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LONG-TERM DEPOSITION PATTERNS OF AIRBORNE WASTES IN THE NORTH-EAST OF ESTONIA

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The deposition loads of fly ash and sulfur have been high in the NE Estonia since the late fifties, when the oil shale energetics, chemical and cement industry achieved the remarkable extent. The combined effects of both pollutants have seriously damaged sensitive ecosystems (forest on podzolic soils and bog). The main effects are related with alkalization due to accumulation of fly ash components and the Sphagnum growth inhibition due to sulfur load. These effects have the time scale of several years or even more.

The pollution loads have been changed during recent 40 years due to launching and reconstruction of enterprises (incl. purification systems) and variations of production capacity. First representative data on air pollution deposition originate from the middle of eighties. Only model estimations could be used to quantify the deposition fluxes before that time, as well as for assessing the future scenarios. The air pollution dispersion and deposition model AEROPOL is used for this purpose. The estimations are based on air pollution emissions per production unit and climatic average dispersion conditions. The model estimations are compared with available measured values. The deposition loads for a possible development in the future are estimated.

Introduction

The ecosystems in the North-East of Estonia have been seriously affected by airborne fly ash and sulfur dioxide for five decades due to oil shale combustion and oil shale based chemical industry. The first important pollution sources were the Kiviõli and the Kohtla-Järve power plants (PP-s) (launched in 1948 and 1949, respectively). During the fifties and the sixties several power plants and chemical factories began to operate. Among them Baltic (1960) and Estonian (1969) PP-s are most important, in respect of energetic capacity, as well as air pollution.

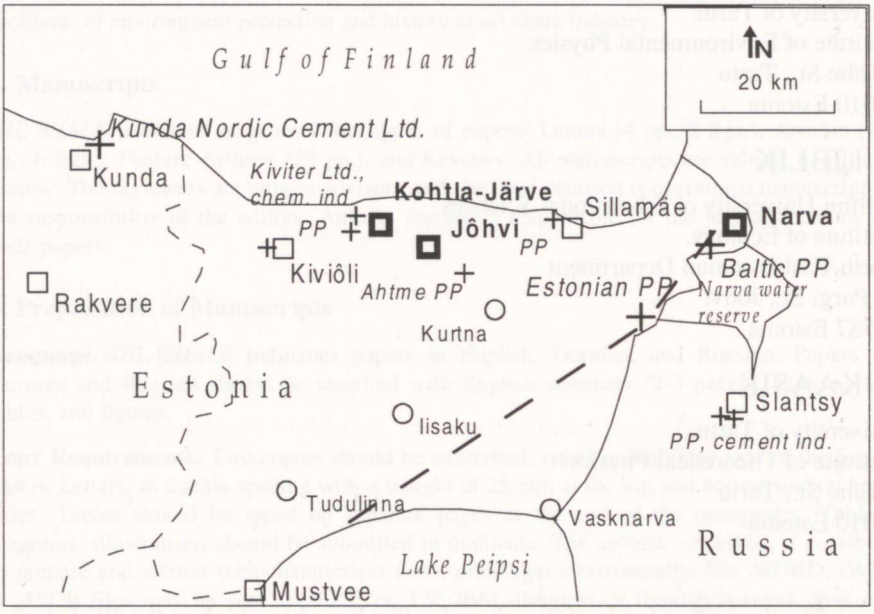

Fig. 1. Schematic map of study area with pollution sources taken into account (crosses). Towns are marked with rectangles, other settlements with circles. Heavy dashed line marks the snow sampling transect 1995/96

The top level of industrial emissions was achieved in the seventies. The electric filters were put into operation at Ahtme (1979) and Kohtla-Järve (1988) PP-s. Both power plants have small electric capacity and produce both electric and thermal energy (for domestic heating). Relatively low stacks (90 and 60 m) made them serious local pollution sources before the filters were launched. The higher stack (150 m) was built for Kohtla-Järve power plant during reconstruction.

In the beginning of nineties the sale of electric energy diminished about twice. Reasons were of economic origin: decreasing demand at the Russian market (partially due to economic fall, partially due to launching Sosnowy Bor nuclear power plant) and restructuring of Estonian economy. The emissions from two large power plants decreased nearly proportionally to

the energy production. As the Kunda cement industry has since 1997 effective electric filters, the cement dust emissions are drastically diminished.

Discussing the air pollution loads in the North-East of Estonia, we have to keep in mind the emissions from the near-border Slantsy region in Russia as well. The power plant in Slantsy (earlier oil shale-fired) was reconstructed, and it has been consuming natural gas since 1995. Dust emissions from Slantsy cement industry have been probably decreased due to production fall, but the data on emissions are irregular. The schematic map with locations of air pollution sources is given in Fig. 1.

The depositions from industrial sources have strongly affected the ecosystems in the most polluted areas [1, 2]. The anthropogenic impact is observed in the spruce stands near large pollution sources [3] and in the forest soils near the border of Russia and Estonia [4]. The oil-shale-specific ash particles were found in the lake sediments in the North-East of Estonia [5, 6].

The most comprehensive studies of pollution level and their ecological consequences should be based on the complex investigations of plant cover, lake sediments and soils. Such a study, however, presumes extensive fieldwork and chemical processing of a large number of samples. An alternative way is the air pollution dispersion and deposition modelling coupled with available field data. On the other hand, the model estimations are needed to estimate, what may happen in the future, if certain decisions or economical reasons induce any development of emission patterns.

This paper is aimed to give an overview of industrial emissions of oil shale fly ash, cement dust and sulfur dioxide and corresponding deposition loads during the industrialisation of this area after World War II and restructuring during last decade. A future scenario is discussed briefly. The results may constitute a basis for retrospective ecological impact estimations and studies on transformations of ecosystems exposed to high loads of sulfur and alkaline particles.

Methods

The air pollution deposition measurements are based on snow samples, which are collected during several winters since 1985. During two winters (1984/85 and 1995/96) the extensive and detailed study coincided with snowy and stable cold winter.

In March 1985, after 110 days of permanent snow cover, 80 snow samples from the north-eastern Estonia were taken. Seven samples were taken from Järve and Rapla districts (100-200 km W) [7, 8]. Forty-eight centimeter thick snow cover had been accumulated from December to March. No remarkable thawing occurred during the accumulation, wind was blowing predominantly from southern directions. The fly ash deposition

fluxes were calculated from the measured calcium fluxes with an assumption that there is 22 % Ca in the oil shale fly ash [9].

In February 1996, after 72 days of permanent snow cover, snow samples were taken from 18 points, including 13 points at the straight line from Baltic to Estonian thermal PP-S and up to 130 km to SW [10] (see Fig. 1). The snow cover was 25-50 cm thick, water equivalent 33-66 l/m^2. Winds were blowing 55 % from SE to SW and 30 % from NW to NE (the rest 15 % was distributed nearly equally between W and E). The fly ash deposition fluxes were calculated from the sum of fluxes of 26 fly ash components, which constitute together 29 % of fly ash [9]. The deposition fluxes of sulfate ion were measured in both field campaigns.

The model estimations are made using the air pollution dispersion and deposition model AEROPOL [11], which has been successfully validated for dispersion estimations in winter conditions [12]. The data on industrial emissions from power plants are gained from [5], and the emissions from Kunda cement factory originate from [13]. During the winter season the energy production and, consequently, the emissions are larger than annual averages. The temporal evolution of fly ash emissions is briefly reported in Fig. 2.

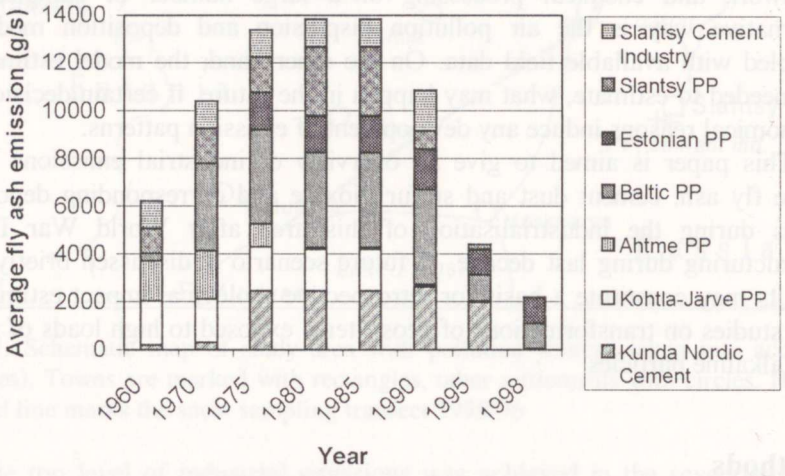

Fig. 2. Temporal evolution of average emissions of oil shale fly ash and cement dust from the seven largest pollution sources in the North-East Estonia

The maximal emissions during 1995 for large power plants (Estonian and Baltic) are about 50 % higher than the average and for smaller ones about twice higher. The average winter-time emissions for model computations were taken at 75 % of maximum, which is well between the maximum and annual average emissions.

The meteorological time series for two winters mentioned above originate from the archive of Estonian Institute of Meteorology and Hydrology. Although the winter-time deposition fluxes are not

representative for entire year (in each particular year or in climatic context), the comparison of measured and computed deposition fluxes give us proofs for deposition modelling precision.

A considerable part of deposited sulfatic sulfur originates from long-range transport and local sources (domestic heating, traffic, etc.). For comparison with measurement, the background load was added to the modelling results. The average SO_4^{2-} load for 1984/85 from seven samples in Järva and Rapla districts [7] was 13.5 mg/m^2 per day, which results in sulfur load 4.5 mg/m^2 per day. For 1995/96 the typical loads of SO_4^{2-} in central and southern Estonia during four winter months were 100 mg/m^2 or slightly more [14], resulting in the background load 0.35 mg/m^2 per day.

The large decrease in deposition loads can be a summary effect of (1) decreasing emissions in Central and western Europe, (2) fall of coal-based energy production in eastern Europe and (3) less use of coal and heavy oil products for central heating in Estonia. Although in both studies only open areas were chosen for snow sampling, the location of each concrete site (neighbourhood of any local source, orography, etc.) may have minor effect to the results. The meteorological conditions during each particular year (wind directions, precipitations) can also affect the deposition loads.

The samples from sites located more than 100 km away are considered here as background samples (free of direct influence of oil shale industry and energetics). Deposition loads from these samples are subtracted from the loads in north-eastern Estonia, when those are compared to the modelled ones. It is assumed, that (1) the long-range transport is nearly homogenous at distances about 100–200 m and (2) effect of local sources (domestic heating, transport, etc.) is nearly the same in north-eastern and central parts of Estonia. Although these assumptions are not strictly valid, we apologise that even roughly estimated background loads should be taken into account to correct the systematic overestimation of loads from the oil shale-fired enterprises due to other sources. This is especially important at distances about 20-50 km from power plants, where deposition loads are in the same range with background ones.

Due to the lack of monitoring we have no direct data on deposition loads before 1985. For earlier time the deposition is estimated using the AEROPOL model. The emissions of solid particles and sulfur dioxide could be estimated back to the sixties from energy, cement and chemical production data [5]. Following years were chosen for model estimations:

1960 - the eve of oil shale energetics: Kohtla-Järve and Ahtme PP-s are working; the first step of Baltic PP is just launched
1970 - the Baltic PP is working with full capacity; Estonian PP- partially
1975 - electric filters are installed at Ahtme PP; Kunda Cement Industry is reconstructed; cement production increased rapidly
1980 - nearly the maximum of oil shale mining and energy production
1990 - demand at Russian energy market is diminishing; the Kohtla-Järve PP has electric filters

1995 - energy production is diminished about twice of maximal
1998 - effective electric filters are installed at Kunda Nordic Cement

Another situation, where only model simulation can give some reliable knowledge, is the possible development in the future. There are simulated the deposition loads for a situation, where Baltic PP is closed due to amortisation and Estonian PP is working with full capacity (approximately at the level of 1980), but has 25 % more effective electric filters as a result of reconstruction.

The air pollution dispersion from a point source is described by a bi-Gaussian (in vertical and cross-wind directions) plume in the AEROPOL model. It is assumed that the fly ash particles reaching the underlying surface are deposited. The wet deposition is calculated, integrating the precipitation-scavenged matter over the path of hydrometeors. The details are given in [11]. While modelling the deposition loads, the sulfur content of deposited fly ash was assumed 7.5 % [15], appearing in sulfate form in the snow water. The kriging algorithm was used to interpolate the deposition loads between snow sampling points.

Results and Discussion

Deposition Loads

The measured and computed deposition loads of solid particles and sulfur in the studied area are shown in Figures 3-7. The term 'solid particles' embraces the oil shale fly ash from power plants and cement dust from the cement industry. The measured sulfur loads were calculated from the content of sulfate ion in snow water. The isolines of deposition loads are shown in Figures 3-4 and 6-7.

The measured loads of oil shale originated solid particles (fly ash and cement dust) from snow samples during winter 1984/85 and corresponding model estimations are given in Fig. 3. The snow sampling points are marked with small diagonal crosses. The highest concentrations (more than 200 mg/m^2 per day) are found near Kohtla-Järve, Kunda and between Estonian PP and Baltic PP. The measured and modelled deposition does not agree well near Slantsy, where the measured loads are several times higher than the modelled ones. Because of sparse set of measurements a few high values can affect interpolation over large areas.

The patterns of sulfur loads (Fig. 4) seem quite similar to the distribution of particles. According to the modelling results most of sulfur is deposited in the content of fly ash or cement dust. The deal of sulfur dioxide deposition is usually 5-15 % (based on model estimations, no experimental proof), because snow-covered areas and falling snowflakes have large resistance to sulfur dioxide adsorption. The small sulfur deposition from the gaseous phase near thermal power plants has been mentioned earlier in several studies [16].

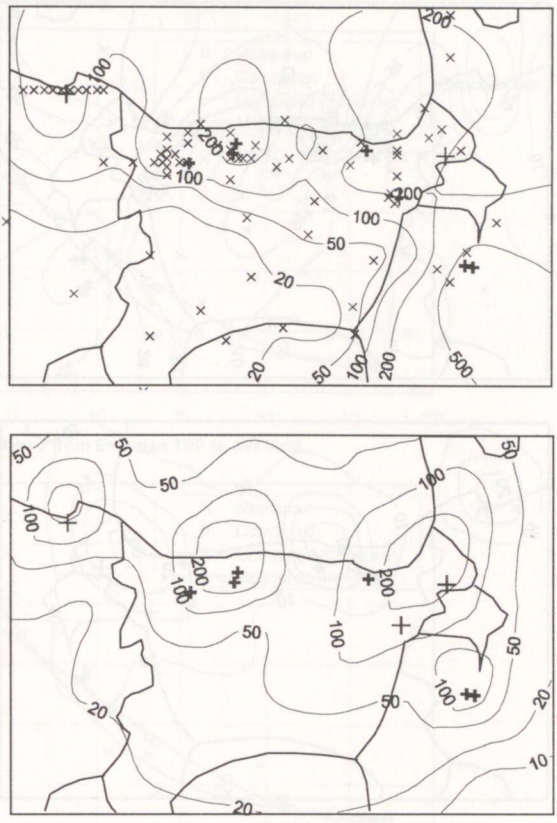

Fig. 3. Measured (*a*) and computed (*b*) deposition loads of solid particles during winter of 1984/85 (mg/m^2 per day, snow sampling points are marked with diagonal crosses)

The model tends to underestimate the sulfur loads not only for Slantsy region but also NE from Narva. A possible reason is SO_2 conversion into the aerosol in the highly elevated plumes of large power plants with following rainout, which is not adequately described in the AEROPOL model.

The deposition during winter 1995/96 is presented in Fig. 5. All the snow samples during this season were taken at a straight line from Estonian PP to the South-West (Fig. 1). The leftmost point was situated near Baltic PP (18 km NE from Estonian PP). The measured and modelled loads coincide fairly with the exception that there is no significant difference of measured ash deposition between first two points (polynomial fitting creates a maximum between them). Both fly ash and sulfur loads are about twice lower than at 1984/85, which is in agreement with industrial emissions.

During both 1984/85 and 1995/96 winters the measured fly ash loads are in agreement with the sulfur fluxes, assuming that (1) the sulfur content of fly ash is 7.5 %, (2) all the sulfur is converted into the sulfatic form before measurement and (3) there are no significant sources of sulfur besides the fly ash. Consequently, the conclusions from model calculations agree in general with those based on the measured loads.

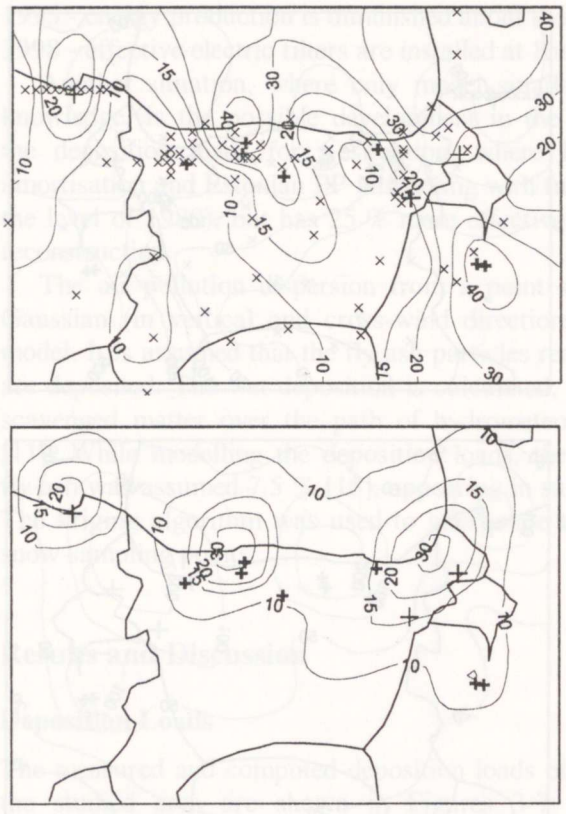

Fig. 4. Measured (*a*) and computed (*b*) sulfur deposition loads, 1984/85 (mg/m^2 per day, snow sampling points are marked with diagonal crosses). The background sulfur load 4.5 mg/m^2 per day is considered in model estimations

The modelled temporal and spatial evolution of deposition of solid particles from the sixties to nowadays and near future is presented in Fig. 6. The evolution of sulfur deposition is given in Fig. 7. The highest average loads were found in the seventies and eighties. The forecast is based on the assumption that the amortised Baltic PP will be closed and the newer Estonian PP will work at the full capacity, but with reconstructed purification system, which is 25 % more effective than now. It is easy to see that the discussed scenario will be highly recommended: the alkaline and sulfur deposition loads near Narva and Narva-Jõesuu will diminish more than twice.

Comparison of solid particle deposition isolines (Fig. 6) with air pollution zones in 1980 and 1990, calculated by maximum and annual average concentrations of oil shale fly ash in the near-surface air layer [17], shows following annual average deposition loads depending on air pollution level, mg/m^2 per day:

In the zone 5 (very high pollution level)	- more than 200
Zone 4 (high level)	- 150-200
Zone 3 (relatively high level)	- 50-150
Zone 2 (moderate level)	- 20-50
Zone 1 (low level)	- 5-20
And zone 0 (very low level)	- below 5

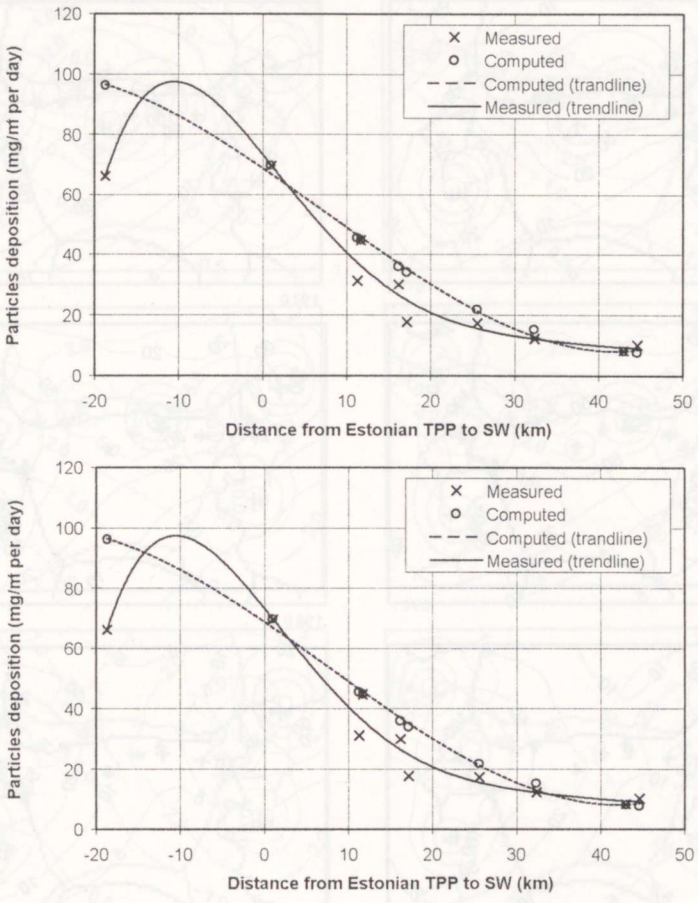

Fig. 5. Measured and computed deposition loads of solid particles and sulfur, 1995/96 (mg/m^2 per day). The background sulfur load 0.35 mg/m^2 per day is considered in model estimations

The data will be used for forecasting of deposition intensity of particles by measured and calculated air pollution data.

Computed data in Fig. 7 show that in 1995-1998 most of north-eastern Estonia is effected by annual average sulfur deposition load below 3 kg S/ha: 0.5 mg/m^2 per day from PP-s as computed by AEROPOL model plus about 0.3-0.4 mg/m^2 background [14].

In Nordic countries the sulfur deposition load on arable lands and forested areas must not exceed 5 kg S/ha [18]. This value is exceeded at distances less than 10-15 km from Narva. During the worst situation period (1970-1980) this level was exceeded at distances up to 30 km from Narva and 10 km around Ahtme and Kohtla-Järve PP-s and Kunda.

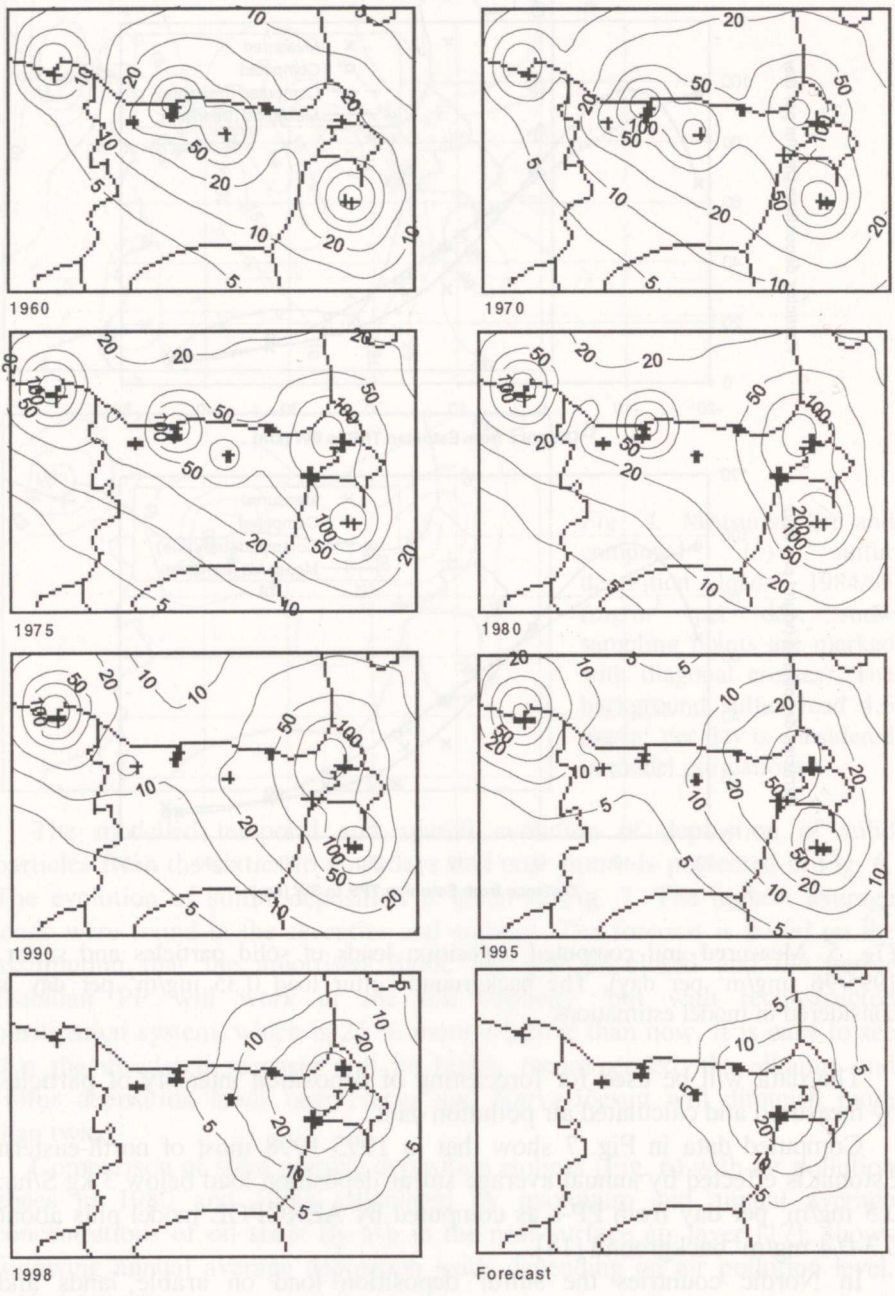

Fig. 6. Computed annual average deposition loads of solid particles from 1960 to 1998 and forecast (mg/m^2 per day)

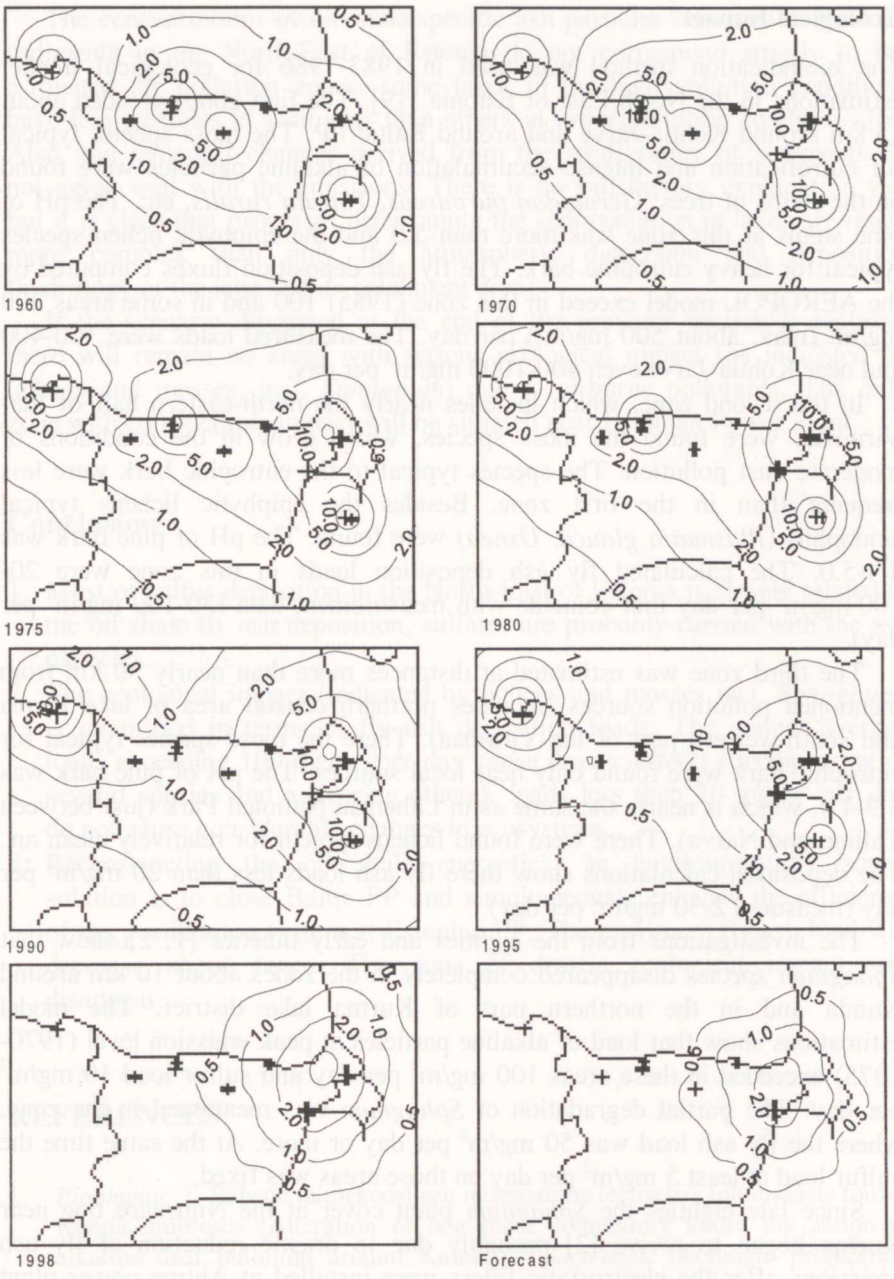

Fig. 7. Computed annual average sulfur deposition loads from 1960 to 1998 and forecast (mg/m^2 per day)

Ecological Impact

The bioindication method was used in 1983-1986 for ecological impact estimations in the North-East of Estonia [19]. The first zone extended about 15 km around Kohtla-Järve and around Baltic PP. The moss species typical for eutrofication and intense accumulation of alkaline particles were found on the stems of trees: *Geratodon purpureus*, *Totrula ruralis*, etc. The pH of pine stems at this zone was more than 5.0 and the epiphytic lichen species typical for heavy eutrophic bark. The fly ash deposition fluxes computed by the AEROPOL model exceed in this zone (1985) 100 and in some areas 200 mg/m^2 (max. about 500 mg/m^2) per day. The measured loads were 100-400 and near Kohtla-Järve even 400-1000 mg/m^2 per day.

In the second zone, which includes nearly the north-eastern half of Ida-Virumaa, were found the moss species, which grow in the conditions of moderate dust pollution. The species typical to the eutrophic bark were less frequent than in the first zone. Besides the epiphytic lichens typical acidophils (*Plasmatia glauca, Usnea)* were found. The pH of pine bark was 4.4-5.0. The calculated fly ash deposition loads in this zone were 20-100 mg/m^2 per day that coincide with measurement data (30-100 mg/m^2 per day).

The third zone was estimated at distances more than nearly 30 km from mentioned pollution sources (includes northern coastal area of lake Peipsi and south-western part of Ida-Virumaa). There the moss species typical for eutrophic bark were found only near local sources. The pH of pine bark was 3.9-4.4, which is nearly the same as in Lahemaa National Park (just between Tallinn and Narva). There were found lichens typical for relatively clean air. The deposition calculations show there fly ash loads less than 20 mg/m^2 per day (measured 2-30 mg/m^2 per day).

The investigations from the eighties and early nineties [1, 2] show that *Sphagnum* species disappeared completely in the zones about 10 km around Kunda and in the northern part of Kurtna lake district. The model estimations show that load of alkaline particles at peak emission level (1970-1975) exceeded in these areas 100 mg/m^2 per day and sulfur load 10 mg/m^2 per day. The partial degradation of *Sphagnum* was mentioned in the zone, where the fly ash load was 50 mg/m^2 per day or more. At the same time the sulfur load at least 5 mg/m^2 per day on those areas was fixed.

Since late eighties the *Sphagnum* plant cover at the Niinsaare bog near Kurtna began to revive [2] probably due to drastic reduction of fly ash emissions after the electrostatic filters were installed at Ahtme power plant 1978. Later the annual average fly ash load near Kurtna did not exceed 30 mg/m^2 per day (model estimation). From this case we have to conclude that the "reaction time" needed for remarkable changes in *Sphagnum* plant cover after the pollution level changed, is nearly 10 years. Consequently, we can expect the ecological effects from electrostatic filter at Kunda Nordic Cement (installed 1997) in next 5-7 years.

The concentrations of oil shale-specific ash particles observed in the lake sediments in the North-East of Estonia do not correspond strictly to the estimated air pollution zones: some lakes in the high pollution conditions have less particles in sediments than others in lower pollution conditions [6]. Also, the temporal changes derived from the stratigraphy of sediments do not agree well with the emissions. There is no satisfactory explanation yet, but it is clear that processes determining the sedimentation in lakes are much more complex than only the atmospheric dispersion and deposition (hydrology of the lake and its catchment area).

If the scenario discussed at the end of the previous section is realised, there will remain no areas with serious ecological impact (as indicated by lichens and mosses, incl. *Sphagnum*) due to airborne pollutants. The only zone with considerable impact will be situated near Estonian Power Plant.

Conclusions

1. Most of sulfur deposition in the North-East of Estonia is closely related to the oil shale fly ash deposition, sulfates are probably carried with the ash particles.
2. The ecological impact (indicated by lichens and mosses incl. *Sphagnum*) is quantified in terms of fly ash deposition loads. The annual average loads exceeding 100 mg/m^2 per day cause drastic effects (disappearing of several species and appearing others), loads less than 20 mg/m^2 per day do not cause considerable changes in ecosystems.
3. Reconstructing the oil shale energetics, an environmentally sound solution is to close Baltic PP and simultaneously enhance the efficiency of the purification system at Estonian PP. The maximum fly ash load will decrease about twice. The zone of drastic ecological impact will disappear.

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