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## Waste Management

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# Recirculation of biomass ashes onto forest soils: ash composition, mineralogy and leaching properties

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## ARTICLE INFO

## Article history:

Received 20 January 2017

Revised 26 July 2017

Accepted 9 September 2017

Available online xxx

## ABSTRACT

In Denmark, increasing amounts of wood ashes are generated from biomass combustion for energy production. The utilisation of ashes on top of forest soil for liming purposes has been proposed as an alternative to landfilling. Danish wood ash samples were collected and characterised with respect to chemical composition, mineralogy and leaching properties (batch leaching at L/S 2 and 10 L/kg, and pH-dependent leaching at 10 L/kg). Large variations in the ash liming properties were observed (ANC<sub>7.5</sub>: 1.8–6.4 meq H<sup>+</sup>/g), indicating that similar soil application dosages may result in different liming effects. High contents of Ca, Si, P, K and Mg were observed in all samples, while the highest contents of S and N were found in fly ashes and mixed ashes (combination of fly and bottom ashes). Similarly, the highest contents of some trace metals, e.g. Cd, Mo and Se, were observed for fly ash. Releases of major, minor and trace elements were affected significantly by pH: high releases of PO<sub>4</sub><sup>3-</sup>, Mg, Zn, Cu and Cd were found for acidic conditions relevant to forest soils, while the highest releases of Mo and Cr were observed in alkaline conditions. Mineral phases were selected based on XRD analyses and the existing literature, and they were applied as inputs for the geochemical modelling of pH-dependent leaching. Mineral dissolution was found adequate for a wide range of major elements and nutrients, while the description of trace elements could be done only for parts of the pH-range. Content and leaching of PAHs were observed below detection limits. The source-term release of Ca, K, Mg, Mn, and P in acidic conditions relevant to forest soils was higher than ten years of atmospheric deposition, in contrast to the relatively low release of Al, Fe and Na. The potential release of Cd was found to be the most critical element compared with soil quality criteria, whereas the maximum theoretical loads of Ba, Cd, Cr, Sr, Mo, Ni, Pb, Sb, Se, Sn and V were relatively low.

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## 1. Introduction

Many European countries are introducing increasing amounts of renewable energy sources at the expense of fossil fuels (European Commission, 2015). In addition to wind and photovoltaic options, biomass often plays an important role in this transition. The Danish district heating system is based on many distributed combined heat-and-power plants supplying both electricity and district heating for the surrounding local region. Many

of these plants utilise wood chips as fuel for combustion, and many of the larger coal power plants are currently being converted into wood pellet combustors, at the expense of coal. This follows the current Danish energy strategy of being fossil fuel independent by 2050 (The Danish Council on Climate Change, 2015). The production of renewable energy from wood chips and wood pellets has shown a significant increase within the last decade, i.e. from 5.7 PJ to 16 PJ in the period 2000–2015 (Danish Energy Agency, 2016). The increased use of woody biomass fuels, however, results in the increased production of wood ashes; in Denmark, about 22,300 tonnes dry weight (dw) of wood ash was produced in 2012 (Skov and Ingerslev, 2013).

The use of wood ashes on forest and agricultural soil is regulated in some European countries, such as Denmark, Finland, Sweden, Austria and Germany (van Eijk et al., 2012). In Denmark, for example, the dosage of ash applied to forest soil (DEPA, 2008) is regulated depending on (i) the content of specific contaminants

*Abbreviations:* ANC, acid neutralisation capacity; BA, bottom ash; CSH, calcium-silicate hydrates; DOC, dissolved organic carbon; dw, dry weight; FA, fly ash; ICP, inductively coupled plasma; LOD, limit of detection; MA, mixed ash; MSWI, municipal solid waste incineration; OCP, octacalcium phosphate; PAH, polycyclic aromatic hydrocarbon; SI, saturation index; SQC, soil quality criteria; TOC, total organic carbon; XRD, X-ray diffraction.

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<https://doi.org/10.1016/j.wasman.2017.09.008>

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in the ash, (ii) the need for plant fertiliser and (iii) the ash's electrical conductivity: a maximum of three tonnes/ha can be applied over a period of 10 years, but not more than three times within the last 75 years. There are approximately 615,000 hectares of forest land in Denmark, which in principle could receive wood ashes. Nevertheless, most of the wood ashes currently produced in Denmark are collected in containers at individual plants and landfilled (Ingerslev et al., 2014), most likely because of the small capacity of the Danish power plants (and therefore relatively small quantities of ashes), stringent legislation limits, concerns about their composition and leachability and the costs associated with the chemical analysis and documentation of ash quality. However, with increasing amounts of wood ashes being generated, landfilling is not a viable solution from a long-term perspective, in that the sustainability of wood combustion requires the continuous renewal of forest biomass and the input of nutrients. The application of wood ashes in forestry may contribute to the recirculation of nutrients (e.g. K, Mg, Ca and P) as well as offer liming effects on the soil (Pitman, 2006) – using three tonnes of wood ashes on top of soil was reported to have a liming effect comparable to one tonne of lime, CaO (Karlton et al., 2008). On the other hand, recirculating wood ashes onto forest soil may also result in undesired releases of contaminants. Specific focus, for example, has been placed on Cd, which has been investigated for its potential bioavailability and toxicity in relation to specific soil ecosystems (e.g. Cruz-Paredes et al., 2017; Fritze et al., 2001; Perkiömäki and Fritze, 2005). Hence, although the potential for increasing the recirculation of wood ashes to forestry may exist in Denmark, further clarification on the consequences of this process is needed.

According to the available literature, the composition of wood ash has been addressed as a function of biomass fuel (Drift et al., 2001; Reimann et al., 2008; Werkelin et al., 2011), furnace operating parameters (Etiégni and Campbell, 1991; Misra et al., 1993; Sarenbo, 2009), combustion technology (Freire et al., 2015; Lanzerstorfer, 2015; Pöykiö et al., 2007) and different ash types (Dahl et al., 2009; Ingerslev et al., 2011; Sano et al., 2013). Several authors have investigated the leaching of wood ash, by applying different methods such as sequential extractions, batch leaching tests and percolation tests (Liodakis et al., 2009; Mellbo et al., 2008; Pöykiö et al., 2012; Sano et al., 2013; Steenari et al., 1998, 1999; Supancic et al., 2014). However, due to the different leaching test conditions applied in these studies, a direct comparison of results taken from individual studies is not possible. Despite a recent study by Freire et al. (2015), who characterised the leaching of a few wood ash samples under different leaching conditions, existing literature is relatively fragmented. Moreover, a systematic evaluation of the variability of leaching from a wider range of ashes, and an evaluation of the leaching mechanisms controlling the release of nutrients and contaminants from these ashes, is largely missing. A consistent evaluation of the leaching properties of these ashes is needed as the basis for future changes in the regulatory framework targeting the minimisation of the landfilling of wood ashes.

The overall aim of this study is to provide a consistent evaluation of the leaching properties of wood ashes, in order to improve the knowledge necessary for recirculating these combustion residues onto forest soils. This includes the following specific objectives: (i) to document variability in ash composition and leaching behaviour over a range of wood ash samples, (ii) to evaluate the effects of pH in relation to leaching for selected ashes, (iii) to identify and interpret key mechanisms controlling leaching by means of geochemical modelling and (iv) to evaluate potential source-term releases onto forest soils with respect to soil quality criteria and atmospheric deposition.

## 2. Materials and methods

### 2.1. Sampling and material handling

Ashes from ten different Danish biomass combustion facilities (see Table S1 in Supplementary Material for more details on the characteristics of these plants) receiving wood chips, mainly from *Picea abies*, were sampled in the period January – March 2014. Depending on the specific technology, freshly generated bottom ash (BA) and fly ash (FA) may be collected at the plant through two separate flows or one single mixed flow (fresh mixed ash: MA). The latter was a specific case of closed ash conveyer systems, where it was not possible to sample BA and FA separately. Consequently, the ratio between BA and FA in the MA samples was not known. A total of sixteen wood ash samples were collected: three BA, four FA and nine MA samples. Sample names used in the following text refer to the ash type, i.e. BA, FA or MA, followed by a number representing the plant, i.e. 1–10, and an optional letter, i.e. a, b or c, indicating the replicate number (where relevant). Upon receipt of the samples at the laboratory, the mass of each sample was reduced by quartering and splitting, using a riffle splitter in accordance with ASTM C702/C702M: 2011. All samples were stored in airtight plastic containers at 10 °C prior to testing and characterisation.

### 2.2. Ash characterisation

Moisture content was determined on 100 g subsamples according to EN 1097-5: 2008. Next, the dry material was crushed in a jaw crusher and then pulverised in a vibratory disc mill (agate discs). Approximately 0.2 g subsamples of the powder were obtained by consecutive riffle splitting and used for determining total organic carbon (TOC; EN 13137: 2001; LECO induction furnace CS-200 Analyser) and contents of C, S, N and H (Elemental Analyser – Vario MACRO cube); triplicate analyses were carried out. The elemental composition of the wood ash samples was determined in duplicates using 0.25 g of pulverised material for microwave-assisted acid digestion (Multiwave Anton Paar 3000) according to EN 13656: 2003, using 6 mL of HNO<sub>3</sub> (65 %), 2 mL of HCl (37 %), 2 mL of HF (40 %) and 12 mL of H<sub>3</sub>BO<sub>3</sub> (10 %). In addition, 2 mL of H<sub>2</sub>O<sub>2</sub> (30 %) was added to enhance the oxidation of residual organic matter. The digestates were analysed by inductively coupled plasma (ICP) mass spectrometry (7700x, Agilent Technologies) for the content of Al, Ag, As, Au, Ba, Be, Ca, Cd, Ce, Co, Cr, Cs, Cu, Er, Eu, Fe, Ga, Gd, Ge, Hf, Ho, In, Ir, K, La, Li, Lu, Mg, Mn, Mo, Na, Nb, Nd, Ni, P, Pb, Pd, Pr, Pt, Rb, Re, Rh, Ru, Sb, Sc, Se, Si, Sm, Sn, Sr, Ta, Tb, Ti, Tl, Tm, V, W, Yb, Zr and Zn, and by ICP optical emission spectrometry (Varian Vista-MPX) for the content of Ca, K, Mn and S. The same procedure was repeated to a reference material, i.e. BCR-176 R, to validate the results.

To facilitate more detailed characterisation, based on the results of the elemental content analyses, two ash samples were selected with the following characteristics: a) one ash sample with the highest Cd content among the sixteen samples (FA-2b, consisting of fly ash) and b) one ash sample complying with the current Danish limit values (DEPA, 2008) for utilising wood ashes on top of forest soils (MA-9c, a mixed ash consisting of both fly ash and bottom ash). As such, the two selected samples represented a “worst case scenario” with regards to the content of Cd, and a “typical situation” with ashes that are today allowed for recirculation in forestry settings. Further characterisation of the two selected samples, i.e. MA-9c and FA-2b, included: (i) determining particle size distribution, (ii) XRD scanning, (iii) analysis of the content of polycyclic aromatic hydrocarbons (PAHs; US EPA, 2008), using GC–MS after

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