



Research paper

Column leaching from a Danish forest soil amended with wood ashes: fate of major and trace elements



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ARTICLE INFO

Keywords:

Biomass

Liming

Release

Soil solution

Exchangeable cations

ABSTRACT

Application of wood ashes onto two Danish forest soil horizons (A- and O-horizons) was investigated through a series of column experiments for ash dosages of 3, 9 and 30 Mg ha⁻¹. Developments in the composition of the percolating soil solutions were investigated both in a short- (below 0.5 m³ m⁻² of infiltrating water) and long-term perspective (until 2.0 m³ m⁻² of infiltrating water). The higher the ash dosage, the higher the percolation of readily soluble elements (K, Cl, Mg and S) occurred within a short-term perspective. This initial washout of soluble elements resulted in the exchange of ions in the soil, thereby causing other soil bound elements to be released and the pH to decrease temporarily. Wood ash application also promoted an increase in the long-term leaching of As, Cu, K, P and Si beyond the O-horizon layer (until ~2.0 m³ m⁻²), while the migration of trace elements through this soil horizon appeared to be of limited concern compared with Danish groundwater quality criteria. Relatively similar effects were observed for both the use of 3 and 9 Mg ha⁻¹ dosages on the composition of the percolating soil solutions. Low mobility of Cd, Co, Cr, Cu, Mo, Ni, Pb, Se, V and Zn was observed. The released amounts were generally limited to a few percentage points of their total contents in the columns. The potential accumulation of trace elements within the forest soil should be evaluated with respect to the specific case, if high ash dosages are intended for spreading.

1. Introduction

Increasing amounts of energy from biomass combustion are being generated in Europe [1]. In Denmark, a considerable increase in the production of renewable energy from wood chips and wood pellets was observed in the period 2000–2015, i.e. from 5.7 PJ to 16 PJ [2]. Wood ashes are produced during the combustion of wood, and because of the increasing amounts of wood being used for energy purposes, more ashes are being generated. About 22,300 Mg dry weight (dw) of wood ashes are estimated to have been generated in 2012 [3]. In Denmark, wood ashes are typically landfilled [4], even though Danish legislation allows for their use on top of agricultural and forest lands for fertilising and liming purposes [5].

Current Danish legislation allows for a maximum wood ash dosage of 3 Mg ha⁻¹ onto forest soil, to be recirculated over a 10-year period, but not more than three times within a 75-year period [5]. More restricted dosages may however be required depending on whether the ash complies with the defined ash quality criteria [5]. As far as the ash quality criteria are complied with, there is no need for any ash pre-

treatment (e.g. hardening) [5]. In line with Danish legislation, the Swedish Forest Agency [6] recommends the use of approximately 2–3 Mg of wood ash per ha of soil to compensate for the removal of base cations and the loss of alkalinity due to harvesting. However, in the case of Sweden, it is recommended to use sufficiently hardened wood ashes [6]. While a wood ash dosage of 3 Mg ha⁻¹ has been reported to have an approximate liming effect comparable to a quicklime dosage of 1 Mg ha⁻¹ [7], its actual neutralisation value has been shown to vary widely across different samples, indicating that similar application dosages may result in different liming effects: neutralising values in the range of 13–92% of pure calcium carbonate were reported by Vance [8] during the analysis of 18 wood ash samples. Likewise, ash composition has also been shown to vary, depending on the type of fuel, the combustion technology and the boiler operating parameters (e.g. Refs. [9–11]).

Whether wood ashes are applied for fertilising or liming purposes, they will be expected to dissolve and their leaching to interact with the soil. In order to address these processes, many studies have focused on the development over time of the ash composition (to account for

Abbreviations: ANC, acid neutralisation capacity; DOC, dissolved organic carbon; dw, dry weight; GQC, groundwater quality criteria; ICP, inductively-coupled plasma; MS, mass spectrometry

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dissolved compounds; e.g. Refs. [12–15]), of the dissolved species in the percolating soil solutions (to evaluate releases beyond specific soil depths; e.g. Refs. [14,16,17]) and of the extractable soil components (to assess the extent of soil retention processes; e.g. Refs. [18–20]). Overall, short-term increases in the levels of Al, sulphate, K and Na have been reported in percolating soil solutions shortly after wood ash application [21,22], while elements of typical environmental concern (often identified as “heavy metals”) have shown low mobility in the soil and no significant increase in the soil percolating water [21,22]. Depending on the dosage, the application of wood ash has generally been shown to increase soil pH, cation exchange capacity and base saturation in the forest floor [18–20,23], and to decrease soil exchangeable acidity [19,21,23] – effects that were documented to persist even 30 years after wood ash application (i.e. long-term effects) [24]. While forest soil and wood ashes may be rather heterogeneous, the results of previous studies focusing on the effects of wood ash applications have generally reported high reproducibility of the experimental results across different replicates (e.g. Refs. [4,12,17]), i.e. limited variations in the analytical values.

In general, a relatively large number of studies have investigated the effects of wood ash application on the soil chemistry (e.g. changes in the cation exchange capacity, composition and pH). On the other hand, the effects on the percolating soil solutions have been often limited to a few key parameters (e.g. pH, electrical conductivity and a few key elements), and to relatively short-term effects (see Refs. [14,16,17,25]). Furthermore, experimental setups differ across different studies, making the results difficult to compare. More studies are needed in order to comprehensively evaluate the effects of different wood ash dosages on the mobility of nutrients and trace elements for diverse soil types and over longer times. The slow release of nutrients from the ash is considered a desired effect, as these may be taken up by local soil organisms and surface plants [6]. Similarly, the release of elements of environmental concern should also be slow, in order to limit potential toxic effects. Monitoring the composition of percolating soil solutions can provide insights into the migration of nutrients and contaminants through the soil, and into the releasing rates.

This study aims to investigate, by means of laboratory column tests, the short- and long-term release of selected inorganic elements from a Danish forest soil amended with wood ash. The topmost soil horizons (O- and A-horizons) were tested against three wood ash dosages, namely 3, 9 and 30 Mg ha⁻¹. The focus was to evaluate whether increasing dosages could potentially result in increasing element mobility overall. Specific objectives of this work were to investigate (i) the development of pH and concentration levels in percolating soil solutions, (ii) the cumulative release of nutrients and typical elements of environmental concern beyond the individual soil horizons and (iii) the residual contents in the columns after 2.0 m³ of water infiltrating per m⁻² of soil.

2. Material and methods

2.1. Ash and CaCO₃

Wood ash from a Danish heat-only boiler facility (55° 56′ 40″ N; 9° 7′ 19″ E Lat/Lon, WGS84), operating with a grate-fired system, was sampled. The combustion plant used wood chips, mainly from Norway spruce (*Picea abies*), as input fuel. The woody biomass came from the thinning of local Norway spruce forests (in the radius of about 50 km from the combustion facility). The trees were felled and dried on-site for about one year. These were then cut into chips and delivered to the combustion facility without any further treatment. The combustion plant burns about 8500 Mg of wood chips every year. Bottom ash and fly ashes are mixed together in a single output flow, which transports the freshly produced ashes into a container. A few hundred kilograms of these mixed ashes were sampled from the container itself. The chemical composition and the basic leaching behaviour of this ash sample was

determined in Maresca et al. [26,27] with the reference “MA-9c” in Maresca et al. [26] and “MA” in Maresca et al. [27].

Wood ash particles > 4 mm were crushed by means of a jaw crusher and mixed with the rest of the ash. The acid neutralisation capacity (ANC) of the ash was determined in accordance with CEN/TS 14997:2006 [28], whereby 30 g of ash was mixed gently with deionised water in a closed batch test (liquid:solid ratio of 10:1) and small amounts of HNO₃ were added following a step-wise procedure. Upon adding acid, the bottles were closed, the material was mixed gently for 25 min, allowed to settle for 5 min and the solution pH was then measured. This procedure was repeated until a final pH of 4.5 (ANC_{4.5}) was measured. The cumulative amounts of acid added and the observed pH of the solution were plotted in the so-called “ANC curve”, which is presented in Fig. S1 in Supplementary Material. The final ANC_{4.5} of the ash was determined at 4.3 meq g⁻¹.

A subsample of laboratory reagent calcium carbonate (min. 95% of CaCO₃) was used during the percolation tests (see Section 2.3). While this calcium carbonate is unlikely to be used for field application, laboratory-grade calcium carbonate was used here as a well-defined source of alkalinity (or ANC). The ANC of calcium carbonate was determined similarly to the ash, resulting in 19 meq g⁻¹ – see Fig. S1.

2.2. Soil sampling and characterisation

Soil was sampled from a test site in the Gedhus plantation, north-west of Denmark (56° 16′ 39″ N; 9° 5′ 7″ E Lat/Lon, WGS84), in a 57-year-old Norway spruce plantation in spring 2014. Note that, in Denmark, Norway spruce represents the most common tree species of local wooded areas [29]. The plantation is a second-generation forest nesting on heathland with a Podzol soil type. Mean annual temperature and precipitation are 8.4 °C and 0.850 m³ m⁻², respectively, in overall agreement with other forest lands in Western Denmark [30]. The site-specific net infiltration below the root nutrient zone was measured earlier at 0.270 ± 0.090 m³ m⁻² year⁻¹ [31]. The Gedhus plantation's test site was specifically established with the scope of testing the effects of wood ash application as part of the ASHBACK project (which this study is also part of) – for more details about the ASHBACK project, please refer to the Acknowledgments section.

The soil was sampled horizon-specifically from the O- and A-horizons (hereafter indicated as O-hor and A-hor). The soil was dried at 50 °C and sieved to < 2 mm and < 4 mm for the mineral and organic soil, respectively. The bulk density of the individual soil horizons was determined in triplicate, based on the soil mass of volume-specific samples. Particle size distribution of A-hor was determined using a Malvern Mastersizer 2000 according to DS/ISO 13320: 2009 [32]. Exchangeable cations were determined according to Stuanes et al. [33] after extracting the soil with a 80.04 kg m⁻³ NH₄NO₃ solution; amorphous and crystalline iron (hydr)-oxides were extracted with ascorbic acid (DS/EN ISO 12782-1 [34]) and dithionite (DS/EN ISO 12782-2 [35]), respectively, whereas aluminium (hydr)-oxides were extracted with ammonium oxalate/oxalic acid (DS/EN ISO 12782-3 [36]). All extracts were analysed by means of inductively coupled plasma (ICP) mass spectrometry (-MS) analysis (iCAP Q ICP, Thermo Fisher Scientific).

Soil samples were ground finely to < 1 mm, and multi-elemental analysis was performed using ICP-MS (Agilent 7900, Agilent Technologies), after digesting the samples in PTFE vessels in a microwave oven with a nitric acid/hydrogen peroxide solution at 250 °C. Nineteen elements were analysed: Al, As, Ca, Cd, Co, Cr, Fe, K, Mg, Mn, Mo, Na, Ni, P, Pb, S, Se, V and Zn. A ground subsample was analysed for C and N concentrations by dry combustion, using a FLASH 20000 EA NC Analyser (Thermo Fisher Scientific).

2.3. Percolation of soil columns

Column percolation experiments were carried out using dry soil

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