

<https://doi.org/10.3176/oil.1995.2.08>

## EMISSIONS FROM ESTONIAN OIL SHALE POWER PLANTS

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*Flue gas emissions from pulverized oil shale fired boilers of Estonian and Baltic power plants have been studied.*

*The concentrations of  $\text{NO}_x$ ,  $\text{CO}$ ,  $\text{C}_x\text{H}_y$ ,  $\text{HCl}$ ,  $\text{HF}$  and polycyclic aromatic hydrocarbons in flue gases have been found to be relatively low and acceptable according to German emission limits, for instance. Desulphurization degree of flue gases by  $\text{SO}_2$  absorption with ash has been found to vary depending on boiler type and operation conditions. In spite of significant sulphur capture (average values for different boilers in the range between 68 and 77 % of the initial sulphur content of the fuel),  $\text{SO}_2$  concentrations in flue gases remain still very high (up to  $2,600 \text{ mg/m}^3$ , 10 %  $\text{O}_2$ ). Very high concentrations of particles, especially at Estonian Power Plant (up to  $6,250 \text{ mg/m}^3$ , 10 %  $\text{O}_2$ ) have been detected. Heavy metal emissions were too high by the reason of particle control insufficiency as well.*

*Yearly emission estimates of this study support the former Estonian ones within the range of 10-15 %.*

### Introduction

Estonian power production is almost entirely based on oil shale combustion. There are two big pulverized oil shale power plants in operation near Narva - the Estonian Power Plant (EPP, 1610 MW) and

the Baltic Power Plant (BPP, 1390 MW), and two small ones in Kohtla-Järve (39 MW) and Ahtme (20 MW). In the top times (the eighties) about 25 million tonnes of oil shale were burnt in power plants every year. By now, power production and correspondingly oil shale utilization have dropped over two times.

The most of the energy related environmental protection problems in Estonia results from the operation of oil shale power plants. The atmospheric emissions of oil shale power plants in North-East of Estonia have caused concern in Estonia as well as in Finland. Main reason for the trouble is extremely high air pollution by fly ash and acidic components of flue gases, especially sulphur oxides, spreading often over the territories of neighbourhood countries - Russia and Finland. As a result, strong acidic deposition effects, e. g. in southern part of Finland, Virolahti, about 80 km to the North from the power plants, have been reported.

Estonian oil shale is rather specific low-grade fossil fuel (LHV about 8-10 MJ/kg), rich in sulphur (about 2 grams per MJ) and mineral matter (about 60 %). After the dissociation of carbonates (Ca and Mg) during combustion process, essential desulphurization of flue gases by ash sulphation in furnace and gas ducts takes place. Process of sulphur capture depends on many factors including boiler construction and operation conditions, fuel preparation, etc. The rates of sulphur capture range from 40 % up to 90 % under different conditions have been reported. The average factors, needed for the simple estimation of total sulphur oxides emission, are still under discussion.

Behaviour of oil shale in the combustion process and the emissions of environmental pollutants have been studied in Estonia for many years. Mainly sulphur and nitrogen oxides and fly ash emission measurements by conventional manual methods have been made. The information about the presence of minor harmful components (such as gas-phase heavy metals, hydrogen chloride, polycyclic aromatic hydrocarbons and others) in flue gases and about the correlation of emission of main pollutants with boiler operating conditions is rather short. To establish such correlations and to elucidate the effect of single factors, continuous measurements and special measurement technique are needed.

In order to achieve additional data on the emissions from Estonian and Baltic power plants based on the measurement technology used in Finland, a joint project of VTT Chemical Technology, Tallinn Technical University, Eesti Energia and IVO was set up. Extended measurements of emissions, the results of which are summarized in this paper were carried out in EPP in September 1992 and in BPP in May 1993 [1].

## Experimental

Flue gas components and their measurement methods are presented in Tables 1 and 2. Measurements by continuously operating instruments offered remarkable progress for the evaluation of emissions. Continuous measurements enable the examination of emissions in different boiler operation modes. During the measurement periods, in the course of



several hours daily, all the continuously operating instruments stored half-minute averages of the concentrations into a data logger.

For many flue gas components manual measurement methods are more reliable than continuously operating ones. Components to be measured and the methods used in this study are presented in Table 2. The disadvantage of manual measurement methods is that only the results of momentary, not varying, emission situations are gained.

A manual method for determination of total particle concentration is standardized in Finland (SFS 3866). Particles are collected isokinetically onto a filter and weighed after sampling. Their mass concentration in flue gases is calculated from the weight of the particles and the sample gas volume.

From the environmental and technical point of view not only the mass, but also the size of particles of flue gases is relevant. Sampling of size fractionated particles was carried out in two different systems. Coarse segregation to large and small particles (cut size 4-6  $\mu\text{m}$ , aerodynamic diameter) was achieved with cyclones and filters. In order to get detailed information on the size distribution of particles smaller than a few micrometers, the impactor sampling was included in the measurement period in 1993.

Elements which penetrated cyclones and filters (mainly heavy metals in gaseous state) were led to absorption bottles installed in the sampling line. 4 %  $\text{K}_2\text{Cr}_2\text{O}_7/20\%$   $\text{HNO}_3$  solution was used for mercury absorption and 30 %  $\text{HNO}_3$  for absorption of other metals. An elementary

**Table 1. Flue Gas Components Measured with Continuously Operating Instruments**

Component	Method
Sulphur dioxide $\text{SO}_2$	Fluorescence
Nitrogen oxides $\text{NO}$ and $\text{NO}_2$	Chemiluminescence
Hydrocarbons $\text{C}_x\text{H}_y$	Flame ionisation
Carbon oxides $\text{CO}$ and $\text{CO}_2$	Infrared absorption
Oxygen $\text{O}_2$	Paramagnetism

**Table 2. Flue Gas Components Measured by Manual Methods**

Component	Method
Total particles	SFS 3866 Isokinetic in-stack sampling
Heavy metals (particulate and gaseous)	VDI 3868 Predraft (adapted) Isokinetic in-stack sampling for particles (two fractions), liquid absorption for gaseous metals
Fractionated particles	Impactor sampling (Bernier Low-Pressure Impactor), five fractions
Polyaromatic hydrocarbons	Isokinetic in-stack sampling for particulate PAH, adsorption to XAD for gaseous PAH
Hydrogen chloride and hydrogen fluoride	Liquid absorption



analysis was made from cyclone and filter samples as well as from absorption samples. Samples were analysed with inductively coupled plasma atomic emission spectrometer (ICP-AES) and with inductively coupled plasma mass spectrometer (ICP-MS), both enabling multi-element analysis. An atomic absorption spectrometer (AAS) was used only for mercury determination and for checking some results of the ICP-AES and the ICP-MS. The ICP-MS analyses were made in the Geological Survey of Finland, the AAS analyses were carried out in the Technical University of Helsinki and in VTT Chemical Technology.

Hydrogen chloride (HCl) was absorbed by distilled water and its concentration was determined by ion chromatography. Hydrogen fluoride (HF) was absorbed by 0.1 M NaOH solution, and the samples were analysed using the ion selective electrode in VTT Chemical Technology.

Polycyclic aromatic hydrocarbons (PAH) were sampled using a sampling line in which the particles were collected on a filter, and the compounds from the gaseous phase were adsorbed using XAD-2 resin. PAH were extracted from the particles and XAD-2 resin with toluene. From the condensates, the compounds were extracted with hexane. Prior to the analysis, the toluene and hexane extracts were combined. The samples were cleaned by dimethylsulphoxide liquid-liquid extraction and silica gel column chromatography. The PAH were analysed by gas chromatography-mass spectrometry using the SIM-technique. As internal standards D<sub>10</sub>-pyrene,  $\beta,\beta'$ -binaphthylene and indeno[1,2,3-c,d]fluoranthene were used. PAH were quantified by using clean reference materials. The whole procedure after sampling was performed in VTT Chemical Technology.

Impactor sampling was performed with a modified Berner low-pressure impactor, with stages from 5 to 11, classifying the particles according to their aerodynamic behaviour into seven different size classes from approx. 0.3 to approx. 16  $\mu\text{m}$ . A cyclone with an aerodynamic cut size of approx. 7  $\mu\text{m}$  was used as a pre-separator for the cascade impactor. This meant that practically only the stages from 5 to 9 were in use in the cascade impactor. Only a small amount of larger particles penetrating the cyclone was collected on the two upper stages of the impactor. Impactor sampling and calculation of the final results were carried out by IVO Group.

In the EPP, seven boilers, out of twelve in operation, were under study. Emissions from the boiler No. 1B were measured thoroughly in order to get more data about the variation of emission levels under different process conditions. All samples in the EPP were taken from the flue gas duct between electrostatic precipitator (ESP) and the fan.

Co-ordination of the various sampling systems was arranged in the following way. Separate sampling systems were connected to the each of four ducts coming from the boiler at the same time. The first one was for continuous registration of flue gas main components ( $\text{O}_2$ ,  $\text{CO}_2$ ,  $\text{CO}$ ,  $\text{SO}_2$ ,  $\text{NO}$ ,  $\text{NO}_2$ ,  $\text{C}_x\text{H}_y$ ), the second one - for heavy metal probe sampling, the third one - for fractionated particle measurements and the fourth one - for PAH, HCl and HF probe sampling (Table 7). Total particle samples, on the contrary, were taken from all the ducts sequentially, because particles are not assumed to be distributed evenly within the flue gas ducts like



gaseous compounds. It was not possible to arrange sampling from all ducts for the other compounds. Consequently, the results concerning concentrations of fractionated particles, particulate heavy metals and particulate PAH are not as representative for the whole boiler as the remaining data. Mass flow measurements were made before each total particle measurement.

In the BPP, two small boilers, Nos. 9 and 15, and one big boiler, No. 19, were selected for measurements. Co-ordination of various sampling systems was arranged in the same way as in the EPP with the exception, that all samples were taken after the fan.

### Plant Operation during the Measurement

In the EPP, fifteen (out of sixteen) boilers (type TP-101) were in use. The steam production capacity of each boiler was 320 t/h (540 °C, 140 bar) and the conventional full load was 280 t/h. The measurements were mainly carried out during a conventional full-load operation of the boilers. Concentrations of the pollutants during underloaded conditions were examined on two boilers, on the boiler No. 2A (40 % of conventional full load) and on the boiler No. 6A (35 % of conventional full load).

BPP has seventeen small tangentially fired boilers (type TP-17) with a steam production capacity of 190 t/h, and eight big front wall fired boilers (type TP-67) with a steam production capacity of 280 t/h. The conventional values are 150 t/h and 260 t/h, respectively. The steam values are 520 °C /100 bar for TP-17 and 510 °C /130 bar for TP-67. The measurements were mainly carried out in the range of 65-100 % of a conventional full-load operation of both big and small boilers.

### Results

The average, minimum and maximum concentrations of SO<sub>2</sub> and NO<sub>x</sub> in the flue gases of different boiler types are shown in Table 3. For comparison, concentrations obtained formerly by Estonian measurement methods [5] are also represented. The results of VTT Chemical Technology are hourly averages, the Estonian results are momentary. Average emissions of SO<sub>2</sub> and NO<sub>x</sub> per input fuel energy unit are presented in Table 4.

CO concentrations have been below the accurate detection level (100 ppm) of the measurement device used, and are therefore not presented. C<sub>x</sub>H<sub>y</sub> concentration in all the measured ducts was predominantly below 10 mg/m<sup>3</sup> (as CH<sub>4</sub>-equivalent) and is therefore not considered further.

Particle concentrations measured in flue gases according to total particle sampling standard (SFS 3866) are shown in Table 5. According to former publications, the particle concentrations vary approximately from 800 to 1,600 mg/m<sup>3</sup> (10 % O<sub>2</sub>) [4], which are considerably lower than the results obtained by VTT Chemical Technology. Average emissions

**Table 3. Average, Minimum and Maximum Values of SO<sub>2</sub> and NO<sub>x</sub> Concentrations in Flue Gases (mg/m<sup>3</sup>)\*<sup>1</sup>**

	SO <sub>2</sub>		NO <sub>x</sub> (as NO <sub>2</sub> )	
	VTT Chemical Technology measurements	Estonian measurements* <sup>2</sup>	VTT Chemical Technology measurements	Estonian measurements* <sup>2</sup>
Estonian PP, boiler type TP-101				
Average	* <sup>3</sup> 1560	1280	* <sup>3</sup> 180	200
Min	770	650	130	140
Max	2280	2160	220	270
Baltic PP, boiler type TP-17				
Average	2600	1980	* <sup>3</sup> 200	160
Min	640	1290	160	90
Max	>3300	3070	210	190
Baltic PP, boiler type TP-67				
Average	* <sup>3</sup> 1680	1720	210	190
Min	1340	1550	190	150
Max	2570	2430	250	310

\*<sup>1</sup> In dry flue gas under normal temperature and pressure, reduced to 10 % O<sub>2</sub>-concentration.

\*<sup>2</sup> Tapupere, O., Ots A., 1989 [5].

\*<sup>3</sup> Average of the concentrations, correlated to the operation regime of the boilers.

**Table 4. Average Emission Factors of SO<sub>2</sub> and NO<sub>x</sub> per Fuel Energy Unit (mg/MJ)\***

	Estonian PP	Baltic PP	
	TP-101	TP-17	TP-67
SO <sub>2</sub>	820	1360	880
NO <sub>x</sub> (as NO <sub>2</sub> )	90	100	110

\* Calculations are based on an average value of 0.274 m<sup>3</sup>/MJ (stoichiometric dry flue gas volume per fuel energy unit) for fuels used in both plants during the years 1992 and 1993.

**Table 5. Average, Minimum and Maximum Values of Particle Concentrations in Flue Gases (mg/m<sup>3</sup>)\***

	Estonian PP	Baltic PP	
	Boiler type (number of measurements)		
	TP-101 (n = 31)	TP-17 (n = 11)	TP-67 (n = 8)
Average	2940	2250	2000
Min	1400	1730	1440
Max	6250	3320	2900

\* In dry flue gases under normal temperature and pressure, reduced to 10 % O<sub>2</sub>-concentration.



Table 6. Average Emission Factors for Particles per Fuel Energy Unit (mg/MJ)\*

Estonian PP	Baltic PP	
TP-101	TP-17	TP-67
1540	1180	1040

\* Calculations are based on an average value of 0.274 m<sup>3</sup>/MJ (stoichiometric dry flue gas volume per fuel energy unit) for fuels used in both plants during the years 1992 and 1993.

Table 7. The Results of Impactor Measurements

Stage	Cut size (μm)	Calculated concentration (mg/nm <sup>3</sup> )				Share of particles collected by impactor (%)*			
		1	2	3	4	1	2	3	4
Estonian PP, Boiler No. 6, TP-101									
Cyclone	7	-	-	906	1574				
11	17	6	50	10	51	0.6	4.4	1.1	6.0
10	8.0	56	91	61	100	5.4	7.9	6.9	11.8
9	3.9	526	531	501	402	50.4	46.2	56.5	47.5
8	2.0	296	304	197	194	28.4	26.5	22.2	22.9
7	0.95	81	89	47	58	7.8	7.7	5.3	6.8
6	0.50	49	59	41	24	4.7	5.1	4.6	2.8
5	0.28	29	26	29	18	2.8	2.3	3.3	2.1
5-11		1043	1149	887	847	100.0	100.0	100.0	100.0
5-11+C		-	-	1790	2420				
Baltic PP, Boiler No. 15, TP-17									
Cyclone	7	2135	2433	1664	1590	5	6	7	8
11	16.5	0	3	(3)	5	0	0.4	(0.4)	0.7
10	7.8	43	45	50	43	5.1	6.4	6.1	5.8
9	3.8	416	340	400	370	49.4	48.4	49.0	49.7
8	1.9	243	203	243	217	28.9	28.9	29.7	29.1
7	0.93	81	67	76	71	9.6	9.5	9.3	9.5
6	0.50	41	31	29	24	4.9	4.4	3.5	3.2
5	0.28	(17)	15	(15)	(15)	(2.0)	2.1	(1.8)	(2.0)
5-11		(842)	703	(817)	(745)	100.0	100.0	100.0	100.0
5-11+C		(2980)	3140	(2480)	(2336)				
Baltic PP, Boiler No. 19, TP-67									
Cyclone	7	1469	989	920	1470	9	10	11	12
11	16.6	6	4	3	3	0.8	0.5	0.3	0.2
10	7.9	36	44	50	60	4.6	5.6	5.2	4.9
9	3.9	401	384	476	611	51.6	48.9	49.8	49.8
8	1.9	223	217	286	377	28.7	27.6	29.9	30.7
7	0.94	72	66	78	110	9.3	8.4	8.2	9.0
6	0.50	(24)	46	38	40	(3.1)	5.9	4.0	3.3
5	0.29	(16)	(25)	26	27	(2.1)	(3.2)	2.7	2.2
5-11		(777)	(786)	956	1228	100.0	100.0	100.0	100.0
5-11+C		(2250)	(1780)	1880	2700				

\* Particles collected by the cyclone are not included.

Table 8. Particulate Heavy Metal Concentrations in Flue Gases\*1

Metal	Particle fraction*2	Estonian PP			Baltic PP		
		Concentration in different particle fractions, $\mu\text{g/g}$	Concentration in flue gas, $\mu\text{g/m}^3$		Concentration in different particle fractions, $\mu\text{g/g}$	Concentration in flue gas, $\mu\text{g/m}^3$	
			In different fractions	Total		In different fractions	Total
Pb	Fine	380	200	600	220	100	300
	Coarse	200	400		110	200	
Cd	Fine	4	2	6	1	1	2
	Coarse	2	4		1	1	
Zn	Fine	380	200	900	220	100	300
	Coarse	300	700		130	200	
Cu	Fine	20	10	40	20	10	30
	Coarse	10	30		10	20	
Ni	Fine	40	30	120	40	20	120
	Coarse	40	90		40	100	
Cr	Fine	50	30	130	80	40	140
	Coarse	60	100		60	100	
Co	Fine	10	5	15	10	4	24
	Coarse	10	10		10	10	
As	Fine	90	60	260	80	40	140
	Coarse	70	200		40	100	
Hg	Fine	n.d.		0.3	0.3	0.2	0.3
	Coarse	0.1	0.3		0.1	0.1	
Mn	Fine	340	200	1000	340	200	800
	Coarse	350	800		360	600	
Mo	Fine	20	10	50	10	10	30
	Coarse	20	40		10	20	
Se	Fine	n.m.			5	2	5
	Coarse	n.m.			1	3	
Tl	Fine	n.m.			10	3	6
	Coarse	n.m.			2	3	

\*1 In dry flue gas under normal temperature and pressure, reduced to 10 %  $\text{O}_2$ -concentration.

\*2 Cut size 6  $\mu\text{m}$  (aerodynamic diameter).

n.m. = not measured, n.d. = not detected.

of particulate matter per input fuel energy unit according to this study are presented in Table 6.

Results of impactor sampling are presented in Table 7. The impactor sampling was made only at one sampling point. Therefore in Table 7, the calculated total particle concentrations, and the concentrations of larger particles, calculated according to the cyclone samples, are not accurate or representative, because the concentration of larger particles varies a lot in the cross-section of the flue gas duct. The concentration of smaller particles, though, is relatively even in each volume unit of flue gas.

In Table 8, the average concentrations of heavy metals in the cyclone (coarse particles) and filter (fine particles) samples are presented. Heavy metal concentrations in flue gases and emission estimates have been



Table 9. HCl and PAH Concentrations in Flue Gases (mg/m<sup>3</sup>)\*

	Estonian PP		Baltic PP	
	TP-101	TP-17	TP-67	
HCl				
Number of measurements	<i>n</i> = 13	<i>n</i> = 2	<i>n</i> = 2	
Average	12	27	49	
Min	6	14	40	
Max	46	40	57	
Total PAH				
Number of measurements	<i>n</i> = 4	<i>n</i> = 1	<i>n</i> = 2	
Average	0.91	0.08	0.08	
Min	0.12		0.04	
Max	3.16		0.12	

\* In dry flue gas under normal temperature and pressure, reduced to 10 % O<sub>2</sub>-concentration.

Table 10. Yearly Emissions of the Narva Power Plants (tonnes per year) in 1992 and in 1993 (Measurements by VTT Chemical Technology in Estonian PP (1992) and Baltic PP (1993))

	Estonian PP			Baltic PP		
	1992	1993	1994	1992	1993	1994
SO <sub>2</sub>	60,400	48,600	48,300	78,000	51,000	49,600
NO <sub>x</sub> (as NO <sub>2</sub> )	6,900	5,400	5,300	7,000	5,000	4,900
Particles	112,300	91,700	91,000	74,000	51,000	49,600

calculated basing on the average heavy metal and the average total particle concentrations of different boilers. It was estimated that 22 % the particles were below 6 µm for all boilers. It is a strong simplification, but because of the lack of samples in the same cut size a more accurate estimation was not possible. All these factors make statistical representations of the heavy metal calculations quite low. HCl and PAH concentrations in the flue gases are presented in Table 9. HF concentrations were below their detection limit, of 0.1 mg/m<sup>3</sup>.

### Estimated Annual Emissions

It is common to evaluate the emissions of a certain plant on a yearly emission basis. Estimating the yearly emissions results in some uncertainties, such as:

- variations of the concentrations depending on plant operating conditions
- variations of the concentrations in different boilers
- variations of the fuel properties (heat value, etc.)

- inaccuracy in the determination of fuel properties (heat value, etc.)
- inaccuracy in the estimation of the emerging flue gas volume per fuel mass unit
- inaccuracy in the fuel input load.

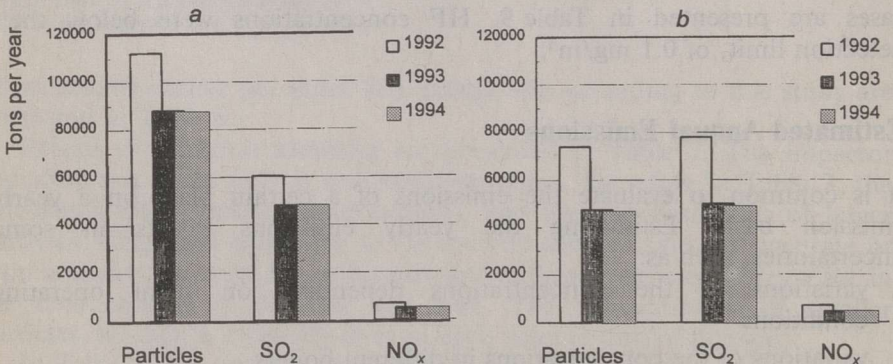
From data collected in this project the variations of pollutant concentration during the plant operation were investigated. For concentration of sulphur dioxide and nitrogen oxides the correlation with steam production capacity for all boiler types (TP-101, TP-17 and TP-67) was examined.

In case of TP-101, SO<sub>2</sub> correlation was positive but not strong (correlation coefficient  $r = 0.5$ ), for TP-67 SO<sub>2</sub> correlation was also positive and slightly stronger ( $r = 0.7$ ), for TP-17 no correlation was observed. Consequently, for estimations of yearly emissions of SO<sub>2</sub>, the average monthly steam production capacity was taken into account for boiler types TP-101 and TP-67.

For NO<sub>x</sub> concentrations a certain correlation with steam production capacity was found for boiler types TP-101 and TP-17 (for both  $r = 0.7$ ). For TP-67 no NO<sub>x</sub> correlation was observed. Consequently, for estimations of yearly emissions of NO<sub>x</sub>, the average monthly steam production capacity was taken into account for boiler types TP-101 and TP-17.

As the manual measurement method was used, the number of particle samples was too low to prove a correlation with steam production capacity. For estimations of yearly particulate emissions, average values were used.

The measurement period was not sufficient to check all the boilers. Consequently, results of the boilers measured had to be extrapolated to the unchecked boilers. According to the Estonian investigations [2], the differences of SO<sub>2</sub> emission between different TP-101 boilers are relatively small (within 11 %), whereas they vary remarkably for different TP-17 and TP-67 boilers, up to 2 and 2.5 times, respectively. Therefore, because of the previously stated reason, and the fact that in the BPP fewer boilers were checked, the estimations of the SO<sub>2</sub> emissions of the BPP are less exact than those of the EPP.



Estimated yearly emissions for (a) Estonian and (b) Baltic power plants



The yearly averages of fuel properties and total fuel input, as stated by Eesti Energia, were used for the calculations. In Table 10 and in Figure, yearly emissions of SO<sub>2</sub>, NO<sub>x</sub> and particles are presented.

On a yearly basis, desulphurization rates (weight per cent of the absorbed by ash sulphur) for different boilers were as follows (years and average sulphur contents of the fuel are shown in parenthesis):

- TP-101 77 % (1992) (1.47 % S)
- TP-17 68 % (1993) (1.5 % S)
- TP-67 75 % (1993) (1.5 % S)

Some estimations were made about the emission of heavy metals in two particle size classes. The estimations were quite rough because of the small number of samples with the same cut size. Nevertheless, the figures give some idea of the emission level. Estimations are presented in Table 11.

**Table 11. Heavy Metal Emissions of Estonian PP in 1992 and of Baltic PP in 1993 (kg per year)**

Metal	Particle fraction*	Estonian PP 1992		Baltic PP 1993	
		In different fractions	Total	In different fractions	Total
Pb	Fine	9000	26000	2000	6000
	Coarse	17000		4000	
Cd	Fine	90	240	10	30
	Coarse	150		20	
Zn	Fine	9000	35000	2000	7000
	Coarse	26000		5000	
Cu	Fine	400	1400	200	600
	Coarse	1000		400	
Ni	Fine	1000	4000	0	1400
	Coarse	3000		1400	
Cr	Fine	1000	6000	1000	3000
	Coarse	5000		2000	
Co	Fine	190	1190	100	300
	Coarse	1000		200	
As	Fine	2000	8000	1000	2400
	Coarse	6000		1400	
Hg	Fine	n.m.	10	3	6
	Coarse	10		3	
Mn	Fine	8000	39000	3000	16000
	Coarse	31000		13000	
Mo	Fine	500	2500	100	500
	Coarse	2000		400	
Se	Fine	n.m.		50	110
	Coarse	n.m.		60	
Tl	Fine	n.m.		100	170
	Coarse	n.m.		70	

\* Cut size 6 µm (aerodynamic diameter).  
n.m. = not measured.

## Discussion

In Finland, there have been public discussions concerning harmful emissions of two oil shale power plants near Narva. Estimates like 250,000 t of SO<sub>2</sub> p.a. have been commonly used. Estonian authorities (Tallinn Technical University, Eesti Energia) have reported values, approximately 60 % of the ones reported in Finland.

The measurements presented in this study support the estimations of Estonians within the range of 10-15 %. In 1992, the annual emission of SO<sub>2</sub> was 138,000 t according to VTT Chemical Technology, and 129,000 t according to Eesti Energia. Average concentrations reported by Estonians tend to be approximately 75-95 % of the measured average concentrations of VTT Chemical Technology, which partly explains the small difference in estimation results. Despite of significant sulphur capture (up to 80 %) by oil shale mineral matter constituents, sulphur dioxide concentration in oil shale power plant flue gases remains very high (average 1500-2600 mg/m<sup>3</sup>) compared, for example, to the limit of the German emission standard (300 mg/m<sup>3</sup>, 10 % O<sub>2</sub>) [3].

NO<sub>x</sub> emissions of oil shale combustion are much lower than typically measured in coal-fired power plants. The highest concentrations were measured for TP-67 boilers - 250 mg/m<sup>3</sup>, 10 % O<sub>2</sub> (as NO<sub>2</sub>), which is clearly below the German emission standard (590 mg/m<sup>3</sup>, 10 % O<sub>2</sub>).

According to this study, particle emission of the EPP is significantly higher than estimated previously. Estimates of VTT Chemical Technology and Eesti Energia for particle emissions converged better for the BPP. Particle emission of the EPP estimated by VTT Chemical Technology was two and a half times higher than the corresponding value declared by Eesti Energia in 1992.

The reasons of different estimations were studied during the measurement period in May 1993. Simultaneous measurements by both teams, VTT Chemical Technology and EPP ones, revealed that the results of the plant own team were systematically much lower than those of VTT Chemical Technology. It was agreed that the discrepancy in the obtained results was due to the aerodynamically faulty design of the measurement system used by the EPP.

All the particle concentrations measured in both plants (1440-6250 mg/m<sup>3</sup>, 10 % O<sub>2</sub>) were remarkably higher than the German emission standard (40 mg/m<sup>3</sup>, 10 % O<sub>2</sub>) permits. The particle size distribution, measured together with the total dust emission, points to the insufficient capacity of the electrostatic precipitators to remove huge particle load of the oil shale power plants. Even larger particles, usually removed relatively easily, penetrate the electrostatic precipitators in large numbers.

According to the German emission standard, the sum of Pb, As, Cd, Cr, Co and Ni in the flue gases of the big boilers (>300 MW, solid fuel) must not exceed 0.5 mg/m<sup>3</sup> (6 % O<sub>2</sub>), i.e. 0.4 mg/m<sup>3</sup> (10 % O<sub>2</sub>). According to this study the average particulate heavy metal concentrations in the flue gases of the BPP exceed the German emission standard moderately. In the case of the EPP the German standard is exceeded three



times. Remarkably high emissions of heavy metals are caused by low efficiency of the particle control devices.

Not only particulate, but also gaseous heavy metal emissions were measured within this project. Only the results for mercury, which exists in flue gases predominantly in its gaseous form, are presented in this paper. The average mercury concentrations in the flue gases (10 % O<sub>2</sub>) were 8 µg/m<sup>3</sup> and 5 µg/m<sup>3</sup> for the EPP and BPP, correspondingly.

Hydrogen chloride concentrations were lower than 100 mg/m<sup>3</sup> in both plants, this number being the emission limit for big boilers (>300 MW, solid fuel) according to the German standard [6].

Hydrogen fluoride concentrations were measured only in the EPP. Concentrations appeared to be lower than the detection limit, 0.1 mg/m<sup>3</sup>.

Polyaromatic hydrocarbon concentrations in the flue gases of the oil shale plants (0.08-3.16 µg/m<sup>3</sup>) were lower than the corresponding values in case of coal-fired power plants in Finland (5-60 µg/m<sup>3</sup>), for instance. Concentrations of such carcinogenic substances as benzo(a)pyrene and dibenz(a,h)anthracene, belonging to the emission standard class of 0.1 mg/m<sup>3</sup> limit in Germany, were under their detection limit (<0.01 µg/m<sup>3</sup>) in all except one measurement. The concentration of 0.06 µg/m<sup>3</sup> for benzo(a)pyrene was detected during the measurement period with exceptionally high total particle concentration in the EPP.

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Presented by A. Ots

Received March 13, 1995