{g}/\text{m}^3$ ) at Lahemaa station.

### 4. Precipitation quality

The annual values of main anions ( $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ,  $\text{Cl}^-$ ) and base cations ( $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{K}^+$ ,  $\text{Na}^+$ ) and pH in bulk precipitation were tested using the Mann-Kendall method and Excel templates prepared by the Finnish Meteorological Institute (Salmi et al, 2002). The results of statistical evaluation of trends (Tab. 2) confirms that downward trends are significant for sulphate and nitrate, less significant for chloride and not remarkable for other compounds.

Table 2. Significant downward trends in precipitation during the period of 1994-2000

	$\text{SO}_4\text{-S}$	$\text{NO}_3\text{-N}$	$\text{Cl}$	$\text{NH}_4\text{-N}$	$\text{Na}$	$\text{K}$
EE09-Lahemaa	*	**	*	+	*	+
EE11-Vilsandi	**	*	*	+		
** 0.01 level of trend significance	* 0.05 level of significance		+ 0.1 level of significance			

The mean annual concentrations of sulphate have fallen about two times during 1994-2001 and annual average concentrations are now 0.4-0.6 mgS/l. It should be mentioned that the seasonal differences between mean concentrations of cold and warm season has decreased as well. The corresponding deposition load of sulphur was approximately 300 mgS/m<sup>2</sup> per year (Fig. 9) .

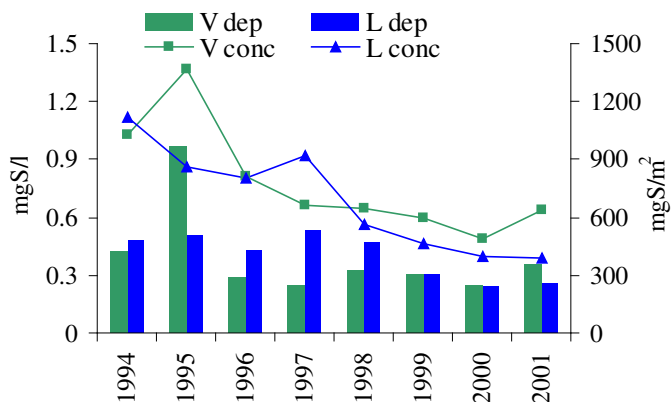


Figure 9. Annual mean concentrations (conc) and deposition (dep) of  $\text{SO}_4\text{-S}$  at Lahemaa (L) and Vilsandi (V).

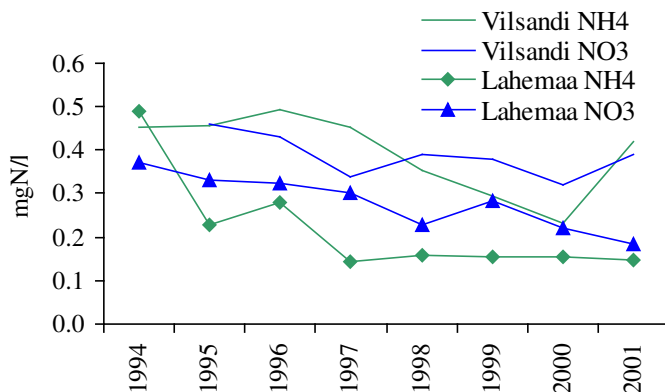


Figure 10. Annual mean concentration of N compounds in precipitations at Lahemaa and Vilsandi.

Although chloride is usually of marine origin, in our case chloride and sulphate anions are acidic components of flue gases of oil shale. Linear decline trends of  $\text{SO}_4^{2-}$  and  $\text{Cl}^-$  annual concentrations followed the same pattern as annual  $\text{SO}_2$  emission and annually combusted oil shale amounts. This trend is confirmed with data from national precipitation network.

The level of both nitrogen compounds ( $\text{NH}_4^+$  and  $\text{NO}_3^-$ ) in precipitation are higher in westerly located island of Vilsandi (Fig. 10). This is in accordance with the results of  $\text{NO}_2$  measurements. Mean annual nitrate concentration decrease has been steeper at Lahemaa station. The last 3-year average values were 0.23 and 0.36 mgN/l for Lahemaa and Vilsandi respectively. Only weak decreasing tendency can be seen in annual mean concentrations of ammonium. The average concentration values were 0.15 and 0.32 mgN/l for Lahemaa and Vilsandi respectively. The total annual wet deposition of nitrogen (nitrate and ammonium) was higher at Vilsandi (358 mgN/m<sup>2</sup>/year) than Lahemaa (247 mgN/m<sup>2</sup>/year).

The mean annual sum of base cations shows that concentrations have decreased though the trend is not linear (Fig. 11). It is complicated to distinguish between marine and industrial origin of base cations since all of them could be emitted in content of fly ash.

Generally the relatively high deposition of base cations neutralizes the acidity of precipitations in Estonia. At Vilsandi and Lahemaa the average annual pH values were rather acidic - 4.5 and 4.7 respectively. As a rule the higher pH values are measured during the warm period and acidic values during the cold period of year. Trend of pH values is presented on Figure 12. The mean annual pH at Lahemaa has slightly decreased, the decreasing trend is clear when only warm period is taken into account (from pH 5.3 in 1995 to pH 4.9 in 2001). At Vilsandi no pH trend is seen, but summer maximum values have decreased.

The precipitation amount at Vilsandi is one of the smallest in Estonia, the last 10 year average has been 565 mm. During the assessment period the yearly sum of precipitation has slightly increased (Fig. 13), mainly due to relatively warmer and precipitation rich winters. Since the data series are relatively short and precipitation amount has been variable, deposition trends were not analyzed.

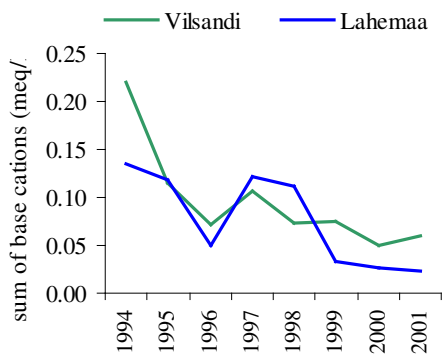


Figure 11. Annual average values of summed basic cations (meq/l) at Vilsandi and Lahemaa stations.

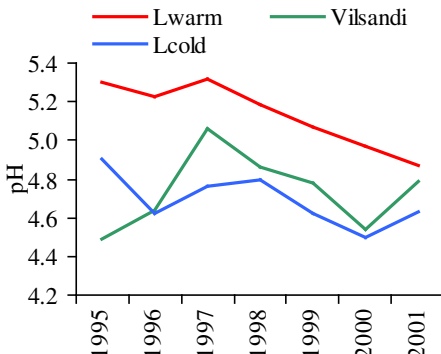


Figure 12. Weighted average values of pH: seasonal means at Lahemaa (Lcold, Lwarm) and annual means at Vilsandi.

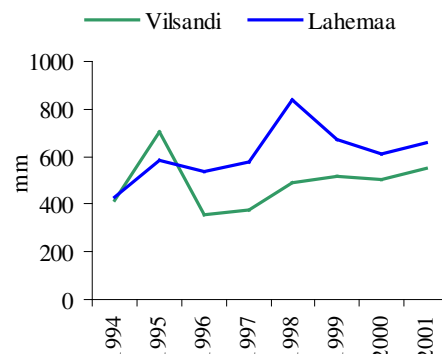


Figure 13. Variation of precipitation amounts (mm) at Lahemaa and Vilsandi.

## 5. Deposition and critical loads

According to data from local precipitation network deposition loads of N and S compounds vary significantly. The higher loads of sulphate occur in NE Estonia (Fig. 14), which used to be the result of the emissions from power plants. Load of nitrogen compounds (Fig. 15) are higher not only in NE Estonia but also in Southern and Western Estonia, which seems to be more associated with long range transport of pollution.

Earlier studies (Oja, 2000) reveal that critical loads for nutrient nitrogen and acidifying level of N+S can be reached in some sensitive ecosystems of Estonia. Comparing two different regions with deposition equal to critical loads it can be said that the NE Estonia is characterized by increased base cation deposition buffering fully the acidic deposition. In Southern Estonia the actual base cation deposition correspond to natural background level and deposition of acidifying compounds may reach or exceed the critical loads in some more sensitive areas.

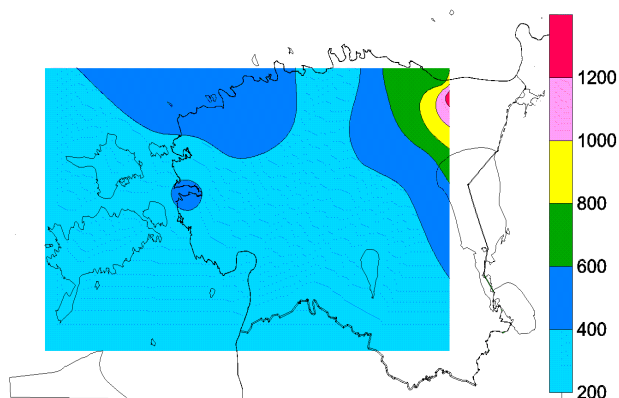


Figure 14. Mean annual sulphur ( $\text{mg S/m}^2 \text{ yr}$ ) deposition in Estonia (2000 – 2002 local precipitation network)

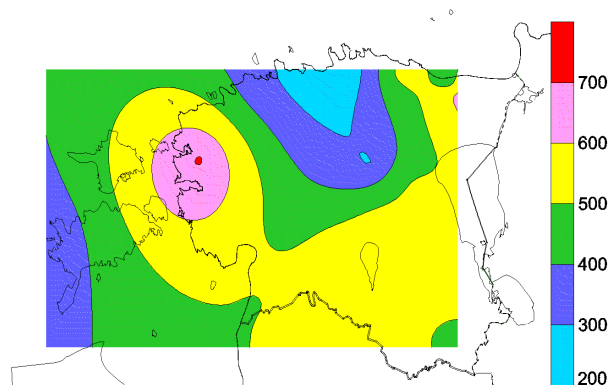


Figure 15. Mean annual nitrogen deposition (oxi+red,  $\text{mg N/m}^2 \text{ yr}$ ) in Estonia (2000 – 2002).

## 6. Conclusions

- There has been a considerable reduction of emissions during the last twenty years in Estonia. The reduction equals to about 60% for sulphur compounds, about 40% for NO<sub>x</sub> and about 70% for ammonia.
- In Estonia, the decline in emissions can be attributed to economic restructuring. The fall in emissions and incoming pollution was especially remarkable in the beginning of the 90's. Unfortunately, there are no proved data of air and precipitation quality.
- Sector analysis shows that air masses with polluted air are mostly transported to Estonia from Western and Southern directions. The location of the Vilsandi station at the western border of Estonia suits best to monitor the background loads of air pollution and deposition coming from long-range transported emissions. The Lahemaa monitoring station in Northern Estonia is quite distant from industrial district (120 km from Northeast Estonian oil-shale complex) and serves as a natural background area for the study of changes in our domestic and long-range transported air pollution and its impact to ecosystems.
- An analysis of surface ozone in relation to transport directions did not show any sector with elevated concentrations.
- In last three years the monthly concentrations of main anions and base cations in bulk precipitation have reached the lowest values of the measuring period.
- The decreasing concentrations of sulphate were connected to a decreasing trend of base cations, which in turn has not resulted in an increased pH level in precipitation. An opposite (decreasing) trend of pH was found at Lahemaa, which could be attributed to reduced emissions of alkaline dust from local power plants and a cement factory in the NE industrial complex. However, the downward trend of pH is normalizing the ecological situation in bogs of North-East Estonia where earlier peat growth was stopped due to alkalization.

## Acknowledgements

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