



Heavy metal concentrations in bottom ash and fly ash fractions from a large-sized (246 MW) fluidized bed boiler with respect to their Finnish forest fertilizer limit values

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ABSTRACT

The total and size fractionated concentrations of As, Cd, Cr, Cu, Ni, Pb and Zn in bottom ash and two fly ash fractions from a large-sized (246 MW) fluidized bed boiler were compared to Finnish statutory limit values for forest fertilizers, which came into force in March 2007. Fly ashes were sampled from the different fields (i.e. electrodes) of the electrostatic precipitator (ESP) unit treating the stack gases. The bottom ash and the fly ash from the first ESP field are suitable for use as a forest fertilizer. Due to the elevated As concentration (40 mg/kg; d.w.), which exceeded its Finnish limit value of 30 mg/kg (d.w.), the fly ash from the second ESP field is not suitable as a forest fertilizer alone. The results of ash sieving indicated that an As concentration of 40 mg/kg (d.w.) for particle size less than 0.125 mm for fly ash 2 from the second ESP electrode field exceeded the As limit value of 30 mg/kg (d.w.). In addition, a Pb concentration of 170 mg/kg (d.w.) for fly ash 1 from the first ESP electrode field for particle size 0.5–2.0 mm exceeded the Pb limit value of 150 mg/kg (d.w.). These two specific fractions are therefore not suitable for use as a forest fertilizer alone.

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1. Introduction

The pulp and paper industry is an energy-intensive industry. Due to the high demand for energy in its processes, most of the energy is produced by combustion of all the appropriate burnable residues, e.g. black liquor from the pulp-making process, clean wood residues (i.e. bark, wood chips and sawdust) from wood-handling plant, sawmill and ground-wood mill, as well as biosludge from activated sludge plant [1,2]. The thermal energy generation from these types of industrial residues is a sustainable, environmental friendly alternative to other energy production processes as it significantly reduces the use of fossil fuels such as heavy fuel oil. According to European Union legislation, the utilization of the energy content of process residues is considered as BAT (i.e. Best Available Techniques) in the pulp and paper industry [3]. The term “Best Available Techniques” refers to both the technology used and the way in which the installation is designed, built, maintained, operated and decommissioned, i.e. the most up-to-date stages of development (or the state-of-the-art) of processes and their methods of operation, the technologies on which they operate, taking into account the practical suitability and cost of a particular technique for preventing,

or where that is not practicable, minimizing emissions to the environment as a whole and achieving a high general level of environmental protection. This kind of waste-to-energy treatment is eco-efficient, since the energy content of the wastes are utilized in the form of heat and electricity, and this saves virgin raw materials therefore [4]. The most important justification for the use of wood residues for energy production is that it could significantly reduce the formation of carbon dioxide emissions. The use of renewable wood-derived residues allows the utilization of natural raw material as an energy source without having any marked effects on the carbon balances in ecosystems [5].

Combustion via bubbling fluidized bed boiler (BFB) is a widely used combustion technology for energy recovery in the modern pulp and paper industry. The BFB is especially suitable for inhomogeneous biofuels such as bark and other wood residues [1]. Fluidized bed combustion technology is suitable for co-combustion of different fuels even fuels with high moisture content. Although the incineration of pulp and paper mill residues using fluidized bed combustion is rapidly becoming the ultimate solution for the final disposal of wastes, the disadvantage of energy generation from biomass is that it produces a considerable amount of ash residue. Ash residue fractions such as bottom ash, which accumulates at the bottom of the fluidized bed boiler and fly ash, which is collected from the flue gas by electrostatic precipitator, wet scrubbing, or a mechanical device such as a multicyclone or a baghouse,

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constitute a major fraction of the solid residues produced by the power plants of pulp and paper mills [6,7].

Due to the fact that most of the heavy metals evaporate in the combustion process and condense on the surface of particulate matter (i.e. fly ash), the different fly ash fractions from an electrostatic precipitator may have a different chemical and physical composition and are therefore variable in their suitability for subsequent utilization. The large and increasing volume of ash residues arising from Finnish pulp and paper mills is motivating a search for disposal options other than that of landfilling. Therefore, there is a growing trend towards the utilization of ash residues in Finnish pulp and paper mills. Careful attention to ash chemistry can maximize the potential of ash residues for use in beneficial applications instead of the reliance on landfill for their disposal. This study is part of a major project, in which the beneficial use potentials of pulp and paper mills' residues are being investigated within the context of industrial symbiosis in order to develop novel applications for various alkaline pulp and paper mill residues [8].

2. Experimental

2.1. Sampling

The bottom and fly ashes investigated in this study originate from a large-size (246 MW) bubbling fluidized bed boiler (BFB) at the power plant of a pulp and paper mill complex located in Finland. At the present time, approximately 50% of the energy produced by the BFB originates from the incineration of clean forest residues (i.e. bark, woodchips and sawdust) and ca. 50% from the incineration of commercial peat fuel. The bottom ash was sampled from the outlet of the boiler and the fly ashes from the boiler's electrostatic precipitator (ESP) (see Fig. 1). The ESP at the power plant has three fields (i.e. electrodes). However, in this study, it was only possible to collect fly ash from the first two (i.e. fly ash 1 and fly ash 2) and not from the last field due to the fact that the first two fields remove particulate matter from the flue gas so effectively that there was no particulate matter left for sampling in the last field. The effectiveness of the removal of particulate matter from the flue gases is illustrated by the fact that the particulate matter concentration in the flue gas emitted from the power plant is only ca. 2 mg/m³ (n).

The sampling of the bottom and fly ashes was carried out over a period of five days, and the individual samples (i.e. one kg per day) were combined to give one composite sample with a weight of 5 kg. The sampling period represented normal process operating conditions for the combustion plant, e.g. O₂ content and temperature. After

sampling, the samples were stored in polyethylene bottles in a refrigerator (+4 °C). A coning and quartering method [9] was applied repeatedly to reduce the ash samples to a size suitable for conducting laboratory analyses. The plant produced about 15,430 tonnes of ash (i.e. ca. 2580 tonnes of bottom ash and ca. 12850 tonnes of fly ash) in 2009. During the last few years, about 30–40% of the fly ash has been used as a hardener in filling local mine cavities. Furthermore, the rest of the fly ash and all bottom ash have been used for landscaping the mill's on-site ash basin.

2.2. Determination of the mineral composition, physical and chemical properties of ashes

For the determination of the mineralogical composition of the bottom ash and fly ash, X-ray diffractograms of powdered samples were obtained with a Siemens D 5000 diffractometer (Siemens AG, Karlsruhe, Germany) using CuK α radiation. The pH and electrical conductivity (EC) of the ashes were determined by a combination pH/EC analyser equipped with a Thermo Orion Sure Flow pH electrode (Turnhout, Belgium) and a Phoenix conductivity electrode (Phoenix Electrode Co., Texas, USA) with a cell constant of 1.0. The determination of pH and electrical conductivity (EC) were carried out according to European standards SFS-EN 12880 and SFS-EN 13037, respectively, at a solid to liquid (i.e. ultrapure water) ratio of 1:5.

Determination of the dry matter content of the bottom and fly ashes was carried out according to European standard SFS-EN 12880. The organic matter content, as measured by the loss-on-ignition (LOI), was determined according to European standard SFS-EN 12879. The total organic carbon (TOC) content was determined according to European standard SFS-EN 13137. The neutralizing (liming effect) value and reactivity value were determined according to the European standards SFS-EN 12945 and SFS-EN 13971, respectively.

For the determination of the metals distribution in different particle size fractions, the bottom ash and fly ashes were dry sieved on an automatic sieve shaker (Retsch Virbo, Haan, Germany) through stainless-steel sieves using a stack of nested sieves (DIN 4188, Retsch 5657, Haan, Germany) with the following particle size fractions: 2, 2-1, 1-0.5, 0.5-0.25, 0.25-0.125, 0.125-0.075 and <0.075 mm. The weight of each size fraction both for the bottom ash and fly ashes was recorded and the percent distribution of weight in each fraction was calculated. For the determination of total metal concentrations in the sieved ashes, the above mentioned particle size fractions were combined in order to obtain the following size fractions: 2.0-0.5, 0.5-0.125 and <0.125 mm. The heavy metal concentrations in these particle size fractions were

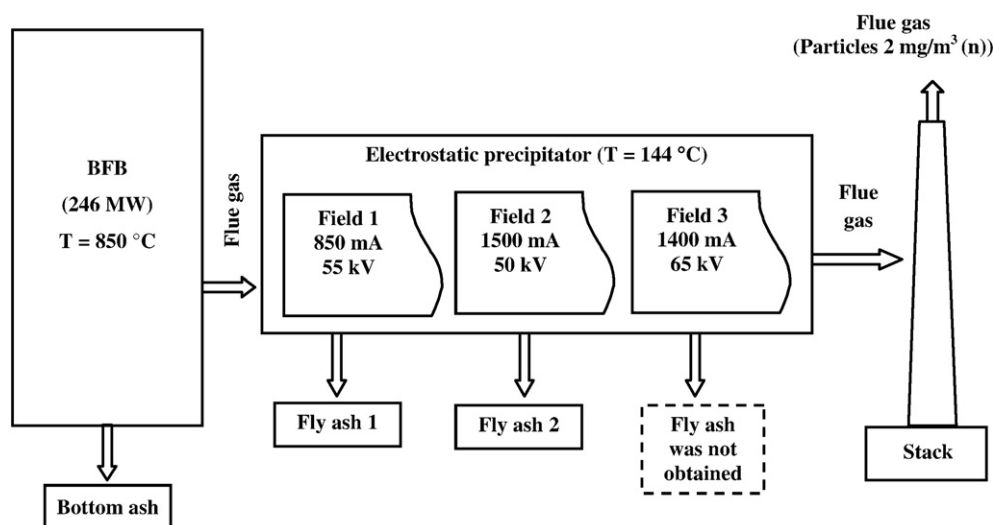


Fig. 1. Schematic picture of the bubbling fluidised bed boiler (BFB), the 3-field electrostatic precipitator, sampling of bottom ash and of fly ashes 1 and 2. Note: It was not possible to collect fly ash from electrode field 3 because electrodes 1 and 2 removed nearly all particulate matter from the flue gas; (picture not at a real scale).

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