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EMISSION OF FINE PARTICULATES FROM OIL SHALE FIRED LARGE BOILERS

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Emissions of fine particulates of grade PM2.5 and PM10 from different oil shale (OS) power plant (PP) boilers were studied. Pulverized (PF) and circulating fluidized-bed (CFB) boilers burning oil shale, biomass and retort gas were investigated. Particle emissions from OS boilers were found to be very fine. Over 90% of emitted particulate matter was in the size range of PM10 and 40-60% under the size of 2.5 μm. Distribution of fine particles by size was found to be depending on OS firing mode (PF or CFB). At CFB firing mode the share of the finest fraction PM2.5 was higher than that at PF firing mode. The distribution varied at the same type of different PF boilers, depending on the efficiency of boiler flue gas cleaning system and value of total suspended particulates' emission (TSP).

Introduction

According to different studies [1] fine particulates (PM2.5 and PM10) in the ambient air present serious health risk due to their ability to pass through the human respiratory organs directly to the lungs or even blood. Harmfulness of the fine particulates is caused by different hazard compounds (heavy metals, carcinogenic compounds, etc.) integrated into particles.

Different sources [2] confirm a significant correlation between PM level in the ambient air and mortality.

Another important feature, making fine particles an attractive research subject, is their transboundary effect – the ability to spread on long distances. Beside motor vehicles and local pollution sources, far large-scale combustion utilities (stationary emission sources) are playing important role in the quality of the ambient air.

Despite renovated electrostatic precipitators (ESP), PP combusting Estonian OS are still remarkable emitters of solid particulates (about 6240 tons in 2009/2010) [3], size distribution of these emissions was unknown until lately.

From beginning of 2008 relevant investigations at different OS fired boilers were started, the results of which are now reported.

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Experimental

Sampling method

The sampling method is based on the principle of impaction. The method is designed for stack measurements at stationary emission sources. The method allows gravimetric determination of concentrations of PM10 and PM2.5 emissions.

PM concentrations were measured with Johnas II cascade impactor. The standards CEN 13284-1 and VDI 2066 were followed [4, 5].

PM10 and PM2.5 mass concentrations are determined by size-selective separation of gas-borne particles basing on different inertia of particles. Flue gas is isokinetically sucked into the cascade impactor. The impactor separates particles above a specific aerodynamic diameter. The aerosol is accelerated in a nozzle and then deflected by 90°. Particles with greater aerodynamic diameters are not able to follow the flow lines of the gas due the mass inertia. They are separated on the collection plate. During sampling the particles are divided into three fractions with aerodynamic diameters greater than 10 μm, between 10 μm and 2.5 μm, and smaller than 2.5 μm.

In-stack sampling was provided isokinetically from the one point of the flue gas duct, usually before the flue gas fan, were flue gas temperatures remain in the limits of 140–200 °C (Fig. 1). The one-point sampling was reasonable because of relatively high TSP concentrations and therefore limited time of sampling. The sampling point location was chosen from the most homogeneous region of the velocity field based on grid measurement

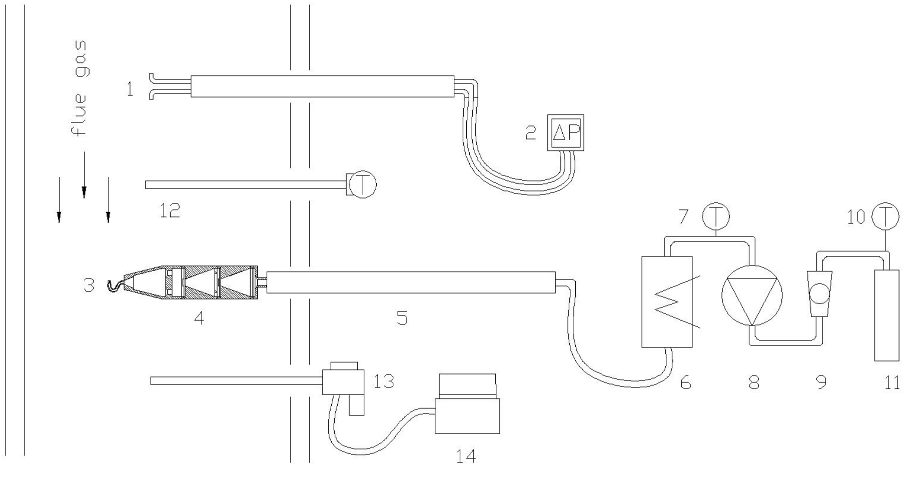

Fig. 1. General measuring setup.

1 – S-type Pitot tube, **2** – differental manometer, **3** – nozzle, **4** – Johnas II cascade impactor, **5** – heated sample probe, **6** – condensator, **7** – thermometer, **8** – pump, **9** – flow meter,**10** – thermometer, **11** – O_2 analyzer, **12** – thermocouple, **13** – gas analysis probe, **14** – FTIR gas analyser.

of flue gas velocities at the cross section (Fig. 2). A suitable nozzle diameter (usually 6–7 mm) of the probe and suction speed were chosen according to the flue gas velocity and isokinetic sampling presumption.

Sampled flue gas amounts (dry) varied from 0.5 to 0.8 Nm^3 and sampling times were from 20 to 40 minutes. The blank test, including the whole procedure without real sampling, was carried out for each test series.

Filters

To avoid chemical effects from acidic components of flue gas micro-quartz fibre filters Munktell type MK360 were used for sampling. The grade MK360 filters ability of catching 0.3 μm particles is better than 99.998%.

Preparation of filter sets was carried out at the laboratory. Filters were heated up to 160 °C for the removal of organic impurities at first. Before and after the test the filters together with holders were dried at 105 °C. After drying, filters were stored in desiccator for over 12 hours, until stable weight. Initial and final weighing of the filters was done at micro-analytical balance with the accuracy of 0.1 mg (readability – 0.01 mg).

Test objects and targets

The general target of the measurements was to determine emission factors of fine particulates, since only partial data about OS PM emissions was available [6]. The PM2.5 and PM10 emission factors were needed for the emission reports of PP and also for the National Emission Inventory (NEI).

Another interest was to find out how the fuel type, the power unit construction and operation mode affect emission of fine particles. Therefore the tests were carried out at different types of OS-fired boilers with addition of woody biomass and the retort gas from the OS processing factory.

Most of the tests were carried through on PF boilers type TP 101 of Eesti PP. The influence of OS and biomass (BM) co-firing on fine PM emissions was studied at Sillamäe PP at PF boilers type TP 35 [7]. Average furnace temperatures at OS CFB and PF boilers are usually in the range of 800 °C and 1400 °C, respectively.

Results and discussion

Reproducibility test was arranged at one of PF boilers to control the reliability of the sampling method, operator skills and influence of possible fluctuations in boiler run (Fig. 2). Single-point sampling was repeated in similar way from the same point of flue gas duct.

At first velocity field was determined at the sampling cross section and then the sampling point location was chosen (Fig. 3).

The results of the test revealed good concurrence, and standard deviation of the repeated measurements remained below 5% in the case of size fractions below 10 µm. Because of relatively low concentration of larger particles (>10 µm), the variation of results was higher, below 10% in that case.

An overview of the results of all tests at Narva PP boilers is given in Fig. 4.

A quite clear correlation between TSP value and size distribution of particulates can be seen in the diagram. The proportion of larger particles

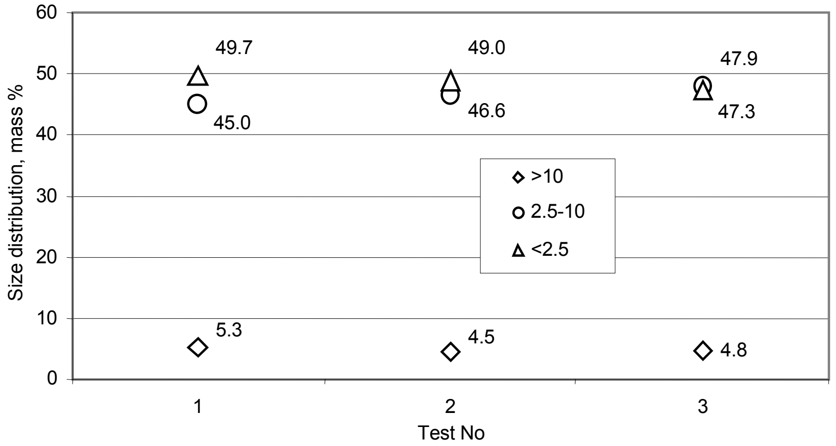

Fig. 2. Reproducibility test at PF boiler TP101 No 7B, ESP D.

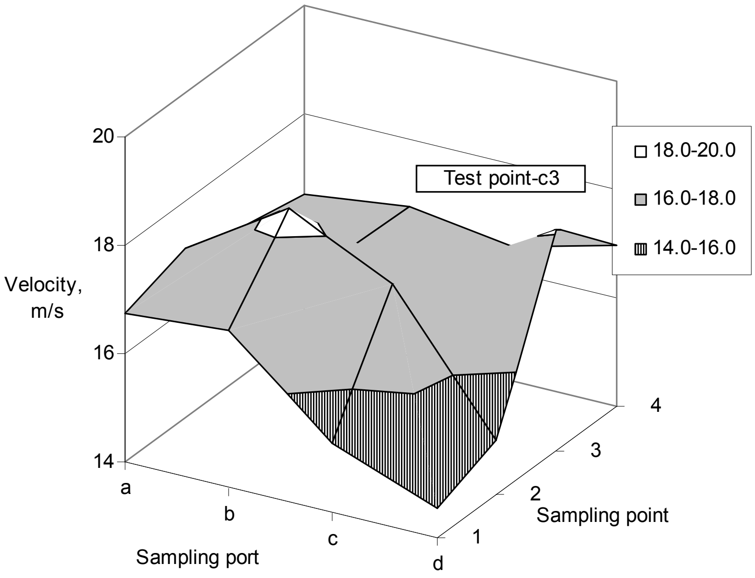

Fig. 3. Flue gas velocity field and the test point location during reproducibility test.

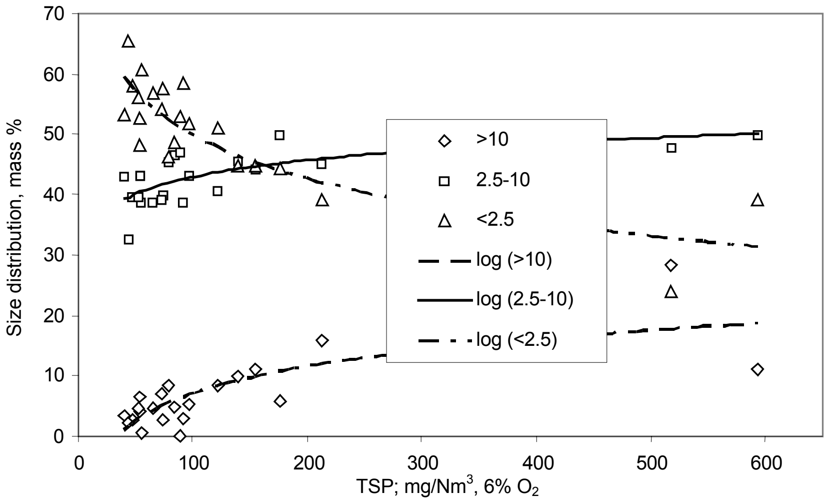

Fig. 4. Size distribution (by mass) of the emission of TSP from Estonian OS fired boilers.

increases concurrently with TSP emission. At usual TSP emission rates (~200 mg/Nm^3) the mass of fractions PM10 and PM2.5 is 90% and 45%, respectively. At lower TSP concentrations (<100 mg/Nm^3), the share of PM10 increases exponentially to 95% and that of PM2.5 to 50%.

In addition to the impactor measurements after the ESP collection of solid particles, at the inlet of one CFB boiler ESP was provided. Particle size distribution of the collected sample was analysed using the method of laser diffraction/scattering and sample dry dispersion. As a result data based on comparison of size distribution of fly ash particles at the inlet and outlet of ESP and relative cleaning efficiency of ESP are presented in Table 1 [8].

The trends of particle size distribution in Fig. 4 and PM concentrations at the inlet and outlet of ESP (Table 1) show that electrostatic precipitators most efficiently clean flue gases from larger OS fly ash particles. ESP efficiency at cleaning flue gas flow from particles of the size >10 μm is 63 times higher than from particles <2.5 μm.

Tests at two different CFB boilers showed little variations (<5%) in size distribution of the finest part (PM2.5) of TSP emissions. These differences remained in the limits of uncertainty according to repeatability test and can

Tab. 1. **Solid particulates at inlet and outlet of ESP of CFB boiler**

Sampling location	Unit	Particle size, μm			Total
		<2.5	2.5–10	>10	
ESP inlet	g/Nm^3, 6% O_2	7.8	22.5	29.8	60.1
ESP outlet	mg/Nm^3 6% O_2	21.3	17.1	1.3	39.7
Efficiency of ESP	%	99.9726	99.9924	99.9996	99.9934

be caused by fluctuations of boilers operation, fuel supply and feeding. The tested CFB boilers are of the same type, but they are located at different power stations.

Comparison of size distribution of TSP emissions from PF (TP101) and CFB boilers reveals that the mass share of fine particles (PM2.5) is about 10% higher in the case of CFB boiler (Fig. 5). Combustion conditions in CFB and PF furnaces, temperature distribution and residence times in gas passes are very different. At PF boilers also ESP-s with lower efficiency are used resulting in more large particulates at the ESP outlet and lower mass share of PM2.5 fraction.

Changes in PF boiler load and co-firing of OS with retort gas (share of gas <10% of thermal capacity) have little effect on size distribution of TSP emissions (Figures 6, 7).

TSP emission value itself is lower at co-firing with retort gas, because of lower ash load to the boiler.

In the case of lower boiler loads the velocities of flue gases remain almost the same, and smaller ash amounts in boiler do not have any significant effect on TSP emissions. The PF boilers are equipped with old fashioned fuel/air control system with limited possibilities to regulate combustion (uncontrolled air penetration).

TSP emissions and their size distribution at co-firing of Estonian OS and BM as sawdust was investigated at the boiler TP35 equipped with three-field ESPs of Sillamäe PP.

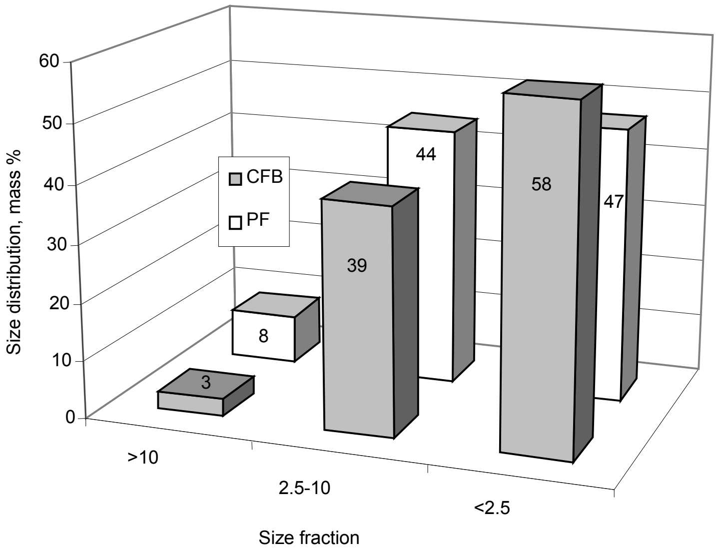

Fig. 5. Size distribution of solid particulates emission - CFB versus PF boilers.

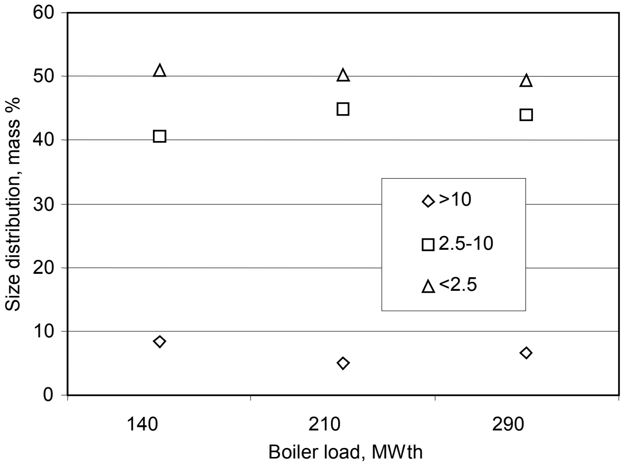

Fig. 6. Particle size distribution of TSP emission at PF boiler different loads.

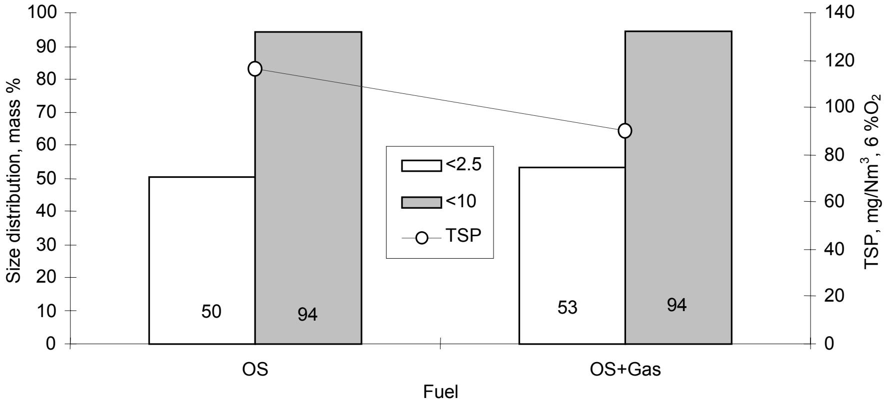

Fig. 7. PM2.5 and PM10 of PF boiler at OS and retort gas co-firing.

Measurements were made at combustion of four different fuels: pure OS; three mixtures of OS with woody BM, with the mass share of BM of 5, 10 and 15 per cent, respectively. From the results of these tests (Fig. 8) follows that the emission of TSP decreases with the increase of the share of BM, the fuel with less ash content. The average ash content of the fuel as received was 43% for OS (815 °C) and 2.8% for BM (550 °C).

Size distribution of the emitted solid particles is quite similar for all cases (Fig. 8). Majority of the particles mass (50–55%) remains between sizes of 2.5 and 10 μm, the share of particles below 2.5 μm is the next in the row (35–40%). The share of the largest particles (>10 μm) is the smallest (10–15%). The trend of the slight fall of the PM2.5 fraction with addition of BM was noticed.

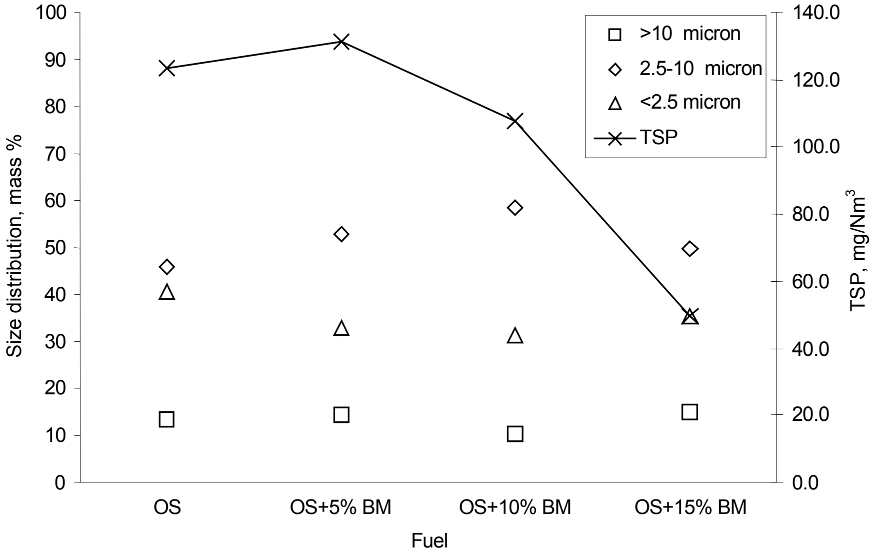

Fig. 8. Emission and size distribution of solid particles at OS and BM co-firing.

As a result a conclusion about similar size distribution of emission of solid particulates from PF boilers TP35 and TP 101 with the same TSP value can be drawn.

Comparison of emissions factors of fine particles of OS and solid fuels used in RAINS model is given in Table 2 [7, 9, 10]. Certainly the factors depend on combustion conditions also in the case of other solid fuels, and presented in Table 2 figures are the averages of very different data. In that sense Table 2 is having only an illustrating purpose.

It follows from Table 2 that PM2.5 emissions at OS firing are higher than at coal firing, remaining at the same level with the waste fuel, but are much lower than those for biomass. Emission of PM10 at OS firing is high and at the same level at co-firing with biomass.

Tab. 2. **Size distribution of solid particulate emissions at different fuels firing, mass %**

Solid fuel	Particle size fraction			
	PM2.5	2.5–10 μm	PM10	>10 μm
Coal	13	39	52	48
Derived coal	30	40	70	30
Biomass	93	3	96	4
Waste	60	30	90	10
Oil shale (PF)	30–59	40–50	80–95	5–20
Oil shale (CFB)	55–65	33–43	96–98	2–4

Conclusions

- Particle emissions from OS-fired large boilers are relatively fine – the share of PM2.5 is between 50–60% and that of PM10 more than 90% in the case of usual TSP values (~200 mg/Nm^3).
- Decrease in TSP value involves a relative increase in the finest fraction (PM2.5) and decrease in the coarsest fraction (>10 μm). The share of intermediate fraction (2.5–10 μm) stays almost unchanged.
- ESP cleans flue gas from particles over 10 μm more efficiently than from particles under 2.5 μm.
- Mass percentage of PM10 is the same for PF and CFB boilers but percentage of PM2.5 is higher for CFB boilers.
- Co-firing Estonian OS and fuels with lower ash content (BM and retort gas) results in lower TSP emission, but does not change relative shares of PM2.5 and PM 10.
- PM2.5 and PM10 total emissions from large OS boilers can be estimated based on regular TSP emission measurements of separate boilers and relevant mass shares of fractions.

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