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REDUCTION OF SULPHUR DIOXIDE EMISSIONS AND TRANSBOUNDARY EFFECTS OF OIL SHALE BASED ENERGY PRODUCTION

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Introduction of a new combustion technology of oil shale – circulating fluidized bed (CFB) process in the Estonian and Baltic Power Plants (Narva PPs) – is an efficient way to reduce the emissions of SO_2 from electric energy production in Estonia. In 2004–2005, the exploitation of CFB boilers in the renovated power unit No 8 at the Estonian PP showed that the mean concentration of SO_2 in exhaust gases was 3–24 mg Nm^{-3}, whereas old-fashioned pulverized oil shale combustion (PC) boilers would have yielded 1920–3000 m Nm^{-3}. After the renovation of the other power unit (No 11) in the Baltic PP the total SO_2 emission from Narva PPs will decrease from 5.7–7.9 t per 1000 t burned oil shale (in 2000–2001) to 4 t (about 38,000 t $year^{-1}$) and in the renovated power units to 0.74 t. Compared to PC, CFB yields twice less emissions of NO_x (about 150 mg Nm^{-3} as NO_2). The modelling (AEROPOL model, developed in Tartu Observatory) indicated that air pollution levels in the surroundings of Narva PPs and transboundary air pollution (to Finland and Russia) would be reduced about 10 times as compared with the period 2000–2003.

Introduction

Oil shale is the most important fossil fuel in Estonia constituting about 87% of the country's total energy balance (motor fuels not included). The use of oil shale (sulphur content about 1.4–1.8%) in power plants is the

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primary source of sulphur dioxide emissions in Estonia. Oil shale processing, utilization of technological gases etc emit only ~15% of the total amount of SO_2. The transboundary effects of SO_2 emission from the two most powerful oil shale-fired power plants near the town of Narva (the Baltic and the Estonian Power Plants, together called *Narva PPs*, with capacities of 1390 MW_e and 1610 MW_e, respectively) are known in the neighbouring countries – in Finland and Russia [1–3]. Due to the decline in electricity production, the total emission of SO_2 diminished from 185–206 thousand t per year in 1990–1991 to the level of 70–85 thousand t per year in 1998–2004 (Fig. 1). At the end of 2001, renovation of Narva PPs began, with the introduction of a new combustion technology of oil shale – circulating fluidized bed (CFB) process [4, 5]. The old-fashioned pulverized oil shale combustion (PC) boilers in two power units with the maximum capacity of 200 MW_e (one in the Estonian PP (No 8) and the other in the Baltic PP (No 11)) were replaced with new efficient and environmentally sound CFB boilers with the maximum power of 215 MW_e. Besides this, in 1997–2002 all operational PC boilers (type TP-67 in the Baltic PP and TP-101 in the Estonian PP) as well as new CFB boilers are supplied with new efficient electrostatic precipitators to reduce the emission of fly ash. In May 2005, the exploitation of eight old worn PC boilers of type TP-17 in the Baltic PP was stopped.

The present paper deals with temporal changes and modelling of the air pollution level of SO_2 and sulphur deposition in pre- and post-renovation periods of Narva PPs. Changes in SO_2 transboundary effects and possible impact of the emissions from power plants on ecosystems after renovation are also discussed.

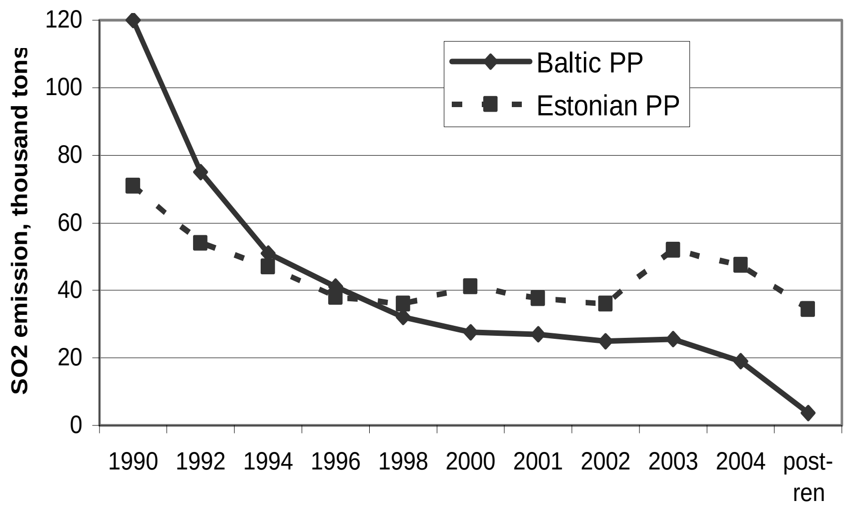

Fig. 1. Emission of SO_2 from Narva PPs in 1990–2004 and after renovation (post-ren) of two power units

Study Area and Methods

For modelling annual mean concentration fields of air pollution in the surface layer of the atmosphere and sulphur deposition loads in the pre-renovation period (1990–1991, 2000–2001) and after the renovation of boilers the AEROPOL Gaussian plume model developed at Tartu Observatory (Estonia) was used [6]. The applied modelling area (Figures 2, 3 and 4) of 270×230 km (~62,000 km^2) included areas influenced by emissions in Estonia (Narva–Tallinn–Jõgeva) and in neighbouring countries – up to 160 km to the north (southern coast of Finland from Helsinki to Hamina) and 80–100 km to the east (Leningrad District, Russia – Slantsy, Kingissepp, Primorsk) from the power plants. In calculations the wet deposition of SO_2 was treated as an exponential decay process with the time scale proportional to the square root of the intensity of precipitation (mm h^{-1}), and dry deposition velocity was set equal to 0.5 cm 7s^{-1}. Concentrations of SO_2 and NO_x in flue gases from CFB boilers were studied by the Gas Purification Laboratory of the Baltic PP, using the gas analyser Testo-350 and gas preparation block Testo-390 by the method of MM 02-2001/SET DIN 33962:1997 (Certificate from 15.04.02 No L084 of the Estonian Accreditation Centre). Data on SO_2 emission in the periods of 1990–1991 and 2000–2001 were obtained from the statistics of Narva PPs.

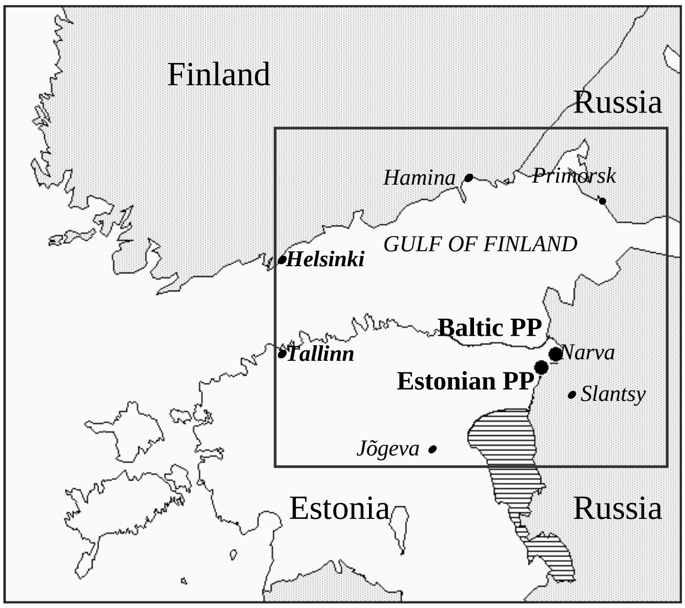

Fig. 2. Location of Narva PPs and the modelling area (square).

Results and Discussion

Reduction of emissions of pollutants

Table 1 shows changes in the emissions of pollutants in the pre- and post-renovation periods of Narva PPs.

Exploitation of the renovated power unit No 8 in the Estonian PP (operation of CFB was started in the end of 2003) has shown that the concentration of SO_2 in exhaust gases is 3–24 mg Nm^{-3} with the mean of 8 mg Nm^{-3} (lower than 200 mg Nm^{-3} is guaranteed by the renovation project), while old-fashioned pulverized oil shale boilers have yielded up to 3000 mg Nm^{-3} [7–9]. At the prognosed rate of oil shale consumption (9.4–9.5 million t per

Table 1. **Emission of pollutants and oil shale consumption in Narva PPs before and after the renovation of two power units, thousand tonnes per year**

Pollutants	Pre-renovation period 2000–2001 (mean factual data)			Prognosis after renovation of power units No 11 (Baltic PP) and No 8 (Estonian PP)		
	Baltic PP*	Estonian PP**	Total	Baltic PP*	Estonian PP**	Total
SO_2 1)	27.1	39.6	66.7	3.621	34.4	38.02
Fly ash 2)	21.9	24.1	46.0	0.333	3.628	3.961
NO_x (as NO_2) 3)	3.29	6.12	9.41	1.522	6.530	8.052
N_2O 4)	–	–	–	0.165	0.145	0.310
HCl	0.532	0.851	1.383	0.356	1.110	1.466
CO 4)	–	–	–	1.090	0.958	2.048
CO_2 5)	2 972	6 368	9 340	1 714	6 166	7 880
Oil shale consumption 6)	3 418	6 945	10 363	2 287 (1 750)	7 130 (1 538)	9 417
SO_2, tons per 1000 t oil shale 6)	7.92	5.7	6.43	1.58 (0.74)	4.82 (0.74)	4.03

* before renovation – TP-17 and TP-67 boilers, after renovation – CFB and TP-67 boilers.
** before renovation – TP-101 boilers, after renovation – CFB and TP-101 boilers.

Concentration of pollutants in flue gases (data from Narva PP-s):

1) *SO_2*: the TP-17 and TP-67 boilers 2200–3000 mg Nm^{-3} and TP-101 boilers 1920 mg Nm^{-3} (880–1360 mg MJ^{-1} [9]), the CFB boilers <200 mg Nm^{-3} (<75 mg MJ^{-1} accordingly to renovation project) and after renovation in power unit No 12 (2 TP-67 boilers) of Baltic PP – 1520 mg Nm^{-3} (570 mg MJ^{-1}).

2) *Fly ash*: TP-67 and TP-101 boilers with old electrostatic precipitators 2100–2800 mg Nm^{-3} and after installation of new electrostatic precipitators – 100–200 mg Nm^{-3}; in the renovated power units – 30 mg Nm^{-3} (11.4 mg MJ^{-1} accordingly to renovation project).

3) *NO_x*: all PC boilers – 240–320 mg Nm^{-3} (90–120 mg MJ^{-1}) [9], CFB boilers – <200 mg Mm^{-3} (<75 mg MJ^{-1}).

4) Only in renovated power units (CFB boilers): N_2O – 30 mg Nm^{-3} and CO – 200 mg Nm^{-3} [4, 5].

5) Emission of CO_2 in PC boilers 0.91 t t^{-1} per oil shale and CFB boilers 0.7 t t^{-1} per oil shale (decomposition of carbonates is lower).

6) In the brackets – corresponding data for renovated power units (CFB boilers).

year, starting from the end of 2005), the total SO_2 emission from Narva PPs will decrease from 5.7–7.9 t per 1000 t burned oil shale in 2000–2001 to about 4 tons after the renovation of two power units (total power of 430 MW_e). In the renovated power units (No 8 in the Estonian PP and No 11 in the Baltic PP – CFB type boilers) the emission of SO_2 is less – 0.74 t per 1000 t oil shale. As a result, the total annual SO_2 emission will be below 38,000 t per year (Fig. 1).

As a result of the installation of CFB boilers, the concentration of NO_x in flue gases has decreased about two times (to 150 mg Nm^{-3}) and the total NO_x emission by Narva PPs to 8,000 t per year, at the same time a low emission of N_2O is possible – 30 mg Nm^{-3} (about 310 t $year^{-1}$) (Table 1). Compared with the pre-renovation period, the emission of HCl will decrease by 20% and that of CO_2 by 22.5%, mainly owing to the reduced fuel consumption and lower decomposition of carbonates in the CFB boilers (Table 1). Diminishing of fly ash emission from 46,000 t $year^{-1}$ in 2000–2001 (about 68,000 t in 1995 [14]) to 3,961 t is a result of the installation of the new more efficient electrostatic precipitators in all the exploited boilers.

Changes in SO_2 transboundary air pollution

After the renovation of two power units the existing air pollution level in the nearest surroundings of the power plants (the town of Narva) as well as transboundary air pollution (to Finland and Russia) will decline essentially.

The model calculations (Fig. 3) show that after renovation of Narva PPs the annual mean concentration of SO_2 in the atmospheric air at the southern coast of Finland will not exceed 0.1–0.2 $\mu g\ m^{-3}$ and in Russia (up to 80 km east from Narva) 0.4–1 $\mu g\ m^{-3}$. Compared with the pre-renovation period (1990–2001), this pollution level is about 2–5 times less. For example, in Kotka and Virolahti (about 35 km east from Hamina) the annual average level of SO_2 was 2–3 $\mu g\ m^{-3}$ in 1999–2000 (data monitored by the Finnish Ministry of Environment). According to computed data, the share of Narva PPs in this SO_2 level might be <0.5 $\mu g\ m^{-3}$ (15–25%), being about 1 $\mu g\ m^{-3}$ during earlier years (1990–1991).

Calculated sulphur deposition loads are given in Fig. 4. In pre-renovation years (1990–2001), the sulphur deposition load caused by Narva PPs was in the range 0.05–0.12 mg S $m^{-2}\ day^{-1}$ at the southern coast of Finland, which constituted about 15–30% of the monitored loads (0.3–0.6 mg S $m^{-2}\ day^{-1}$ in 1985–1993) [10]. In 1990–1998, the daily deposition load of S in Virolahti increased up to 0.57–2.99 mg S m^{-2} [11], but this could not be caused by Narva PPs because their emissions of SO_2 decreased continuously (Fig. 1).

As a result of the exploitation of CFB boilers, the mean deposition of sulphur from flue gases of Narva PPs will drop below 0.02 mg S $m^{-2}\ day^{-1}$ in southern Finland and down to 0.02–0.5 mg S $m^{-2}\ day^{-1}$ in Russia (up to 80 km east of Narva), which is 2–10 times less than AEROPOL-calculated data for the pre-renovation period (2000–2001) (Fig. 4).

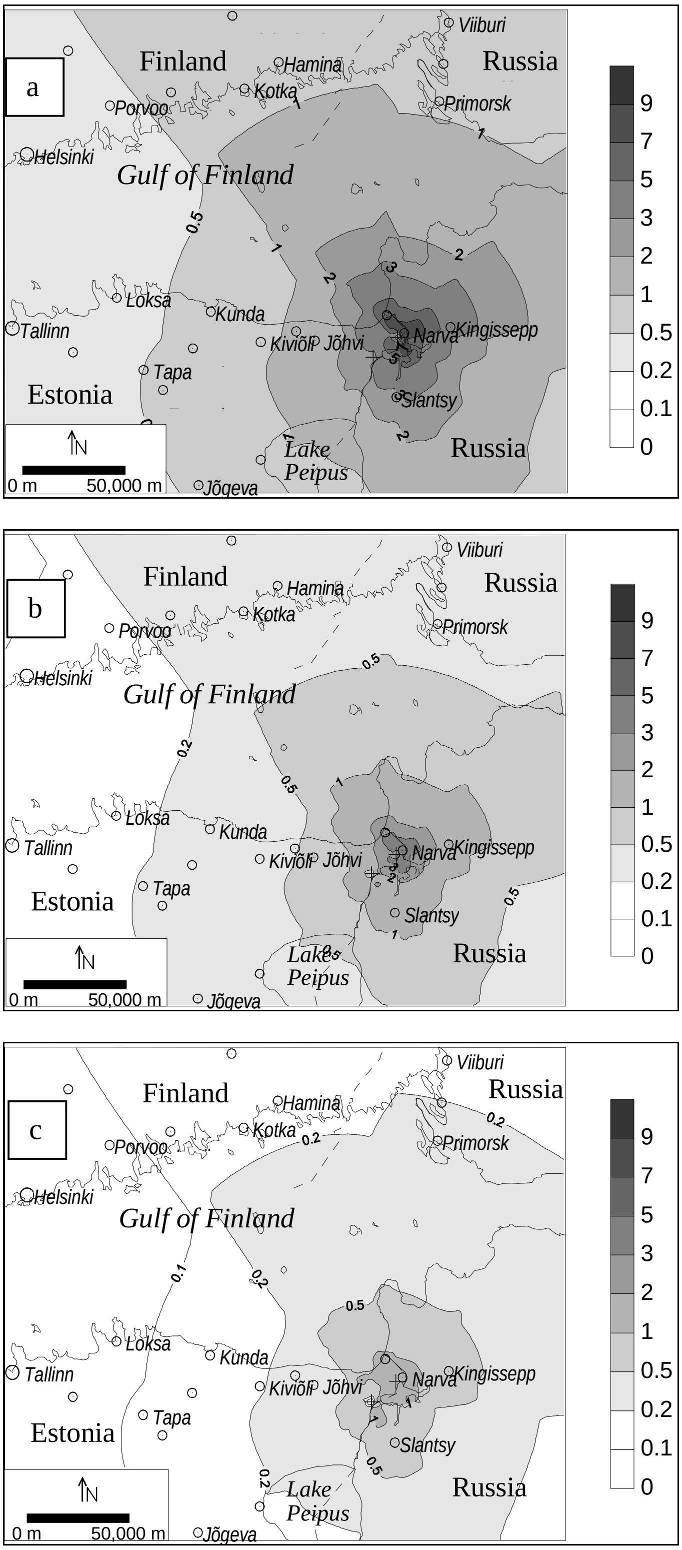

Fig. 3. Annual mean concentration of SO_2 in the surface layer of the atmosphere in the surroundings of Narva PPs, µg m^{-3}. Top-down: (a) during 1990–1991, (b) 2000–2001 and (c) after renovation of two power units. In calculations the following emission intensities of SO_2 with flue gases were used: in 1990–1991 – 6255–9170 g s^{-1}, in 2000–2001 – 4000–5530 g s^{-1} and after renovation of power units – 1890–3165 g s^{-1}.

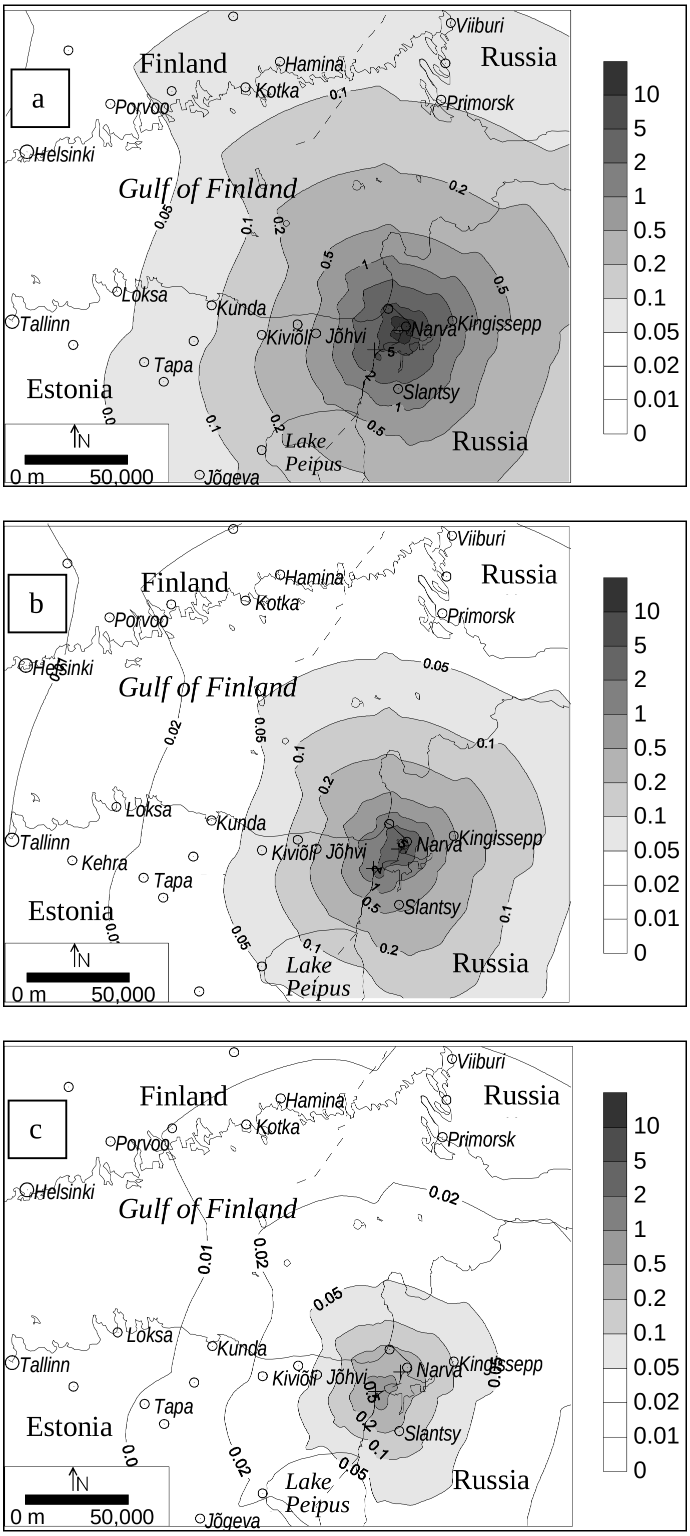

Fig. 4. Annual mean deposition of sulphur from emissions of Narva PPs, mg S m^{-2} day^{-1}. Top-down: (a) during 1990–1991, (b) 2000–2001 and (c) after renovation of two power units. Annual emission data of sulphur see Table 2.

Table 2. **Annual emission of sulphur from Narva PP*s* and deposition on the modelling area (Fig. 4), tonnes per year**

Source or deposition area	1990–1991	2000–2001	After renovation
Emission			
–as SO_2	95 563	39 704	19 913
–with fly ash*	10 363	2 947	440
TOTAL	105 926	42 651	20 353
Deposition			
–northern Estonia	2 125	779	229
–southern coast of Finland	178	68	31
–in Russia	4 197	1 324	379
–aquatory of the Gulf of Finland	1 806	618	213
TOTAL	8 305	2 789	852

* average 5.8% S in fly ash

Compared to the years of 2000–2001, in southern Finland and in the basin of the Gulf of Finland, the sulphur deposition will be reduced 2.2–2.8 times, in Russia (Leningrad District) about 3.5 times and in northern Estonia 3.4 times and more (Table 2).

Effects on the ecosystems

The concentrations of SO_2 in the ambient air and the deposition of S caused by Narva PPs will not endanger human health and ecosystems after renovation, as these concentrations will be 2–20 times and more below the critical level for ecosystems and significantly below the air pollution norms as well.

In order to prevent acidification, the sulphur deposition load must not exceed 1.37 mg S m^{-2} day^{-1} (5 kg S ha^{-1}) in Nordic countries [12]. This critical load is more than 50 times higher than the calculated S deposition caused by Narva PPs in southern Finland after renovation (0.01–0.02 mg S m^{-2} day^{-1} and below) (Fig. 4).

In north-eastern Estonia, the critical load of sulphur deposition with respect to terrestrial ecosystems is 9 mg S m^{-2} day^{-1}(33 kg S ha^{-1}) [13]. During the period of very high S deposition (up to 50 mg S m^{-2} day^{-1} before 1990), high deposition of base cations from oil shale fly ash fully buffered acid deposition in NE Estonia [14]. In the future, the effects of acidic pollutants on the ecosystems will become stronger in this region as a result of long-range transport. Installation of new electrostatic precipitators (1997–2002) and the renovation of power units will cause a decrease in the emissions of alkaline fly ash (CaO and other alkaline oxides) into the atmosphere by 92% compared to the years 2000–2001 (Table 1). Calculations have shown that daily deposition of Ca^{2+} and S (mg S m^{-2}) in the close surroundings of Narva PPs changed from S/Ca^{2+} = (0.5–20)/(1.1–50) in 1990 to the level of $S/Ca^{2+} = (0.05–1)/(0.045–1.1)$ after the renovation of two power units. This means that the role of base cations in NE Estonia is essentially decreasing. Thanks to the continuous decrease in alkaline deposition, pH of precipitation in NE Estonia has also been continuously falling after the 1990s: from 7.5–9.5 to 5.8–7.1 in 1994–1997 and to 5.16–5.7 in 2003 [15].

By the end of 2005, the calculated maximum value of short-term concentrations (1-hour average) of SO_2 in the air of Narva will decrease ~5.3 times (to 75 $\mu g\ m^{-3}$ at the limit value in the ambient air of 350 $\mu g\ m^{-3}$ [16]). The annual mean concentration (for example in Narva and Kurtna Landscape Reserve – about 40 km to SW from Narva) should not exceed 0.5–2 $\mu g\ m^{-3}$, which is essentially lower than the critical pollution level of SO_2 for ecosystems (for sensitive lichens, 10 $\mu g\ m^{-3}$ [14, 17]).

Conclusions

Installing the circulating fluidized bed (CFB) boilers based on a new combustion technology of oil shale in Narva PPs is an efficient way to reduce the emissions of SO_2 and NO_x from the production of electric energy in Estonia. The concentrations of SO_2 and NO_x in the flue gas from CFB power units are more than 100 and 2 times lower, respectively, as compared to the old PC-type boilers. This guarantees the fulfillment of the EU Directive 2001/80/EEC. Decline in SO_2 emissions from oil shale power plants in Estonia is an important factor in decreasing the acidification of lake water and forest soil in southern Finland as well as in Leningrad District in Russia situated to the east from the town of Narva. The renovation process of power units will be continued also in the future.

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