

Polycyclic aromatic hydrocarbons in ash: Determination of total and leachable concentrations

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Total PAH content and the particle weathering rate are more important influences on the rate of migration of PAHs from recycled wood ash into forest soils than leaching.

Abstract

Before wood ash can be used as a soil fertilizer, concentrations of environmentally hazardous compounds must be investigated. In this study, total and leachable concentrations of 16 polycyclic aromatic hydrocarbons (PAHs) were determined in four ash samples and one green liquor sludge. The ash sample with the highest carbon content also contained high levels of PAHs; three of the ash samples had total concentrations exceeding the limit permitted by the Swedish Forest Agency for recycling to forest soils. The leachable concentrations were higher for the non-stabilized samples; this was probably due to colloid-facilitated transport of the contaminants in these samples. However, the leachable concentrations were overall relatively low in all the samples studied. The amounts of PAHs introduced to forest soils by additions of stabilized, recyclable ash products will be determined primarily by the rate of weathering of the ash particles and the total concentration of contaminants. © 2007 Elsevier Ltd. All rights reserved.

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) comprise a large category of stable hydrophobic organic compounds that are highly persistent in the environment. Sixteen of the PAHs are listed as priority pollutants by the United States Environmental Protection Agency (US-EPA) due to their toxic, mutagenic and carcinogenic effects (ATSDR, 1995). They are formed during incomplete combustion at levels that depend both on the fuel burned (Yasuda et al., 1989; Johansson and van Bavel, 2003) and the combustion conditions (temperature, oxygen, residence time and turbulence) (Andersson and Marklund, 1998; Blumenstock et al., 2000). PAHs formed in the combustion chamber are subsequently distributed between

flue gases and the combustion residues (Davies et al., 1976; Oehme et al., 1987; Luthe et al., 1998). Most of the PAHs are associated with particulate material and, in combustion plants with air pollution control, the PAHs either remain in the bottom ash or are removed from the flue gases by the precipitators to form fly ash. As yet there are no regulatory limits on PAH concentrations in ashes from the combustion of municipal solid waste (MSW) or biofuel in Sweden; current regulations used to determine whether the ash is classed as hazardous waste are concerned only with inorganic pollutants.

The dominant energy sources in Swedish district heating plants are biofuels, such as harvest residues and wood chips; a smaller number of plants generate power from MSW incineration. Although plants of both types produce substantial amounts of ashes annually, the ashes from MSW often contain high concentrations of heavy metals and other environmentally hazardous compounds. These are treated as hazardous waste and must be landfilled. Wood ash from the burning of

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clean biofuels, on the other hand, generally contains low amounts of heavy metals. Nutrients from the wood and bark are retained in the ashes, and can be recycled to forests to promote sustainable wood production. In Sweden such recycling of wood ash is regulated by the Swedish Forest Agency (SFA). At present, the only recommendation pertaining to PAHs in wood ash destined for recycling states that the concentration of PAHs should not exceed 2 mg kg^{-1} for the sum of the 16 US-EPA PAHs (SFA, 2001).

Although the contents of PAHs in ashes of various origins was recently reported by Johansson and van Bavel (2003), there is still very little published data on PAHs in wood ash. Possible aqueous emissions due to leaching of PAHs from ash have received even less attention. In the study reported here, the total and leachable concentrations of 16 PAHs were quantified in two wood ash products, one green liquor sludge/lime mud mixture intended for recycling to forests soils, and two ashes from the incineration of MSW.

2. Materials and methods

2.1. Samples

Five samples of different origins were selected for the investigation (Table 1). Two of the samples were combustion residues originating from the incineration of MSW and destined for landfill disposal (denoted L1 and L2, respectively, in Table 1, where L stands for ash for Landfill). L1 was a fly ash from a circulating fluidized bed boiler and L2 a bottom ash from a bubbling fluidized bed boiler. The other three samples (R1, R2 and R3) were destined for forest recycling (where R stands for ash for Recycling). R1 was a mixture of green liquor sludge mixed with lime mud (residual products from the recycling of chemicals in the pulp industry) and fly ash collected from a bubbling fluidized bed boiler. Sample R2 was a combustion residue (fly ash) from a converted Wanderrost boiler (adapted to be top loaded with wood powder). Sample R3 was a mixture of green liquor sludge and lime mud. Thus, it contained no ash, but for simplicity all five samples are henceforth termed ashes in the paper. The green liquor sludge was similar to wood ash in that much of the inorganic material it contained was from wood, but differed from biofuels in that the organic material had been removed chemically in the pulp production process, rather than burnt in a combustion chamber.

2.2. PAH extraction

In order to quantify the concentrations of the amounts of the 16 US-EPA PAHs (individual and total) in the samples they were extracted by mixing 3 g

of each sample with 20 g of toluene-washed sand, adding ^{13}C -labelled internal standards and 15 ml of acetic acid, then subjecting them to Soxhlet–Dean–Stark extraction for 48 h with toluene (400 ml) as the extraction solvent. The extract obtained from each sample was concentrated and cleaned up by transferring 25% of each extract to a silica column and eluting with cyclopentane (60 ml). The resulting eluate was concentrated to 1 ml in toluene and a recovery standard was added, before PAH determination was carried out by gas chromatography–low resolution mass spectrometry using a Thermoquest TraceGC gas chromatograph coupled to an MD 800 mass spectrometer (see Liljelind et al., 2003, for a more detailed description).

2.3. Leaching of PAHs

Leaching of PAHs from the ashes was studied, using the apparatus described below, under both alkaline and acidic conditions. The ash samples R1, R2 and L2 were initially leached at $\text{pH} > 10$, to accumulated liquid/solid (L/S) ratios of 580, 675 and 810 L kg^{-1} , with deionized water. Leaching was then continued with water adjusted to $\text{pH} 3.0$ to accumulated L/S ratios of 1370, 1570 and 1650 L kg^{-1} , respectively. The leaching of PAHs from the samples R3 and L1 was studied only at $\text{pH} 3.0$ and were leached to accumulated L/S ratios of 1060 and 780 L kg^{-1} .

2.3.1. Experimental apparatus

The leaching of PAHs was assessed using a column method with on-line filtration and solid phase extraction (SPE). The leaching device and the SPE method are described in more detail in Enell et al. (2004), and are illustrated in Fig. 1. The experimental apparatus consisted of five separate vertical glass columns (one for each sample) through which water was pumped from bottom to top. The effluent was filtered ($0.7 \mu\text{m}$) before being passed through an SPE cartridge to trap the leached PAHs. The SPE cartridges were replaced and the PAHs they had collected were analyzed after approximately 80 L of effluent had passed through them.

The leaching experiments under alkaline conditions were performed with recirculating water (Fig. 1a), since the SPE cartridges were expected to adsorb all hydrophobic organic compounds effectively, including the PAHs (Enell et al., 2004). The eluent used for the alkaline experiments was initially adjusted to $\text{pH} 7$, but increased almost instantly to $\text{pH} > 10$ in all samples studied (R1, R2 and L2), due to the very high alkalinity of the ashes.

It was not feasible to use recycled water for the experiments conducted under acidic conditions, so influent water from a reservoir, adjusted to $\text{pH} 3$ (with HCl), was used for these cases (Fig. 1b). The average flow of leachate was 5 ml min^{-1} in both the alkaline and acidic experiments.

The glass columns (internal diameter 5 cm, height 30 cm) were packed with a mixture of ash (approx. 250 g) and sand (approx. 500 g), the latter being added to increase the hydraulic conductivity of the sample matrix. To prevent photolysis of the PAHs, the leaching apparatus was kept in darkness at all times except during maintenance (e.g. to change SPE cartridges and filters).

Table 1
Characteristics of the five samples chosen for investigation

Sample	Composition	Combusted material	Boiler	Stabilized	Intended use	TOC %
R1	Fly ash, green liquor and lime mud	Wood chips	BFB ^c	Granules	Recycling	8
R2	Fly ash	Wood powder	CWB ^d	Pellets	Recycling	21
R3	Green liquor and lime mud	Wood chips	Soda recovery unit	Yes ^f	Recycling	10
L1	Fly ash	MSW ^a	CFB ^c	Yes ^f	Landfill	4
L2	Bottom ash	MSW ^a , RDF ^b	BFB ^c	No	Landfill	0.005

^a Municipal solid waste.

^b Refuse-derived fuel, obtained from a fraction of dry waste material (paper, plastic etc.) that is sorted and shredded before incineration.

^c Bubbling fluidized bed.

^d Converted Wanderrost boiler.

^e Circulating fluidized bed.

^f The stabilization procedure is described in Section 2.

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