Forest Ecology and Management 424 (2018) 494–504

Contents lists available at ScienceDirect

Forest Ecology and Management

journal homepage: www.elsevier.com/locate/foreco

Effects of ash application on nutrient and heavy metal fluxes in the soil and soil solution in a Norway spruce plantation in Denmark

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ARTICLE INFO

Keywords:
Wood ash
Heavy metal speciation
Forest
Nutrient fluxes
Fertilisation
Soil solution

ABSTRACT

Wood ash application is used in forests to counteract nutrient losses and acidification due to intensified harvesting methods, where harvest residues are removed. The aim of the study was to investigate the effect of ash application at doses of 0, 3, 4.5 and 6 Mg ha^{-1} with three replicates on soil and soil solution chemical properties in a mature Norway spruce plantation in Denmark from 2014 to 2017. Un-replicated extreme scenarios of 15 and 30 Mg ha^{-1} were included. Soil solution from 15 to 60 cm depth were sampled each month over a period of 2 years and 9 months after ash application and soil samples were obtained after 2.5 years. The fluxes of nutrients and heavy metals through the soil were calculated based on water fluxes modelled with CoupModel. Geochemical modelling was applied on selected soil solution data to investigate changes in heavy metal complexation and mobility. Application of ash to forest soil increased both soil pH and nutrient concentrations in the O-horizon but not in the mineral soil. No changes were detected in the soil solution chemistry or in the speciation of heavy metal at doses ranging up to 30 Mg ha^{-1}, and there was therefore no change in the mobility of metals caused by ash application. The un-replicated extreme scenario suggested that the O-horizon concentration of Cd, Co, Cr, Cu and Zn could increase proportionally with increasing ash application dose, while no indications of changes in the mineral soil below 5 cm could be seen. Among the experimentally replicated treatments, only the Cd concentration at dose 6 Mg ha^{-1} exceeded the Danish legislation limits, in the O-horizon of 0.5 ppm.

1. Introduction

The focus on replacing fossil fuels with renewable energy sources has increased the use of biomass as an energy source in Europe as a response to mitigate climate change and enhance energy security (Bentsen and Felby, 2012; EU, 2009). The increased use of biomass for energy production will increase export of nutrients from the forest and the amount of ash produced during the incineration, and improvements of the sustainable management of the ash is therefore necessary (James et al., 2012). Overall, the produced ash can be landfilled as waste, be used in construction projects, or recycled in agriculture or forest ecosystems (Knapp and Insam, 2011). The effect of using ash in forest systems has been studied in Northern European countries for decades, with a main focus on the fertilising and liming effects of ashes (Aronsson et al. 2004; Karltun et al., 2008). To optimise the energy potential in forests, an intensification of harvest methods has occurred where harvesting residues are also included, rather than left behind to decay (Achat et al., 2015). This may have negative impacts, as it can reduce the concentration of soil cations and enhance soil acidification (Thiffault et al., 2011). Reintroducing the nutrients to the forest system by spreading ash has been introduced to increase the sustainability of the intensified harvesting (Karltun et al., 2008). Wood ash consists of a mixture of inorganic components such as silicates, oxides, hydroxides, sulphates, chlorides and a smaller part of organic phases such as charcoal (Vassilev et al., 2013). The composition can differ markedly between ashes and is determined overall by combustion type, input biomass and burning temperature (Pitman, 2006). Ashes are highly alkaline due to the content of oxides, hydroxides and carbonates (Vassilev et al., 2013) and contain high amounts of nutrients such as P, Ca, Mg and K (Ingerslev et al., 2011). Ash application is used in forests as a fertiliser to counteract nutrient losses and acidification (Huotari et al., 2015; Karltun et al., 2008), but it may have negative effects to the forest ecosystem due to the content of heavy metals (Olsson et al., 2017), dioxin and PAH's (Johansson and van Bavel, 2003; Pohlandt and Marutzky, 1994) and due to the risk of increased NO_3 leaching caused by increased nitrification (Kreutzer, 1995). The application of ash

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https://doi.org/10.1016/j.foreco.2018.05.005
Received 28 September 2017; Received in revised form 3 April 2018; Accepted 3 May 2018
Available online 21 May 2018
0378-1127/

cannot only change the total concentration of heavy metals, but also the mobility of heavy metals in the soil, as ash application changes the soil pH and adds inorganic sorbents, which can change the speciation of the heavy metals (Violante et al., 2010). Generally, an increased pH will lead to a reduced solubility of heavy metals (Shaheen et al., 2013), but, if the speciation of the heavy metal changes by increasing the fraction of complexed metals over free metal ions, it can lead to a decrease in the amount of heavy metals sorbed to minerals and thereby increase the mobility of the metal (Violante et al., 2010). To avoid the harmful effects of ash application, the current Danish legislation allows ash to be spread at a dose of maximum 3 Mg dry matter ha^{-1}. Ash can be applied with a minimum of 10-year intervals but no more than three times during 75 years (DEPA, 2008).

The specific aim of this study was to investigate the initial effects of different doses of ash application to a Norway spruce (*Picea abies* (L.)) plantation on soil and soil solution chemistry in West Denmark. This study not only includes effects on pH and nutrient changes, but also in-depth analysis of changes in heavy metal accumulation and mobility.

2. Methods

2.1. Field site description

A field experiment was established in a mature 57-year-old Norway Spruce (*Picea abies* (L.)) plantation in 2014 at Gedhus Plantation, NW Denmark (56° 16.39′ N; 9° 05.10′ E Lat/Lon, datum: WGS 84). The plantation is a second generation of plantation forest after heathland. The stand density is 457 trees ha^{-1}. One growing season after the establishment of the experiment, the average tree height was 18.8 m and the diameter at breast height was 253 cm. The stand had not been fertilised before this study commenced and the individual plots were homogeneous with respect to stand data. The area has an elevation of 51 m above sea level. The mean annual temperature is 8.4 °C and the annual mean precipitation is 850 mm (Wang, 2013). The soil has developed in glacio-fluvial sand. The soil type is an Albic Podzol (FAO, 2014) with the horizons O (6–0 cm), A (0–10 cm), E (10–20 cm), Bh (20–30 cm), Bs (30–45 cm) and C (45- cm). Texture, density and chemical composition can be seen in Table 1. Bulk density was determined horizon specifically based on the soil mass of volume specific samples in replicates of 3. Particle size distribution was determined using a Malvern Mastersizer 2000 following the DS/ISO 13320 standard. The CEC, C and N concentration was determined as described below.

2.2. Wood ash characteristics

The utilised ash was a mixed ash including bottom and fly ash from the district heating plant in Brande, Denmark, firing with coniferous wood chips (Table 2). The ash was directly collected from the heating plant two weeks prior to spreading without any stabilising pre-treatments as the ash already met the requirements set by the Danish Environmental Protection Agency (DEPA, 2008) for the utilisation of wood ashes for fertilisation purposes in forest ecosystems.

Table 2
Chemical composition of applied ash (from Maresca et al., 2018). MC: moisture content, TOC: total organic carbon, relative standard deviation is shown in brackets.

		Ash
pH		12.7
MC	%	0.15 (0.35 %)
TOC		5.84 (2.5 %)
Al	$g\ kg^{-1}$	12.4 (4.7 %)
Fe		6.72 (1.9 %)
Ca		135 (2.8 %)
K		39.4 (3.8 %)
Mg		12.7 (2.3 %)
Mn		7.43 (2.7 %)
Na		10.5 (8.0 %)
P		10.0 (2.6 %)
S		245 (5.3 %)
Cd	$mg\ kg^{-1}$	3.99 (3.4 %)
Co		5.49 (0.21 %)
Cr		26.5 (3.1 %)
Ni		16.1 (2.0 %)
Pb		13.8 (4.6 %)
Zn		340 (3.2 %)

2.3. Field experiment

The experiment was carried out as a randomised plot design. Each of the four treatments was made in a 20 × 25 m plot with three replicates, giving a total of 12 sample plots. A buffer zone of 3 m to separate each plot was established. The experimentally replicated treatments were Control (no ash application), 3, 4.5 and 6 Mg ash ha^{-1}. Additionally, two un-replicated extreme scenario plots of 7 × 8 m were included and treated with 15 and 30 Mg ash ha^{-1}, respectively. The ash was applied by hand in April 2014.

2.4. Soil samples

Soil samples were collected 2.5 years after ash application with a 50 mm diameter auger down to a depth of 60 cm. On each plot five samples were randomly collected covering the plot and divided into five depth-specific samples of O-horizon, 0–5, 5–15, 15–30 and 30–60 cm mineral soil, the samples were bulked pr. plot, giving a total of three replicates pr. treatment pr. depth. The soil was dried at 50 °C and sieved to < 2 mm and < 4 mm for the mineral and organic soil, respectively. A subsample was ground in a ball mill to < 20 µm before total C and N analysis by dry combustion (DUMAS method) using a FLASH 2000 NC Analyzer (Thermo Fisher Scientific, Waltham, MA, USA). Soil pH was determined after extraction with 0.01 M $CaCl_2$ in ratio 1:2.5 (w/v) and 1:10 (w/v) for mineral and organic soil respectively, using a Metrohm 827 pH lab meter. The concentration of exchangeable cations was determined by extraction with 1 M NH_4NO_3 before analyses by ICP-MS (iCAP Q ICP, Thermo Fisher Scientific) (Stuanes et al., 1984). Multi-elemental analysis of the heavy metal content of Cd, Co, Cr, Cu, Ni, Pb and Zn was performed by HNO_3

Table 1
Physicochemical properties of the soil at Gedhus plantation listed for the given horizons (n = 18).

Horizon	Depth	Bulk density	Texture				pH	C	N	CEC
	cm	$g\ cm^{-3}$	Clay	Silt	Fine sand	Sand		%		$cmol(+)\ kg^{-1}$
O	6-0	0.18 ±0.04					3.78	44.4 ±4.0	1.27 ±0.3	12.9 ±1.7
A	0-10	0.65 ±0.08	0.4	5.6	20.8	73.1	3.82	6.14 ±1.5	0.17 ±0.06	4.87 ±1.0
E	10-20	1.50 ±0.04	0.5	3.8	22.8	72.9	4.37	1.11 ±0.1	0.03 ±0.003	1.29 ±0.2
Bh	20-30	1.20 ±0.05	0.1	3.3	10.8	85.8	4.04	6.03 ±0.8	0.23 ±0.04	13.7 ±0.6
Bs	30-45	1.42 ±0.04	0.0	0.0	3.0	97.0	4.61	2.61 ±3.0	0.09 ±0.1	2.70 ±0.6
C	45-	1.49 ±0.02	0.0	0.0	1.9	98.1	4.92	0.21 ±0.2	0.01 ±0.006	0.95 ±0.08