



Investigation of polycyclic aromatic hydrocarbon content in fly ash and bottom ash of biomass incineration plants in relation to the operating temperature and unburned carbon content



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HIGHLIGHTS

- The 16 EPA PAHs in 96 ash samples of biomass incineration plants were investigated.
- The highest PAH content was observed in phytomass fly ash derived at 500 to 750 °C.
- The LMW, MMW, HMW and total PAHs in ash correlated significantly.
- The total PAHs present in ash increased with increased levels of unburned carbon.
- The ashes suitable for soil applications as an organic amendment were suggested.

GRAPHICAL ABSTRACT



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ABSTRACT

Purpose: The use of biomass fuels in incineration power plants is increasing worldwide. The produced ashes may pose a serious threat to the environment due to the presence of polycyclic aromatic hydrocarbons (PAHs), because some PAHs are potent carcinogens, mutagens and teratogens. The objective of this study was to investigate the content of total and individual PAHs in fly and bottom ash derived from incineration of phytomass and dendromass, because the data on PAH content in biomass ashes is limited. Various operating temperatures of incineration were examined and the relationship between total PAH content and unburned carbon in ashes was also considered.

Methods: The analysis of PAHs was carried out in fly and bottom ash samples collected from various biomass incineration plants. PAH determination was performed using gas chromatography coupled with mass spectrometry. The correlations between the low, medium and high molecular weight PAHs and each other in ashes were conducted. The relationship between PAH content and unburned carbon, determined as a loss on ignition (L.O.I.) in biomass ashes, was performed using regression analysis.

Results and discussion: The PAH content in biomass ashes varied from 41.1 ± 1.8 to $53,800.9 \pm 13,818.4$ ng/g dw. This variation may be explained by the differences in boiler operating conditions and biomass fuel composition. The correlation coefficients for PAHs in ash ranged from 0.8025 to 0.9790. The regression models were designed and the coefficients of determination varied from 0.908 to 0.980.

Conclusions: The PAH content in ash varied widely with fuel type and the effect of operating temperature on PAH content in ash was evident. Fly ashes contained higher amounts of PAHs than bottom ashes. The low molecular

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weight PAHs prevailed in tested ashes. The exponential relationship between the PAH content and L.O.I. for fly ashes and the linear for bottom ashes was observed.

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1. Introduction

Biomass is a type of biological material that can act as a renewable fuel source for energy production. The thermochemical process of conversion of biomass fuels, such as industrial grown plants, fast growing trees, fuel wood, wood chips, forest and harvest residues and many others, is possible to use for the production of useful heat and electricity (McKendry, 2002; James et al., 2012). The energy derived from biomass combustion contributes to the development of environmental, social and economic sustainability. The direct combustion of biomass fuels in incineration plants is considered to be the most promising energy source to mitigate greenhouse gas emissions (Khan et al., 2009; Atkins et al., 2010).

Biomass combustion produces fly ash and bottom ash as a waste-product of the biomass conversion processes (Bridgwater et al., 1999; Johansson and van Bavel, 2003a). Fly ash represents the solid component mixed with unburnt particles of fuel, which is deposited on the precipitator inside the fluidized, circulating fluidized or bubbling fluidized boilers. The bottom ash consists of solid bed particles located in the bottom part of the furnace (Pitman, 2006; Freire et al., 2015).

The ash derived from biomass combustion is rich in macro- and micronutrients and has the potential to be utilized as a mineral fertilizer in agriculture (Ferreiro et al., 2011). The use of this waste material may also reduce landfill disposal (Reijnders, 2005). However, the ash derived from biomass combustion can contain high amounts of hazardous metals that may cause serious health risks (Aronsson and Ekelund, 2004; Pöykiö et al., 2009).

The conditions of biomass combustion can also lead to the formation of polycyclic aromatic hydrocarbons (PAHs) in biomass ash (Chagger et al., 2000; Enell et al., 2008). The most frequent formation mechanism of PAHs is pyrolysis and subsequent pyrosynthesis during the incomplete combustion of biomass. PAH formation via high and low temperature pyrolytic reaction has also been described (McGrath et al., 2001; Morf et al., 2002; Sharma and Hajaligol, 2003; Hays et al., 2005). The formation of PAHs during biomass combustion is strongly dependent on the type of biomass fuels used, their physical and chemical properties and the operating conditions of combustion (Jenkins et al., 1998; Ross et al., 2002).

The PAHs represent persistent organic pollutants (POPs) which are widely distributed in the environment as a consequence of human industrialization (Wheatley and Sadhra, 2004; Bignal et al., 2008). These lipophilic compounds consist of 2 to 7 fused aromatic benzene rings in linear, angular or cluster arrangements. This arrangement predicts their stability in the environment and stable PAHs tend to accumulate mainly in soils and sediments. The physical-chemical properties, distribution and behaviour of PAHs in the environment also vary considerably with molecular weight. The lower molecular weight PAHs (LMW PAHs), containing 2–3 rings, are mobile in the environment, whereas the medium molecular weight PAHs (MMW PAHs) containing 4 rings and higher molecular weight PAHs (HMW PAHs), containing 5–7 rings, are relatively non-mobile. With increasing molecular weight, the melting point, boiling point and lipophilicity of PAHs with respect to $\log K_{ow}$ (n -octanol-water partition coefficient) grows and the water solubility decreases, suggesting increased solubility in lipid compounds (Eisler, 2000).

Sixteen basic PAHs are included in the United States Environmental Protection Agency (16 US EPA PAHs) priority pollutant list, because they may pose serious threats to the environment and biota (USEPA, 2016). The interest of researchers in PAH monitoring in ash has increased recently due to the possibility of recycling nutrients from ash to soil,

because PAHs could be absorbed and assimilated by plants and subsequently enter into the food chain of animals and humans (Demirbas, 2005; Park et al., 2012). Individual PAH compounds have various toxicities and some PAHs are known to have carcinogenic, teratogenic and mutagenic properties. For instance, naphthalene has been described as the most acutely toxic PAH and benzo[*a*]pyrene has been identified as carcinogen to humans (Juhasz and Naidu, 2000).

Most previous research has focused on PAHs in ashes derived from municipal solid wastes and wood biomass, and very few studies have characterized the distribution of low, medium and high molecular weight PAHs found in ashes taken from commercial biomass incineration plants operated between 250 and 1000 °C. The data on PAH content in fly and bottom ashes derived from phytomass (including agricultural crop residues) in literature are still limited and the relationship between the unburned combustible carbon and total PAH content in various biomass ashes has not been studied sufficiently.

The main objective of this work was to investigate the total and individual content of the 16 US EPA PAHs in fly ash and bottom ash derived from phytomass and dendromass derived from various operating temperatures of biomass incineration. The relationship between the total PAH content and unburned carbon in ashes was also considered.

2. Experimental

2.1. Ash sampling and preparation

Ninety six ash samples were taken from 48 commercial biomass incineration plants located in the Czech Republic. Fly ash and bottom ash samples were collected from each incineration plant. The fly ashes could be a mixture of ashes collected at the superheater, economizer, and electrostatic precipitator. All bottom ashes were collected from the tank below the fluidized or grate boilers. The approximate amount of 2 kg of each ash sample was taken in four random replications which were further mixed thoroughly. The samples were put into dark glass bottles and transported to the laboratory for further analysis. The tested samples were air-dried to a constant mass in the laboratory, pulverised by a Retch fraction mill (Retch, Germany), passed through a 2.0 mm sieve and subsequently homogenized. A total of 50.0 g of each homogenized ash sample was grounded to a fine powder by a vibration mill with diameter 15–20 µm. If necessary, the samples of ash were stored in a refrigerator at –8/–10 °C in Petri dishes covered with aluminium foil.

Before PAH-specific analysis, each ash sample was diluted in deionized water at a ratio of 5:1 (solid to liquid) and then the pH value was measured after 24 h according to Johansson and van Bavel (2003a). For pH determination, a WTW pH 340i meter with glass ion selective electrode (WTW, Germany) was used. The amount of unburned combustible carbon in each ash sample was determined as a loss on ignition (L.O.I.), when 10.0 g of dry ash sample was heated in a crucible placed in a furnace for 2 h at 105 °C and then the temperature increased to 1000 °C at a rate of 10 °C/min. The L.O.I. was calculated, as follows: $L.O.I. (\text{mass } \%) = ((\text{Weight}_{105} - \text{Weight}_{1000}) / \text{Weight}_{105}) \times 100$, where Weight_{105} is the weight of sample after heating at 105 °C and Weight_{1000} is the weight of sample after ignition at 1000 °C. This is a standard method used in many European countries for determination of residual organic matter represented by unburned carbon in ash (Stubington and Wang, 2000). The L.O.I. analyses were performed in the Institute of Rock Structure and Mechanics, Academy of Sciences of the Czech Republic. The analyses of PAHs were performed in the

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