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Chemical composition and size of particles in emissions of a coal-fired power plant with flue gas desulfurization

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ABSTRACT

Globally more than a quarter of the total primary energy supply is based on coal combustion. The emissions of coal-fired power plants (CFPPs) are regulated in many industrialized countries and therefore power plants use cleaning techniques to minimize emissions such as sulfur dioxide (SO_2) and particles. In this study, the particulate emissions from coal combustion were investigated at a CFPP (506 MW) used for combined heat and power production in Helsinki, Finland. Fine particle samples (PM_1) were collected after electrostatic precipitator before the desulfurization plant (DSP), including flue gas desulfurization unit (FGD) and baghouse filters, and simultaneously in the smokestack to study the influence of DSP to particulate mass and chemistry. The DSP removed over 97% of particle mass in flue gas. Trace metals were removed efficiently but contribution of some ionic compounds increased in the FGD process. The particle properties were studied in more detail in the smokestack including particle size distribution measurements and size-segregating sampling to study chemical composition and morphology of particles. The particulate emissions from the CFPP were relatively small, consisting mainly of products and reagents of the FGD process (e.g., $CaSO_4$, NaCl) and partly of the primary emissions from the coal combustion (e.g., mineral ash and reaction products of gas phase components). The maximum in particle volume was detected at 0.68 μm. PM_1 contributed on average 62 ± 5% to PM_{10} mass. Besides particulate matter, also the gas-phase emission of mercury was studied because coal combustion is one of the major sources of mercury found in the environment. The mercury emissions were within the proposed limits in the EU.

1. Introduction

Coal is still one of the major fuels used in energy production despite awareness of the potential adverse environmental and health impacts, and high contribution to CO_2 emissions and therefore to global warming. In 2010 about one-fourth of

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the total primary energy supply of the world and one-third in the EU was contributed by coal (IEA, 2012). The major attention has been paid to CO_2 emissions of the coal combustion but coal-fired power plants (CFPPs) may be also significant sources of other air pollutants such as SO_x, NO_x, particulate matter (PM) and trace pollutants (Córdoba et al., 2012a). Emissions influenced by CFPPs vary depending on the coal quality, combustion technique and cleaning methods. In Europe, the emissions are regulated by the European Commission's Large Combustion Plants Directive (directive 2001/80/EC), which set requirements for quantity and quality of the emissions from power plants.

Emissions of CFPPs can be reduced using advanced combustion technologies such as low-NO_x burners and flue gas recycling. In addition, electrostatic precipitators (ESP), flue gas desulfurization (FGD) techniques and baghouse filters are used to reduce PM and sulfur dioxide (SO_2) emissions. ESP is usually used as a first particle removal technique after boiler. The ESP system captures a major part of the particles (ca. 99% of particulate mass according to Helble, 2000). After ESP, FGD technology is used to abate SO_2 emissions. Based on the study of Córdoba et al. (2012b), the FGD system can remove over 80% of primary gas-phase emissions (e.g., SO_2, F^-, Cl^-, B, and Hg) from the flue gases. Furthermore, FGD can be followed by baghouse filters that remove most of the end product dust of the FGD system and primary particles that have penetrated ESP. The use of both ESP and FGD with a baghouse decreases remarkably the gaseous and particle emissions of coal. The flue gas emissions after cleaning processes are affected by the chemicals used in the desulfurization process and include only a fraction of the primary emissions caused by coal combustion.

Particles emitted from coal combustion can be formed through transfer of inherent mineral matter into PM without any phase change (coarse particles) and through nucleation, condensation and coagulation of vaporized species (ultrafine and fine particles) (e.g., Ninomiya et al., 2004; Yoo et al., 2005). Toxic elements have been found to be enriched in ultrafine and fine particles (Kauppinen & Pakkanen, 1990; Yoo et al., 2005). In addition, Helble (2000) predicted metal enrichment to be dependent on metal volatility on particles. Hazardous particles are enriched in the size range that has a higher probability to penetrate through common flue gas cleaning equipment, such as ESP and baghouse filters, than the bulk of coarse fly ash particles formed in the combustion process. Characteristics of particle emissions originated from coal combustion depend on the chemical composition and properties of the coal used (Kauppinen & Pakkanen, 1990). Mineralogical properties of coal vary remarkably depending on the origin (Goodarzi, 2002).

Coal includes toxic elements (e.g., trace metals such as Cu, Fe, Hg, Ni, V, Zn) that are released into the atmosphere as gas and particle emissions causing adverse health effects (e.g., Córdoba et al., 2012a; Huggins et al., 2004; Lighty et al., 2000; Linak et al., 2000). In addition to chemical composition, particle size has been identified to be a potential factor to human health; especially fine particles are found to correlate with various health impacts (e.g., Knol et al., 2009; Laden et al., 2000). Hazardous elements released in coal combustion can also result in environmental problems such as pollution of soil and groundwater, especially in the case of disposal of solid by-product ashes produced by coal combustion.

Hg is one of the major concerns of coal combustion emissions. Coal burning is estimated to be one of the major sources of anthropogenic Hg emissions even though with high uncertainty (Pirrone et al., 2010; UNEP, 2013). Hg emissions from fossil fuel burning account for about 45% of the total amount of Hg emissions (Rallo et al., 2012). Hg is a toxic and hazardous metal pollutant that accumulates in the food chain, inducing diseases and even can be fatal (Tchounwou et al., 2003). Hg emissions of coal combustion are regulated in the USA and the EU has adopted the European Mercury Strategy. This strategy aims to reduce human exposure to Hg through reducing the emissions, supply and demand of Hg in the EU. The revision of the mercury strategy puts strong pressure on more stringent emission regulations of Hg from CFPPs. Proposals for the Hg emission limits of the coal combustion are done and the suggested emission limit value is in the range of 1.5–3.0 $\mu g\ Nm^{-3}$. Starting from 2016 coal-fired power stations have to measure Hg emissions periodically on a yearly basis.

In this study, the emission characteristics of a coal-fired boiler were investigated with respect to the chemical and physical properties of particle emissions. The focus of the study was in particles that are emitted to the atmosphere from the CFPP after desulfurization unit and baghouse filters. However, in order to see the changes in particle characteristics caused by the desulfurization process, particles were studied before the desulfurization unit. The chemistry of fine particles was studied from emissions before and after desulfurization. The physical and chemical size distributions and mass concentrations of particles, as well as the size, shape and composition of the selected particles were studied after desulfurization (i.e., from the smokestack). Because of the increasing interest to regulate Hg emissions of coal combustion, gaseous Hg emissions were determined from the smokestack emissions.

2. Methods

2.1. Measurement site and fuel description

The measurement campaign was carried out in a CFPP in Helsinki, Finland, during January 26–31, 2011. The power plant produces district heat and electricity by combined heat and power generation. The fuel power of the boiler in the studied power plant unit is 506 MW, the electrical power is 160 MW and the corresponding district heat power is 300 MW. The operation efficiency is 0.89. During the experiment, the boiler operated on average with a power of 440 MW and with stable operation conditions.

The fuel used at the power plant was bituminous coal from Russia. The fuel properties and composition details of the used coal are given in Supplementary data. The typical impurities in Russian coal include sulfides (e.g., pyrite (FeS_2)), carbonates (e.g., calcite ($CaCO_3$)), clay minerals (e.g., kaolinite ($[Al(OH)_2]_2SiO_5$)), and tectosilicates (e.g., quartz (SiO_2) and