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Electrostatic Precipitator Collection Efficiency and Trace Element Emissions from Co-Combustion of Biomass and Recovered Fuel in Fluidized-Bed Combustion

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Particle and trace element emissions from energy production have continuously been subject to tightening regulations. At the same time, not enough is known on the effect of different combustion processes and different fuels and fuel mixtures on the particle characteristics and particle removal device operation. In this investigation, electrostatic precipitator fractional collection efficiency and trace metal emissions were determined experimentally at a 66 MW biomass-fueled bubbling fluidized-bed combustion plant. The measurements were carried out at the inlet and outlet of the two-field electrostatic precipitator (ESP) at the flue gas temperature of 130-150 °C. Two fuel mixtures were investigated: biomass fuel containing 70% wood residue and 30% peat and biomass with recovered fuel containing 70% wood residue, 18% peat, and 12% recovered fuel. The particle mass concentration at the ESP inlet was 510-1400 mg/Nm³. Particle emission at the ESP outlet was 2.3-6.4 mg/Nm³. Total ESP collection efficiency was 99.2-99.8%. Collection efficiency had a minimum in particle size range of 0.1–2 μ m. In this size range, collection efficiency was 96-97%. The emission of the trace metals As, Cd, Co, Cr, Cu, Mn, Ni, Pb, Sb, Tl, and V was well below the regulation values set by EU directive for waste incineration and co-incineration.

Introduction

Fine particles emitted to the atmosphere have gained increasing attention lately because of the adverse health effects associated to fine particles in recent studies (1). Energy production by combustion is one of the major stationary sources of fine particles, and it has therefore been the subject of tightening regulations as regard to emissions. Trace elements are mainly found in the particulate phase at the particle removal device operating temperatures, with the exception of Hg and possibly Se (2, 3). However, many of them are volatile during combustion, and consequently, they may be enriched in the fine particle fraction that penetrates easily through the particle removal devices.

The European Union has set Directive 2000/76/EC to regulate the harmful emissions, including particles and 12 toxic trace elements, from waste incineration. The directive also applies to co-incineration of waste materials with other fuels. At the same time, the amount of waste is steadily increasing, and waste incineration including co-incineration is a viable option to reduce the amount of waste. Therefore, the issue of particle and trace element emissions from waste co-combustion needs to be addressed.

The particle formation and trace element behavior during combustion of solid fuels have been extensively investigated, and a fairly good description of these issues is available in the literature (4-8). The particle emission to the atmosphere is determined by the combination of the particle formation and transformations in a specific combustion process and fuel and the particulate removal system that is used. The solid fuel combustion-generated particle emissions typically consist of two types of particles: fine particles approximately $0.1-1 \, \mu \text{m}$ in diameter that are formed from the ash-forming species that are volatilized during combustion and residual ash particles larger than 1 μ m in diameter that are formed from mineral impurities in the fuels (4). In fluidized-bed combustion (FBC), also a third particle type is found that consists of fragmented bed material (9). The relative abundance of the different particle types depends on the fuel and combustion system as well as combustion conditions. Typically, for example, higher combustion temperature increases the amount of fine particles. Also, biomass combustion generally produces more fine particles than, for example, coal combustion due to the different mode of occurrence of the ash-forming compounds in the fuels. Many trace elements are relatively volatile during combustion and are consequently enriched in the fine particle fraction.

Electrostatic precipitators (ESPs) and fabric filters are commonly used to remove particles from the flue gases of power plants and waste incinerators when relatively low emission values are needed. The ESP is generally accepted as a reliable and efficient particulate control device with low operating and maintenance costs. It has also proved to be able to achieve the required emissions levels for most major applications. Fabric filters offer high collection efficiency for particles and can be combined with absorbents for gaseous emission control but have relatively high maintenance costs. Most of the particulate removal devices capture coarse particles efficiently, whereas fine particle collection efficiency depends on the device and the characteristics of the particles to be captured as well as gas conditions and other factors. Typically, ESPs have a penetration window in the particle size range of $0.1-1 \mu m$. In pulverized coal combustion, even 10% of the particles in this size range may penetrate the ESP (10-12). Fabric filter collection efficiency also decreases with decreasing particle size, but with a well-maintained fabric filter, the maximum penetration is typically significantly lower than with ESPs. Insignificant literature data are available on the fractional collection efficiency of ESPs or fabric filters in biomass combustion or on waste and biomass co-combustion. However, particle characteristics and other conditions are different from, for example, pulverized coal combustion; therefore, it can be expected that particle removal device operation may differ significantly. This, in turn, is critical in respect to the particle and trace element emissions to the atmosphere.

In this investigation, we determined the fractional collection efficiency of a two-field ESP during biomass combustion and biomass—waste co-combustion in a bubbling fluidized bed. In addition, we determined the emissions of

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TABLE 1. Ash Contents of Fuels and Chemical Analysis of Major Elements in Fuel Ash

element	wood residue (%)	refuse (%)	peat (%)	total in fuel no REF (%)	total in fuel with REF (%)
ash	1.5	10	3.0	1.9	2.8
Ca	27.5	12.9	12.6	20.6	18.2
CI	0.217	2.31	0.355	0.282	1.154
K	5.89	2.00	0.531	3.38	3.15
Na	0.883	2.97	0.156	0.543	1.65
S	0.299	1.62	2.23	1.20	1.25
Si	2.89	16.5	7.11	4.86	9.62

the trace elements that are regulated by EU directive on waste incineration. Mercury was not included in this investigation despite its importance while assessing trace element emissions from combustion processes because, at the temperatures of the typical particulate removal devices, mercury may occur both in the vapor phase and in the particulate form. This investigation concentrated only in the emissions in the particulate matter; therefore, mercury was not included in the study.

Methods

Plant Description and Fuels. Fine particle measurements were carried out at a 66 MW Forssa biomass-fueled CHP bubbling fluidized-bed plant between February 13 and February 16, 2001. The measurements were carried out upstream and downstream of the two-field ESP at the flue gas temperature of 130–150 °C. The particle mass size distribution was determined with Berner-type low-pressure impactor (BLPI), mass concentration was determined with a tapered element microbalance (TEOM) and BLPI, and number size distribution was determined with an electrical low-pressure impactor (ELPI). The values of process parameters and gas composition were collected continuously.

Two different combinations of biomass fuels were studied. During the first 2 d of measurements, 30% peat and 70% wood residue were burned. Then, the effect of high-chlorine fuel was tested by adding approximately 12% recovered fuel (REF) to 18% peat and 70% wood residue for the next 2 d. Numbers are the percentages from total fuel power. Sand was used as bed material. Even though the aim was to keep the power plant conditions as stable as possible during the measurements with the two fuel combinations, the load of the power plant varied slightly from day to day and even during the measurements. However, the gas concentrations were fairly stable during the measurements with O2 concentration before the economizer between 4.5 and 5.5%, CO concentration at 8-10 ppm, and SO₂ concentration very low at about 1 ppm. HCl concentration was below 1 ppm without REF and with REF was higher, up to about 15-20 ppm. Bed temperature was typically about 770-830 °C without REF feed and with REF was slightly higher at about 810-840 °C.

Samples of fly ash from ESP hoppers, bottom ash, sand, and fuels were collected periodically for analysis. Fuel samples were taken from the feeder line on the day of the measurements. For the analysis, the fuels were ashed at 500 °C. The ashes of wood residue, peat, and REF were analyzed by XRF, IC, and ICP-MS/AES. The analyses were carried out at the Geological Survey of Finland, which has been accredited according to Standard EN 45001 and ISO IEC Guide to use the aforementioned methods.

The ash contents and the concentrations of the major ash elements in the fuel ash are shown in Table 1. Chlorine content in REF (2.3%) is approximately 10 times higher than in wood residue and peat (Table 2). Also, as expected, many of the trace element concentrations are higher in REF than

in the biomass fuels. The REF-containing fuel mixture contains significantly more Cd, Cr, Cu, Pb, and Sb than the fuel without REF.

The trace element concentrations of the fuel ash, the ESPcollected ash, and the bottom ash are shown in Table 2. ESP 1 was collected from the first ESP field, and ESP 2 was collected from the second field. The specific surface area of ESP ash from the first field was 25.3 m²/g and from the second field was 14.0 m²/g. Typically, the coarsest particles and most of the mass are collected in the first ESP field. Scanning electron microscope (SEM) analysis of the fly ash particles showed that the coarse mode particles ($D_p > 1 \mu m$) were porous agglomerates whereas the fine mode particles ($D_p \le 1 \mu m$) were mostly single, close to spherical particles, or small agglomerates with only a few primary particles. This explains the somewhat surprising result of a larger specific surface area of the particles collected in the first ESP field as compared to the particles collected in the second ESP field. This result is consistent with earlier studies from fluidized-bed combustion of biomass where the coarse fly ash particles were found to have a very porous structure (13).

As, Cd, Mn, Pb, Sb, Tl, and V were more concentrated in the second ESP field than in the first ESP field with both fuel mixtures. Other trace metals were enriched either in the first or second field, depending on the fuel. Some of the trace element concentrations were significantly higher in the fuel ash than in the ESP-collected ash. This was presumably due to the heterogeneous nature of the waste fuel.

Aerosol Measurements. The fly ash particle mass size distributions at the ESP inlet and outlet were determined by collecting size-classified fly ash samples with an 11-stage, multijet compressible flow BLPI (θ). The collections were carried out by inserting the impactor inside the flue gas channel. To avoid overloading of the impactor, a precutter cyclone was used prior to the impactor at the ESP inlet to collect particles larger than 8 μ m. Thin, greased aluminum (Al) and polycarbonate (poreless Nuclepore) films were used as impaction substrates at both measurement locations. BLPI samples were taken at approximately the same time at the inlet and outlet of the ESP, although the measurement before ESP lasted 0.5–1.5 min and after ESP lasted 15–60 min. The measurement system configuration is presented in Figure 1.

Aerosol samples were analyzed for inorganic ions (SO₄, Cl, NO₃, Na, NH₄, K, Mg, and Ca) using two Dionex DX500 ion chromatographs. Samples were first wet with 0.5 mL of methanol and extracted with 4.5 mL of deionized water (Millipore Alpha-Q) as described by Teinilä et al. (14). The resulting solutions were mixed by rotating the test tubes for 10 min. For anions, a sample solution was manually injected through a 1000- μ L loop to an AG 11 guard and an AS 11 analytical column. The eluent for anions was 0.4-25 mM NaOH (gradient). A 300- μ L loop, a CG 12A guard column, and a CS 12A analytical column were used for cations. The eluent for cations was 20 mM methanesulfonic acid. The ions in both methods were detected by conductivity detector.

Elements in aerosol samples were analyzed using an ELAN 6100 ICP-MS instrument (PerkinElmer SCIEX). For ICP-MS analyses, samples were extracted in a mixture of concentrated HNO $_3$ and concentrated HF, following the method of Jalkanen and Häsänen (15). Analyzed elements were As, Cd, Co, Cr, Cu, Mn, Ni, Pb, Sb, Ti, and V. Rhodium was used as an internal standard. Laboratory filter blanks were made both for IC and ICP-MS to determine the backgrounds of analytes for sample handling and storage.

Number size distributions of the fly ash particles in the size range of $0.01-3~\mu m$ were determined at the ESP outlet with an ELPI (16). Prior to the ELPI, the flue gas was diluted with a porous tube diluter to quench the sample gas flow.

The total particle mass concentration at the ESP outlet was measured with a tapered element oscillating microbal-

TABLE 2. Trace Element Contents in Fuel Ash, ESP-Collected Ash from Fields 1 and 2, and Bottom Ash during Combustion with and without REF Addition

element	wood residue (ppm)	refuse (ppm)	peat (ppm)	total in fuel no REF (ppm)	total in fuel with REF (ppm)	ESP 1 no REF (ppm)	ESP 2 no REF (ppm)	ESP 1 with REF (ppm)	ESP 2 with REF (ppm)	bottom ash no REF (ppm)	bottom ash with REF (ppm)
As	3	37	108	52	38	13	43	12	34	6.9	5.9
Cd	6.0	24	1.9	4.1	13	1.5	3.6	2.4	6.7	0.017	0.033
Co	15	67	78	44	50	4.2	3.6	2.0	4.5	1.6	6.9
Cr	90	1 020	143	115	505	24	40	67	55	14	16
Cu	111	2 800	245	174	1 310	390	320	300	480	140	na
Mn	14 100	1 650	6 760	10 700	7 240	1 300	3 200	1 800	3 400	730	700
Ni	95	209	138	115	153	8.8	27	53	25	4.2	4.9
Pb	128	1 100	40	87	533	35	61	60	160	14	3.2
Sb	0	79	0	0	34	3.5	8.8	11	40	2.0	4.2
ΤI	0	0	0	0	0	0.45	0.70	0.47	1.4	0.062	0.032
V	15	52	350	172	97	27	60	24	40	9.1	8.1

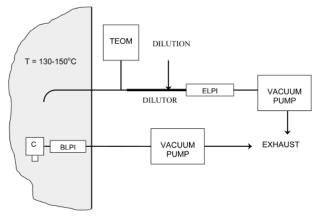


FIGURE 1. Schematic diagram of the experimental setup. A precutter cyclone (C) was only used at the ESP inlet in combination with BLPI.

TABLE 3. Particle Mass Concentrations at the ESP Inlet and Oulet As Determined by Low-Pressure Impactors with and without REF Feed

		particles <	particles < 0.5 μ m		
	fuel	(mg/Nm ³)	(%)	(mg/Nm ³)	
inlet	no REF	26	3.1	830	
	no REF	25	4.9	510	
	no REF	40	5.4	740	
	with REF	85	6.1	1 400	
	with REF	62	6.2	1 000	
	with REF	72	9.0	800	
outlet	no REF	1.1	22	5.1	
	no REF	0.79	23	3.4	
	no REF	1.4	24	5.6	
	with REF	3.1	49	6.4	
	with REF	0.92	40	2.3	

ance (TEOM) (17). TEOM samples were undiluted, and the measurements lasted less than 30 min each.

Results

Fly Ash Particle Size Distributions. Fly ash particle mass concentration at the ESP inlet was 510–830 mg/Nm³ without REF feeding and 800–1400 mg/Nm³ with REF (Table 3). The outlet concentrations as determined with low-pressure impactors were 3.4–5.6 and 2.3–6.4 mg/Nm³ without and with REF feeding, respectively. The continuous mass monitor TEOM gave outlet concentrations of 4.5–9 mg/Nm³ with both fuels The lowest value for the outlet concentration with REF feeding was measured when the boiler load was significantly lower than typically during this test campaign.

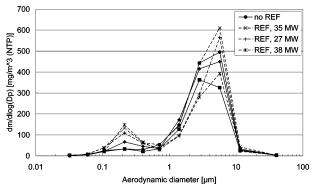


FIGURE 2. Particle mass size distributions at the ESP inlet as determined with low-pressure impactor. The particles collected in the precutter cyclone are not included in the figure.

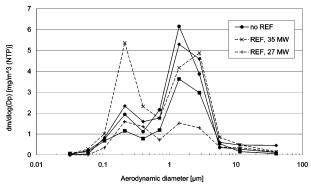


FIGURE 3. Particle mass size distributions at the ESP outlet.

The typical boiler load during the measurements was 36-44 MW_{fuel}, whereas during the outlet sampling, which gave the low mass concentration of 2.3 mg/Nm³, the boiler load was only 27 MW_{fuel}.

Fly ash consisted of two distinctly different particle modes at both the ESP inlet and the ESP outlet (Figures 2 and 3). A fine particle mode appeared at the particle size of 0.1-0.4 μm with both fuels, and a coarse particle mode appeared at the particle size greater than 1 μm . Mass concentration of the fine mode was significantly lower without REF than with REF, with concentrations of 25–40 and 62–85 mg/Nm³, respectively, at the ESP inlet. The fine particles contained on average 4.5% and 7.0% of the total particle mass at the ESP inlet, without and with REF, respectively. At the ESP outlet, the fine mode contained 23% and 44% of the total particle mass, without and with REF feed, respectively.

The enhanced fine mode fraction at the ESP outlet was caused by the less efficient particle collection of the ESP at the fine mode particle size range than in the coarse particle

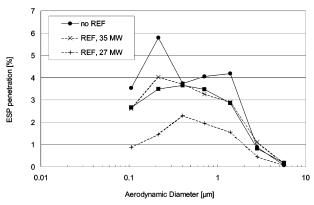


FIGURE 4. ESP penetration as a function of particle size as determined with simultaneous low-pressure impactor measurements.

sizes. It can also be seen from Table 3 that with certain fuels—especially ones with high chlorine contents—a significant fraction of the particle emission comes from the fine mode particles. In this case, almost half of the particle emission was caused by fine mode particles during REF firing.

The main components in the fine mode particles, as determined with the IC and ICP-MS analysis, were Na, K, and SO_4 without REF feed and Cl, K, Na, and SO_4 with REF feed. The increased fine mode particle concentration with REF was mainly due to the higher chlorine content of the fuel. Chlorine is known to increase the amount of alkalis in the vapor phase in post-combustion conditions. The fine particle composition agrees well with earlier studies where the fine mode particles in biomass combustion have been found to contain mainly alkali salts that have condensed from the gas phase (8, 9). The coarse residual ash mode showed little variation with the two different fuels.

ESP Collection Efficiency. ESP collection efficiency as a function of particle size was determined with simultaneous low-pressure impactor measurements in the particle size range of 0.1–6 μ m. Particle penetration through the ESP is defined as

penetration =
$$[100 \times \text{concn}(\text{outlet})/\text{concn}(\text{inlet})]\%$$
 (1)

ESP collection efficiency is then

collection efficiency =
$$(100 - penetration)\%$$
 (2)

The total collection efficiency of the ESP was 99.2-99.4% without REF feed and 99.6-99.8% with REF feed. The collection efficiency was 99.9% or greater for particles larger than 5 μ m (Figure 4). However, the collection efficiency decreased (penetration increased) with decreasing particle size. The collection efficiency was lowest—and penetration highest—at the particle size of about $0.2-0.5 \mu m$. At this size range, the penetration through the ESP was approximately 3–4%. At particle sizes smaller than 0.2 μ m, penetration seemed to decrease again with decreasing particle size. This is consistent with earlier results from ESP collection efficiency at pulverized coal-fired boilers, where a clear penetration window has been detected between 0.1 and 1 μ m (12). However, in this study, the mass collected in the smallest impactor stages was so low that no reliable penetration curve could be determined for particle sizes smaller than 0.1 μ m. More detailed measurements using, for example, number size distribution determinations would be required to confirm the shape of the penetration curve below 0.1 μ m. ESP penetration of ions (Cl, SO₄, Na and K) followed pretty well the penetration of the total mass of the particles.

The effect of ESP cleaning and soot-blowing in the boiler on the particle emissions was studied with the continuous

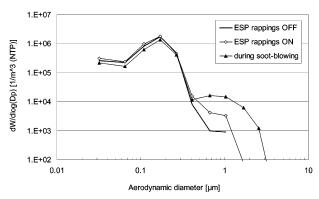


FIGURE 5. Particle number size distributions at the ESP outlet as determined with ELPI during addition of REF. The effect of ESP cleaning (rapping) and soot-blowing in the boiler.

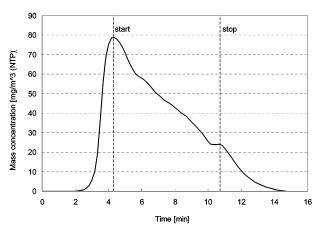


FIGURE 6. Mass concentration at the ESP outlet during soot-blowing in the boiler as determined with the continuous mass monitor TEOM. "Start" and "Stop" in the figure denote the starting and stopping time of the TEOM measurement.

mass monitor TEOM and with ELPI. ESP cleaning (rapping) was found to have limited effect on the emission. In the measurements without REF, the mass concentration increased from 5 to 8 mg/Nm³ and with REF increased only from 9 to 10 mg/Nm³. There was a slight increase in the concentration of particles larger than 0.4 μ m seen in the ELPI measurements (Figure 5).

Soot-blowing in the boiler had a more pronounced effect on the emission. The soot-blowing is usually carried out once a day in the boiler; each soot-blowing sequence lasting approximately 1 h. A TEOM measurement was started 10 min after the beginning of the soot-blowing when REF was included in the fuel mixture (Figure 6). "Start" in the Figure 6 denotes the starting time of the TEOM measurement. The mass concentration at the ESP outlet at the start of the measurement was almost 80 mg/Nm³, about 10-fold the normal emission. The mass concentration decreased rapidly during the soot-blowing, and after less than 10 min measurement (20 min from the start of the soot-blowing), the concentration had dropped to 25 mg/Nm3. However, during the whole soot-blowing period the mass concentration remained well above the normal, "non-soot-blowing" value of less than 10 mg/Nm3.

ELPI measurement (Figure 5) was started about 30 min after the beginning of the soot-blowing. It shows substantially increased particle number concentration for particles above the size of 0.4 μ m.

Emission of the Trace Elements. The emissions of trace elements as determined by impactor measurements were very low for both fuels (Table 4). The particle emission limit values set by the EU directive for waste incineration are 50

TABLE 4. Trace Metal Concentrations (NTP) in the Particles at the ESP Outlet

element	emission, no REF (µg/Nm³)	emission, with REF (µg/Nm³)
As Cd Co Cr Cu Mn Ni Pb Sb TI V	0.79 0.040 0.39 7.2 2.8 57 2.1 1.1 0.035 0.032 0.78	0.31 0.11 0.23 17 4.2 14 6.1 3.2 0.61 0.037 0.22
$\begin{array}{c} \text{Cd} + \text{TI} \\ \text{As} + \text{Co} + \text{Cr} + \text{Cu} + \text{Mn} + \\ \text{Ni} + \text{Pb} + \text{Sb} + \text{V} \end{array}$	0.072 72	0.15 46

 $\mu g/Nm^3$ for the total of Cd + Tl and 500 $\mu g/Nm^3$ for the total of As + Co + Cr + Cu + Mn + Ni + Pb + Sb + V. The measured values of 0.072 and 0.15 $\mu g/Nm^3$ for Cd + Tl and of 72 and 46 $\mu g/Nm^3$ for the other metals were well below the limit values. Mercury emissions were not included in this investigation.

The measured mass concentration in the sample that was chemically analyzed at ESP outlet with REF was quite low as compared to other mass concentrations from ESP outlet (2.3 mg/Nm³ as compared to 6.4 mg/Nm³). This was presumably due to the low and decreasing load during this measurement. However, even considering the typical three times higher mass concentration and, consequently, three times higher emission of trace metals, the emission values would be well below the limit values set by the EU directive for waste incineration and co-incineration. Also, in Table 4, the emission values for Cr and Ni are slightly overestimated as there was some contamination of these elements in the samples because of the use of stainless steel impactor material. The real emission for these two elements would be even lower than measured in these samples. Despite the low total emission value measured during REF feeding, presumably due to the low boiler load, the emission of Cd, Cr, Cu, Ni, Pb, and Sb clearly increased with REF addition to the fuel.

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