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HISTORY OF FLY ASH EMISSION AND PALAEORECORDS OF ATMOSPHERIC DEPOSITION IN THE OIL SHALE COMBUSTION AREA

**J.-M. PUNNING
V. LIBLIK
T. ALLIKSAAR**

Institute of Ecology
Tallinn, Estonia

This paper presents a comprehensive analysis of the history of oil shale mining and combustion in northeastern Estonia, the formation of atmospheric influxes and spatio-temporal distribution of deposited impurities. The data calculated for the subsurface air layer and the distribution of impurities in lake sediments are in a very weak correlation. These show an important role of the subsurface turbulence, landscape topography, as well as the deposition pathways and processes of influxes into the lake.

Introduction

The impact of anthropogenic factors on different ecosystems can be assessed by examining the relationships between the biogeochemical cycling and characteristics of ecosystems like changes in lake fauna and flora, state of forests, ecophysiological transformations in species, etc. Changes in the atmosphere show well the ability of man to alter the environment. The transportation and biogeochemical cycling processes are the fastest in the air and therefore much attention is paid to the modelling of the pollutant fields in the atmosphere [1-5].

For the landscape and ecosystem studies it is especially important to understand the processes assisting the formation of near-surface pollution fields and influxes of atmospheric impurities to the earth's surface. Several attempts have been made to find the correlations between the distribution of different compounds in the atmosphere and deposited matter using various models. As these models must be rather specific depending on the structure of the studied landscape, as well on the

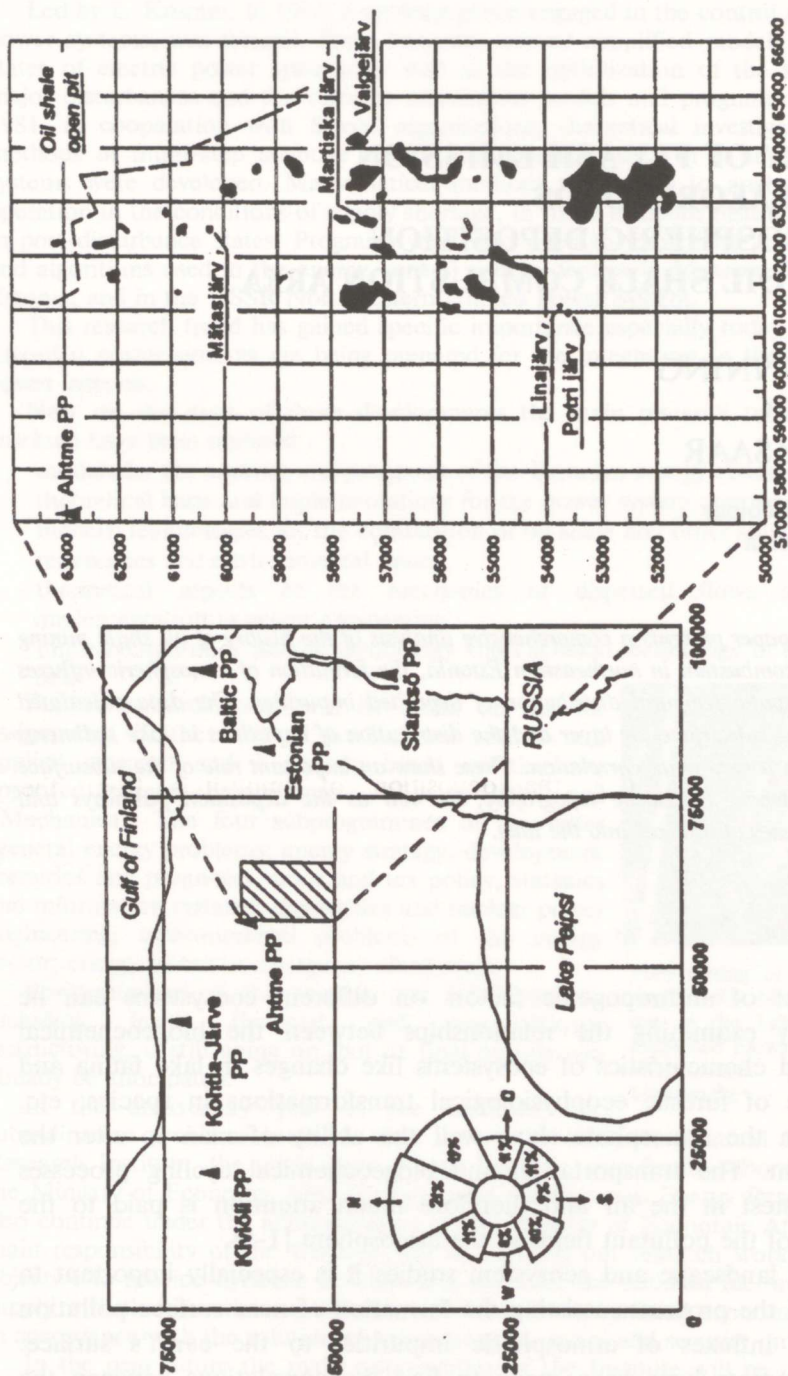


Fig. 1. Location of the study area, power plants (PP, \blacktriangle), studied lakes and the wind-rose (average for years 1958-1991) of the region. The distances by X, Y-axis in metres are given

applied spatio-temporal scale, they need verification. In the global scale good results have been obtained by using specific impurities like cesium isotopes emitted from a point source during the accident at Chernobyl nuclear power station in May 1986 [6].

In a short-term scale the main problem lies in establishing the monitoring set. Actually, monitoring data reflect influx values for certain time intervals and their interpolation often leads to wrong results.

In a long-term scale sediment profiles are widely used to study the variances in the physical environment and their impact on the ecosystems [7, 8]. Major advantages of this approach are integration of data in sediments and a possibility of eliminating short-term and nontypical fluctuations, also its cheapness in comparison with the continuous monitoring programme. Naturally, this calibration is complicated, because some additional problems, such as the multitude of factors influencing the formation and preservation of primary information in sediments, must be considered as well.

In the case of aerial migration the spreading of pollutants in the atmosphere, their spatio-temporal distribution and outwashing on the land surface deserve special attention. There are many models for describing the extraction of pollutants from the atmosphere, as well as the advective and turbulent motions in the atmosphere and their influence on the matter released into the air [9]. The transformation processes of impurities and their atmospheric deposition pathways (dry or wet) have significance, too [10].

In mosaic landscapes the deposition of impurities from the atmosphere and their spatial distribution on the land surface are extremely complicated because of the multitude of physical processes near the surface caused by the roughness of landscape.

We have made an attempt to study the spatio-temporal regularities in the distribution of fly ash in northeastern Estonia using historical data about the combustion of oil shale at power plants and emission of fly ash to the atmosphere. Both approaches: the modelling of the distribution of impurities in near-surface air layers and their spatio-temporal distribution in lake sediments were used and the corresponding data compared.

History of Human Impact on the Study Area

The study area - the Kurtna Landscape Reserve (LR) - is located in northeastern Estonia in the Ida-Viru district, about 15 km SE of the town of Jõhvi (Fig. 1). The LR lies in a transitional zone between a densely populated and heavily industrialized oil shale mining region, which is one of the areas most affected by man in Estonia, and a sparsely inhabited territory with big forests and mires. About 18-30 km west and southwest

Table 1. Power Plants (with dates of installation) and Their Average Fly Ash Emissions [13-16]

Average height of stacks, m	Period, years	Combustion of oil shale, kilotonnes	Emission of fly ash		
			t yr ⁻¹	g s ⁻¹ (max)	g s ⁻¹ (mean)
Kohta-Järve PP, installed in 1949					
60	1959-1960	978	58000	2900	1830
	1969-1970	936	57250	2800	1815
	1975	958	58400	3100	1850
	1979-1980	909	55500	2975	1750
	1988	545	28200	1900	895
150	1990	500	1464	81	46
Ahtme PP, installed in 1951					
95	1959-1960	674	53000	3145	1700
	1969-1970	821	64000	3650	2030
	1975	383	29950	2375	950
	1979-1980	357	2360	175	75
	1988	426	2000	185	65
	1990	401	2230	178	70
Baltic PP, installed in 1960					
160	1960	820	9800	500	310
	1969-1970	10350	120000	4800	3800
	1975	11090	124000	4850	3930
	1979-1980	12475	125000	4850	3965
	1988	10720	104200	4230	3300
	1990	11300	125060	4758	3965
Estonian PP, installed in 1969					
250	1970	2700	13500	642	425
	1975	9985	50000	1950	1585
	1979-1980	12230	48420	2540	1535
	1988	10980	51300	2600	1625
	1990	11170	53620	2805	1700
Slantsy PP (Russia)					
100	1960	400	32500	1855	1030
	1975	575	46100	2550	1460
	1979-1980	1130	91000	4800	2880
	1988	900	72880	3880	2310
	1990	657	52550	2685	1665

of the LR Baltic and Estonian Power Plants (PP), and 5-8 km to the southeast Ahtme PP are located. In addition to local power plants, emissions from Slantsy PP in Leningrad District (Russia), about 30 km to the southeast from the study area, reach the Kurtna LR as well (Fig. 1).

At present oil shale mining and processing affect most severely the area under study. The Estonian oil shale fields are the world's largest oil shale deposits of commercial significance, which have been exploited since 1916. Before World War II, about half of the oil shale extracted was used for the production of fuel oil and motor fuels, including gasoline. After the Second World War, in 1949-1969, new oil shale fired power plants were erected (Table 1; Fig.1) and at the end of the 1970s

about 27 million tonnes of oil shale was burned in the power plants. Owing to a high mineral content of this fuel the power plants and retorting plants produce huge amounts of wastes. The bulk of the wastes is caught and directed by tubes to sedimentation basins or transported onto terricones. However, more than 220 kilotonnes of fly ash passed through the filters and was emitted into the atmosphere annually in the 70s and 80s (up to 322 kilotonnes in 1980) (Table 1; Fig. 2).

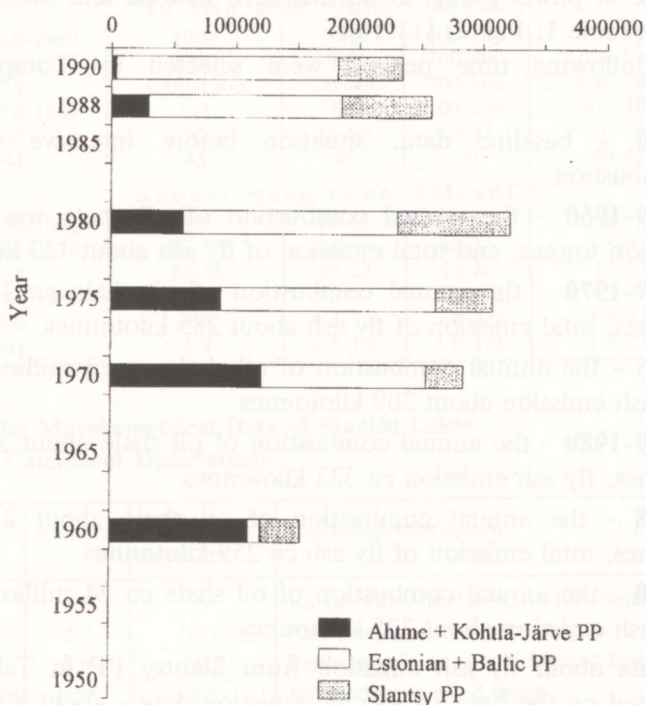


Fig. 2. Fly ash emission from power plants in northeastern Estonia and nearby situated Slantsy PP (Russia)

The alkaline oil shale fly ash is characterized by high concentrations of several heavy metals, carbonates, sandy-clay minerals and also harmful organic substances. Depending on the type of boilers and purification systems for outlet gases, the emission per energy unit varies largely. Heavy metals can be found relatively abundantly in the oil shale fly ash: the content (mg kg^{-1}) of Ni is 53, Pb - 163, As - 43, Cd - 2, Mn - 300, Hg - 1 [11]. As the content of organic matter in oil shale is relatively low (30-40 %), the total amount of residues, including gaseous and dust products, is big. Fly ash passing electrofilters consists mainly of CaO (30.5 %), Al_2O_3 (8.8 %), K_2O (6.8 %), Fe_2O_3 (4.2 %) and MgO (2 %) [12].

Methods

The main aim of the current study was to simulate spatio-temporal distribution of the calculated concentrations of fly ash in the overground air layer and deposition intensity of the ash on the land surface at different emission loads. To realize this idea the following data and methods were used:

- (i) Historical data about the emissions of fly ash during combustion of oil shale at power plants in northeastern Estonia and Slantsy PP in Russia (Table 1; Fig. 2) [13-16].

The following time periods were selected for comprehensive analyses:

- **1940** - baseline data, situation before intensive oil shale combustion
- **1959-1960** - the annual combustion of oil shale was about 3 million tonnes, and total emission of fly ash about 153 kilotonnes
- **1969-1970** - the annual combustion of oil shale ca 15 million tonnes, total emission of fly ash about 285 kilotonnes
- **1975** - the annual combustion of oil shale ca 23 million tonnes, fly ash emission about 309 kilotonnes
- **1979-1980** - the annual combustion of oil shale about 27 million tonnes, fly ash emission ca 322 kilotonnes
- **1988** - the annual combustion of oil shale about 25 million tonnes, total emission of fly ash ca 259 kilotonnes
- **1990** - the annual combustion of oil shale ca 24 million tonnes, fly ash emission about 235 kilotonnes

The data about fly ash emission from Slantsy PP in Table 1 are calculated on the basis of specific emission data - about 80 kg of fly ash per 1 t of oil shale [13].

- (ii) The maps on the distribution of fly ash in the near-surface atmosphere were compiled calculating short-time (0.5 hour) maximum (MaxC) and annual average concentration (MeanC) fields of fly ash in the overground air layer using the semi-empirical modelling method [3, 17]. To calculate MaxC and MeanC values, the data on maximum emission intensity of fly ash in the winter period and annual average emission intensity were used, respectively (Table 2). Recurrence of wind directions for the whole observation period was calculated as an average of many years (1958-1991) using data obtained at the Jõhvi Meteorological Station (Fig. 1). The average annual wind velocities of 3.7-4.9 m s⁻¹ at different wind directions and the maximum speed of 95 % probability of 9 m s⁻¹ were used as mean values. It should be pointed out that the obtained

values describe concentration fields near the land surface, dry deposition being the main deposition mechanism.

Table 2. Calculated Concentrations of Fly Ash in the Overground Air Layer in Different Areas of Kurtna Kame Field, $\mu\text{g m}^{-3}$

Year	Lakes (Fig. 1)			
	Mätasjärv	Martiska	Valgejärv	Linajärv, Potri
	30 min max. conc. (MaxC)			
1958-1960	1000	800	600-700	500-600
1968-1970	1200	900	700-800	600-650
1975	1000-1200	700-800	700-800	650-700
1978-1980	115	95	100-110	100-105
1990	90	85-90	90-95	70-75
1994	45	45	50	40-45
	Annual mean conc. (MeanC)			
1958-1960	20	10-15	7-8	6-7
1968-1970	25-30	13-15	10-11	8-9
1975	30-35	20-22	15-16	12-13
1978-1980	4.3	3.9	3.9-4.0	3.3-3.4
1990	3.7	3.3-3.4	3.4-3.5	2.8-2.9
1994	1.9	1.7-1.8	1.8-1.9	1.5-1.6

Table 3. The Morphometrical Data of Studied Lakes and Their Catchment Descriptions

Lake	Area, ha	Maximum depth, m	Catchment description
Mätasjärv	0.5	9	Kames with pine forest, on the shores forested bog
Martiska	2.7	6.5	Pine forest, partly forested bog
Valgejärv	8.6	10.5	Partly pine forest, partly forested bog
Potri	0.56	6.5	Forested bog, mixed forest
Linajärv	1.0	7.2	Forested raised bog, partly transitional mire, on the kames pine forest

(iii) To assess the distribution of deposited fly ash compounds on the earth's surface, the geochemical data on the upper parts of sediment cores from different lakes of the Kurtna Kame Field (lakes Mätasjärv, Martiska, Valgejärv, Potri and Linajärv - Fig. 1) were used. These are all relatively small closed lakes located in glaciokarstic depressions. They are surrounded by forests and their shores are paludified (Table 3).

Palaeoecological investigations were carried out in the Kurtna LR in 1981-1991 [8]. The upper parts of sediment cores were sampled continuously and subsamples at 1-2 cm intervals were taken for geochemical analysis. The accumulation time of these subsamples varies

from 2.5 to 5-6 years. In the samples the concentration of heavy metals was measured using atomic absorption and neutron activation methods, and the distribution of spheroidal fly ash particles (SFAP) as the indicators of fuel combustion was determined with the Renberg-Wik method [18-20]. To estimate the age of separate layers, the indicators of human impact, such as morphology of the curves of heavy metals [21] and SFAP [22], cesium as the marker of nuclear explosions and ^{210}Pb method [21], were used.

In the current study the age scale was compiled using mainly linear extrapolation. Therefore the layers, showing a rapid increase in the concentration of SFAP, were presumed as having accumulated in 1950. The weight of dry mass was summed for all sublayers above the 1950-year layer. The age of every analysed layer was calculated so that the accumulation rate of matter over this period was taken constant. The data obtained are in good accordance with other datings [19, 23]. Using the concentration data and sedimentation rates, the influxes of chemical compounds and SFAP per cm^2 in a year were calculated.

Results and Discussion

The main source of the atmospheric pollution in northeastern Estonia is fly ash emitted from thermal power plants operating on oil shale [8]. The influx of impurities on the landscape depends on various factors, such as the parameters of sources (height above ground level), emissions (temperature, flow-out velocity, intensity per time unit), meteorological characteristics (wind speed, direction, atmospheric stratification conditions), surface profile, deposition pathways, etc. The real short-term influx values for different impurities vary largely, as is vividly seen from monitoring data. Therefore, the trend of atmospheric influxes could be better studied using the integrated concentration values of compounds. These data are available and can be obtained by analysing lake sediments.

A special problem is the correct selection of indicators reflecting the regularities of the distribution and deposition of compounds in fuel gases. The correlations between concentrations of chemical elements and spheroidal fly ash particles (SFAP) in sediment cores show that SFAP correlate best with K and Na (0.98-0.82 in the studied lakes). The correlations between Ca and SFAP are less and vary in a wide range (from 0.93 in Lake Linajärv up to 0.37 in Lake Mätasjärv). The variations are probably caused by differences in volatility of chemical compounds (mainly oxides), as well by the postsedimentational migration of elements in lakes depending on their hydrochemical characteristics.

SFAP, formed during the combustion of fossil fuels, are relatively resistant to biogeochemical transformation and migration processes in

lakes. Therefore their distribution patterns in sediments are more appropriate for studying the impact of oil shale combustion than chemical indicators.

The general regularity in the distribution of SFAP in lake sediment cores is revealed in the similarity in their curves (Fig. 3a). It shows an increase in the SFAP concentration upwards in the cores, which has correlation with the combustion history of oil shale at power plants in the temporal scale. However, there are also certain differences between the curves from different lakes: cores from lakes Martiska and Potri have distinct influx maximums of SFAP and heavy metals in layers formed in the mid-1980s, but in the cores from lakes Mätasjärv, Valgejärv and Linajärv the concentration of SFAP increases up to surface layers (1989-1990).

The total quantity of SFAP influxes per sediment surface unit vary markedly in lakes under study. Figure 4 shows that the influx of SFAP to different lakes differs up to 3-4 times throughout the whole considered interval. The lowest influx values are recorded in Lake Valgejärv in the eastern part of the Kurtna LR and highest in lakes Potri and Linajärv in its southern part (Fig. 1). The same influx ratios can be obtained by calculating values per gram of mineral matter, because loss on ignition values in different lakes differ only slightly [23].

The values of overground concentrations of oil shale fly ash were calculated for the areas of studied lakes and the concentration maps for all reference periods were obtained (Fig. 5). By the calculations, the influence of all power plants located in northeastern Estonia on the Kurtna LR at different wind directions was taken into account. The impact of Slantsy PP was not considered when calculating and compiling the maps.

The overground pollution level at certain locality depends on its distance from pollution sources and wind direction. Seemingly, the computed highest level of fly ash pollution was reached in the study area in 1968-1975 (Table 2) (without Slantsy PP). After 1977 the air pollution level with fly ash decreased (Fig. 3b), because in Ahtme PP the electrostatic precipitators were installed in 1976-1977. As a consequence, the yearly emission of fly ash from this PP fell from 80000 t to 2366 t (Table 1). Influenced by the dominating wind direction in the nearest vicinity, in the region of Lake Mätasjärv, 8 km southeast from Ahtme PP, the calculated maximum fly ash concentration values (about 1000-1200 $\mu\text{g m}^{-3}$ in 1958-1975) decreased about 9-10 times, whereas mean concentration values (20-35 $\mu\text{g m}^{-3}$) decreased 7-8 times. In the region of Lake Potri and Linajärv (about 14 km from Ahtme PP), ash concentration in the air decreased less (3.5-6 times) due to a higher share of Baltic and Estonian PPs in dust pollution.

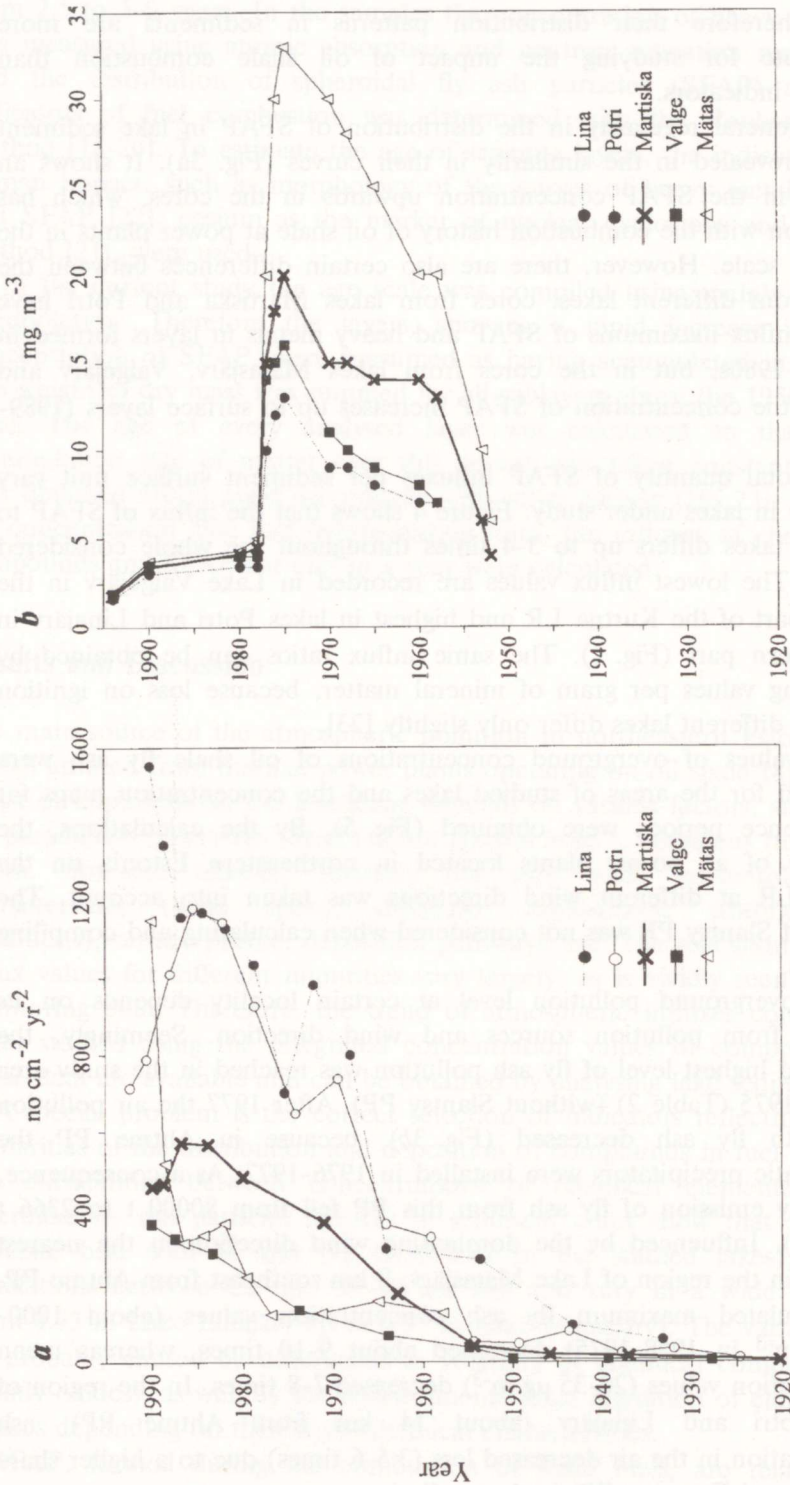


Fig. 3. Temporal distribution of spherical fly ash particles in the sediments of studied lakes (a) and calculated mean fly ash concentration in their subsurface atmosphere (b)

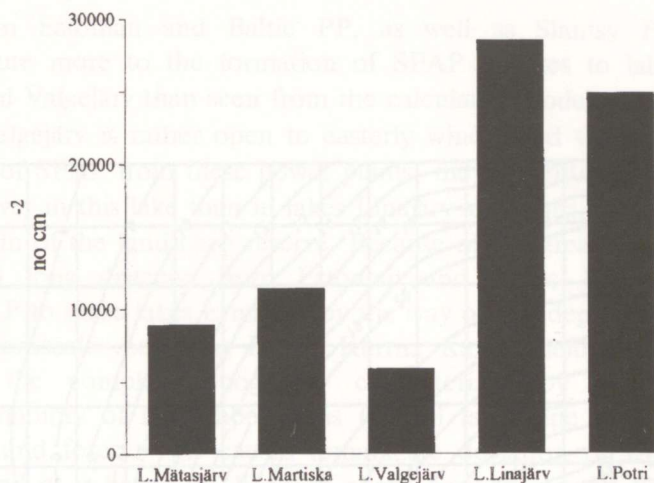


Fig. 4. Total sum (for years 1950-1990) of spherical fly ash particles accumulated in the sediments of different lakes

Comparison of the historical records about the energy generation and development of emission purification technologies, expressed as the amount of emitted fly ash (Table 1, Fig. 2) supported by SFAP influx curves in the sediments (Fig. 3a), shows that the Ahtme and Kohtla-Järve PP may serve as the main sources of pollutants, deposited by the dry way, for the lakes Mätasjärv, Martiska and probably also Valgejärv. The influx of SFAP to lake sediments largely depends on the position of the lake with respect to relief forms and its catchment structure. According to calculations, the highest fly ash concentrations in the air should occur above Lake Mätasjärv, but actually the influx of SFAP into lake sediments is one of the lowest here (Fig. 3a,b). Most probably this may be related to the location of Lake Mätasjärv in a small and deep kettle hole. The forest growing closely around the waters inhibits the fall of air currents above the lake. In the sediments from Lake Martiska the concentration of SFAP increased until the middle of the 80s. Afterwards the influx of SFAP to Lake Martiska decreased, as well as the calculated values of fly ash in the overground air layer above this lake. At the same time, the influx of SFAP into lakes Mätasjärv and Valgejärv continues to increase, probably due to the emissions from Estonian and Baltic PP.

Weak correlations between the computed concentration fields and temporal distribution of various indicators of fly ash emissions (SFAP, chemical elements) in sediments might also be explained by differences in vertical air mass characteristics and deposition pathways. Depending on their volatility and air mass characteristics, the pollutants will deposit either by a dry or wet way. This leads to great seasonal or even day and night differences in the deposition parameters, and may cause big variation of correlations between fly ash components from different sediment cores.

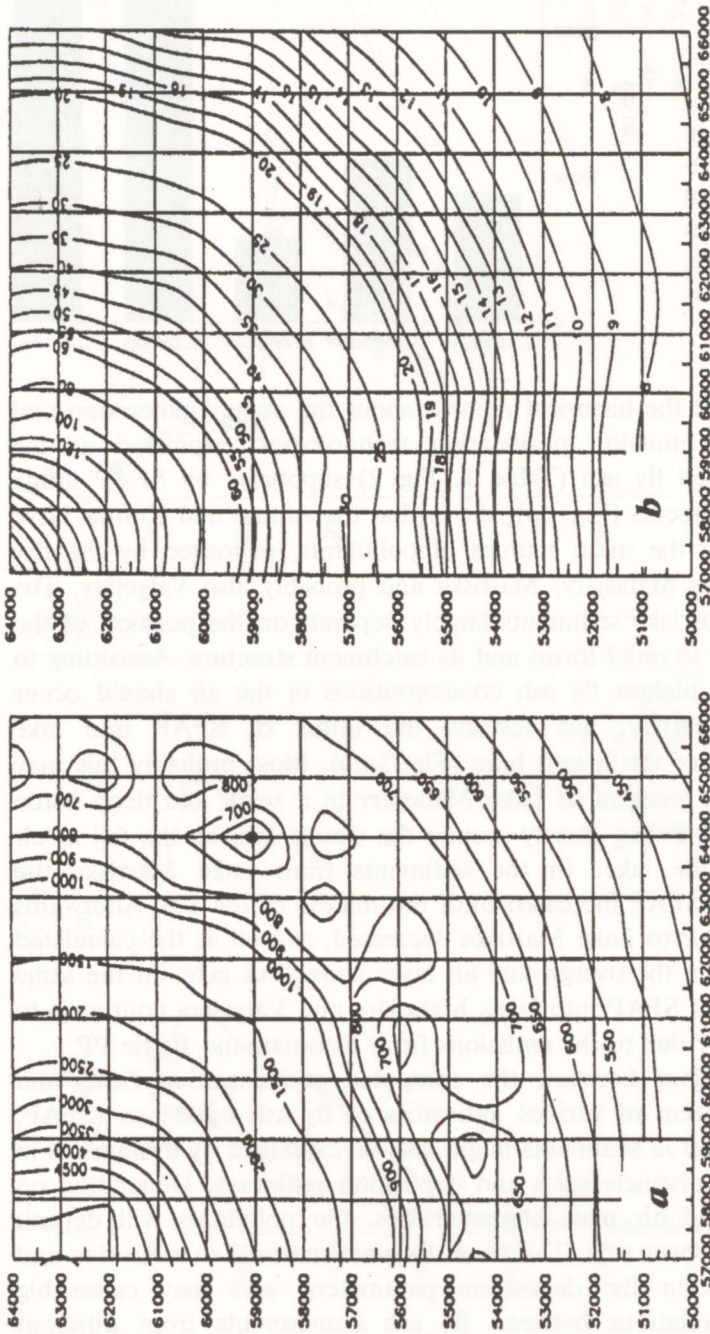


Fig. 5. Oil shale fly ash concentrations ($\mu\text{g m}^{-3}$) in the overground air layer of Kurlna LR in 1975 (location of study area given on Fig. 1): a - maximum short-time values (MaxC), b - annual mean values (MeanC)

Emissions from Estonian and Baltic PP, as well as Slantsy PP, obviously contribute more to the formation of SFAP influxes to lakes Linajärv, Potri and Valgejärv than seen from the calculated model values. Although Lake Valgejärv is rather open to easterly winds, and therefore also to the influx of SFAP from these power plants, the concentration of SFAP is much lower in this lake than in lakes Linajärv and Potri, located at the south margin of the landscape reserve. Because of the great height of chimneys and long distance from Estonian and Baltic PP, the deposition of SFAP to these lakes is mainly by the way of wet deposition.

Meso- and microscale modelling in the Kurtna Kame Field is also complicated by the complex topography characterized by a high morphological variability of landscape forms (glacial landscape of hills and depressions) and forest. This causes remarkable deviation of wind direction and speed at a height of 10-20 m above ground. As the flow field is altered by the surface features, the transport and location of the plume area is difficult to estimate. All this causes differences between calculated fly ash concentration fields over the land surface and estimated SFAP concentrations in lake sediments.

Conclusions

Comparative analysis of the calculated distribution of the fly ash emitted from oil shale combustion over the Kurtna Kame Field with data obtained by the analysis of sediments in lakes show high variability. There are certain similarities between general spatio-temporal trends of spherical fly ash particles (SFAP) and chemical element concentrations in lake sediment cores, and oil shale combustion history of nearby located big power plants, but the absolute influx values of emission impurities into different lakes differ from computed subsurface atmospheric fields.

Essential spatial differences occur between computed maps of fly ash distribution in the overground atmosphere and concentrations of SFAP, K, Ca and other elements measured in sediments related to emitted fly ash. The differences were biggest between computed mean and measured values in the southern part of the area in the 1980s and later. Differences between the computed maximum and measured values are less but the regularities are the same. Reasons for these phenomena may be different.

First, the computed model of the formation of fly ash fields in the atmosphere operates with mean wind direction and its share in the transport of fly ash from different emission sources. In the calculations the structure of wind, temperature and turbulence through the vertical profile is taken as homogeneous. The models, using the maximum values of emissions, seem to correspond better to the real situation. Actually,

atmospheric turbulence and wind fluctuations play a very important role in the transport of air pollutants [10].

Second, the calculated model values might be compared mainly with the spatio-temporal distribution of influxes caused by dry deposition under stable atmospheric conditions. Unfortunately it is not possible to make out which part of impurities in the sediments has deposited by a dry and which part by a wet sedimentation pathway. Naturally, the outwash is more effective by wet deposition and SFAP from a much wider atmospheric layer are washed out. Therefore the share of Baltic and Estonian PP in the formation of influxes from the atmosphere is greater than it follows from computed atmospheric fly ash fields. Differences in the absolute values of SFAP influxes to lake sediments evidently speak about a higher impact of these power plants than shown by imitation models.

The palaeoecological method is a good tool to estimate real influxes from the atmosphere during certain periods, providing also possibilities of verifying complicated models describing the transport and transformation of impurities as well as near-surface turbulence. Naturally, at the same time, using palaeoecological data for the assessment of long-term changes in the influx of atmospheric impurities, one must consider the complications arising with the postsedimentational changes and redeposition of sediments.

This study is the first attempt to compare modelled subsurface atmospheric fields with data of long-term deposition.

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