The effect of granulated wood-ash fertilization on soil properties and greenhouse gas (GHG) emissions in boreal peatland forests

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The amount of wood ash produced in power plants is increasing with increasing use of forest biomass in energy production. Wood ash can be recycled as fertilizer especially in boreal peatland forests naturally rich in nitrogen. Improved nutrient availability and increases in soil pH can enhance microbial activities, decomposition of organic matter and greenhouse gas (GHG) emissions. We studied the effects of granulated wood-ash on soil chemical properties, vegetation characteristics, decomposition rate and fluxes of nitrous oxide (N₂O) and methane (CH₄) in boreal peatland forests. In addition to the field measurements, we conducted laboratory experiments. Wood-ash fertilization changed soil chemical properties, altered understory vegetation, increased tree growth and decomposition rate but there were no significant changes in the N₂O and CH₄ fluxes in situ, whereas in laboratory incubations ash decreased the N₂O production rate. The results show that there is no major risk of increasing GHG emissions after granulated wood-ash fertilization in boreal peatland forests.

Introduction

The EU countries have committed to increase their use of energy from renewable sources up to 20% by 2020. The target for Finland is even higher, 38%. To achieve this goal, the use of bioenergy is increasing rapidly in Finland, for example production of forest chips is to be doubled to 13.5 million m³ by 2020 (Ministry of Employment and Economy 2010). At the moment, about 400 000 tonnes of wood or mixed wood and peat ash is produced in power plants annually (Energiateollisuus 2013). The increase in the use of forest chips for energy generation implies a considerable increase in the wood-ash production. Previously, wood ash was considered waste and was disposed of in landfills or used e.g. in road construction. However, nutrients in ash can also be recycled as fertilizer especially in peatland forestry (Moilanen et al. 2002, Hytönen et al. 2003, 2012). Currently, ash is spread as forest fertilizer annually to nearly 10 000 ha, mostly drained peatland forests (Finnish Statistical Yearbook of Forestry 2011). Except nitrogen, wood ash contains other nutrients, like phosphorus, potassium and boron.
which can be recycled as fertilizer at nitrogen-rich peatland forest sites. However, in mineral soils where tree growth is limited by a shortage of nitrogen, ash fertilization does not improve tree stand growth (Jacobson 2003, Aronsson and Ekelund 2004, Moilanen et al. 2013). Besides plant nutrients, wood ash contains cadmium and other heavy metals such as mercury, lead, arsenic, cobalt, chromium and nickel (Demeyer et al. 2001, Aros and Ekelund 2004). Therefore, it cannot be applied as fertilizer in agricultural soils. Wood ash is alkaline (pH 10–13) increasing soil pH (Aronsson and Ekelund 2004) and can therefore lower toxicity of aluminium and manganese, which are soluble in acidic conditions (Demeyer et al. 2001).

Drained peat soils are significant sources of carbon dioxide (CO$_2$) and nutrient-rich peat soils can also be sources of nitrous oxide (N$_2$O), but drained peat soils emit less methane (CH$_4$) than natural peatlands (Maljanen et al. 2010a). When wood ash is used as fertilizer, it is important to know if the enhanced microbial activities resulting from changes in soil chemical characteristics (Demeyer et al. 2001) would increase the greenhouse gas (GHG) emissions. Most of the studies on the effects of wood ash on tree stand growth, on soil nutrient status and on GHG balance were conducted with dust-like loose ash. Today, mostly granulated or self-hardened ash is used because it is easier and cheaper to spread (Väätäinen et al. 2011), and granulation and hardening allows for the slow release of nutrients from ash (Nieminen et al. 2005, Pitman 2006). Thus, granulated ash would increase soil pH less than loose ash. Therefore, CO$_2$ and N$_2$O production could be affected less by granulated ash than with the loose ash studied earlier. However, since ash fertilization is long-lasting, it is possible that the short-term effects differ from the long-term effects. To test these hypotheses we measured soil chemical properties, N$_2$O and CH$_4$ flux rates and soil respiration (CO$_2$ production rate) in three Finnish boreal peatland forests with different nutrient characteristics fertilized with granulated wood-ash. We also studied the immediate effect of ash treatments on N$_2$O production in laboratory experiments. Results are then compared with the earlier results obtained in the experiments with loose or self-hardened ash.

**Material and methods**

**Study sites**

The three study sites were located in western Finland, near Kannus, within 40 km of each other (Table 1). Granulated wood-ash (5000 kg ha$^{-1}$, dry ash) was used at all sites. Before spreading, the moisture content of ashes was determined and ash dry-weight amounts were calculated accordingly. Ash was applied at Site 1 in May 2010 and at Sites 2 and 3 in 2003. The size of the study plots varied from 32 m$^2$ (Site 1) to 1500 m$^2$ (Sites 2 and 3). The ash applied at Site 1 contained P 10, K 25, Ca 160, Mg 19 and B 0.25 g kg$^{-1}$ as total amounts and the ash applied at Sites 2 and 3 contained P 15, K 38, Ca 167, Mg 27 and B 0.32 g kg$^{-1}$. Our measurements at Sites 1 and 2 started in May 2010 and at Site 3 in May 2011.

The studied peatland forests were drained to improve forest growth (Site 1 in the 1960s, Site 2 in the early 1990s and Site 3 in 1974). Sites 1 and 2 are classified according to Laine et al. (2012) as Vaccinium vitis-idaea type peatland forests (Ptkg) and Site 3 was originally less fertile low sedge bog developing towards Cladonia type peatland forest (Jätkg). In Finland Ptkg is the most common drained peatland forest type covering 37% of the total 5.7 million ha drained for peatland forestry. The Jätkg type peatland forests cover less than 2% of the total drained peatland area (Laine et al. 2012, Turunen 2008).

The long term average (LTA) annual temperature (1971–2001) in Kannus is 2.8 °C and the average annual precipitation is 561 mm (Drebs et al. 2002). Of the total precipitation approximately 50% falls as snow. Snow cover typically appears in mid-November and melts in late April. The coldest month is February (−9.2 °C), and the warmest is July (15.8 °C) (Drebs et al. 2002). The LTA maximum snow depth is 44 cm (obtained in mid-March).

**Environmental variables**

Soil temperature was measured manually (depth 0–20 cm) close to the chambers from non-frozen soil during gas sampling. At Site 1, soil tem-
perature was also recorded using temperature probes (109 Thermistor Probe) and a data logger (CR200, Campbell Scientific, UK). Soil temperatures at Sites 2 and 3 were recorded with iButton® temperature loggers (Dallas Semiconductor Corp., USA). Air temperature and daily precipitation were recorded at the Toholampi weather station within 20 km of the study sites. Water table depth (WT) was measured in groundwater pipes (n = 4–5) installed at the study plots. WT was not recorded during the winter months when the topsoil was frozen.

Soil samples for analyses of pH, electrical conductivity (EC), nitrate (NO$_3^-$), ammonium (NH$_4^+$) and dissolved organic carbon (DOC) were collected from a depth of 0–20 cm (n = 5, pooled for the analysis) three times in 2011 and 2012 growing seasons (June, July/August and September). NO$_3^-$ was extracted with distilled H$_2$O and NH$_4^+$ with 1M KCl solution. The amount of NO$_3^-$ was analyzed using an ion chromatograph (DX 120, Dionex Corporation, USA) and NH$_4^+$ with a spectrophotometer (Ultrospec 3000 Pro, Biochrom, UK) using the method

Table 1. Characteristics of the study sites (analysis from pooled soil samples, depth 0–20 cm). C = Control, A = Ash fertilization (5000 kg granulated wood-ash per ha). Asterisks (*) indicate significant difference between ash and control at p < 0.05 (one-way ANOVA), see text for details.

| Site          | Location          | Dominant tree species | Age of trees | Peatland classification according to Laine et al. (2012) | Site volume (m$^3$ ha$^{-1}$) | Increase in stem wood volume in 5 years (m$^3$ ha$^{-1}$) | pH (H$_2$O)$^a$ | EC (µS cm$^{-1}$)$^a$ | NO$_3^-$N (µg g$^{-1}$)$^a$ | NH$_4^+$-N (µg g$^{-1}$)$^a$ | DOC (mg g$^{-1}$)$^a$ | Cl$^-$ (µg g$^{-1}$)$^a$ | SO$_4^{2-}$ (µg g$^{-1}$)$^a$ | Peat depth (cm)$^b$ | Bulk density (g cm$^{-3}$)$^b$ | Degree of decomposition$^b$ (H$^c$) | Organic matter (OM, %)$^b$ | C (%)$^b$ | N (%)$^b$ | C:N$^b$ | P (mg g$^{-1}$)$^b$ | K (mg g$^{-1}$)$^b$ | B (µg g$^{-1}$)$^b$ | Ca (mg g$^{-1}$)$^b$ | Mg (mg g$^{-1}$)$^b$ | Mn (µg g$^{-1}$)$^b$ | Cd (µg g$^{-1}$)$^b$
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<td>9.5 ± 10</td>
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<td>0.9 ± 1.1</td>
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<td>2.7</td>
<td>18</td>
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$^a$ Average values from growing seasons 2011 and 2012 ± SD (n = 6, six sampling times).

$^b$ Measured once in June 2011 from a pooled sample.

$^c$ von Post scale.
of Fawcett and Scott (1960). DOC concentrations in soil were measured from 0.25 M K$_2$SO$_4$ extracts with TOC analyzer (Shimadzu TOC Vcph, Shimadzu Scientific, Japan).

Cellulose and teabag decomposition rate tests were carried out in 2011, from June 6 to 11 September. Pieces of birch cellulose ($5 \times 10$ cm) were dried at 105 °C, stabilized for two hours at room temperature and weighed. A cellulose piece was inserted in a plastic net (mesh size 1 mm) and buried in the peat at each site at the depths of 5 and 20 cm ($n = 5$). After the in situ incubation period, the ingrown roots and mosses were cleaned off, and the pieces were dried and weighed in the same way as before installation. Cellulose decomposability was calculated from the weight loss. In addition to cellulose, green and rooibos teas were used in the decomposition tests (Keuskamp et al. 2013). Two tea varieties in nylon mesh bags (Lipton® Rooibos Tea and Lipton® Green Tea) were buried at a depth of 7 cm ($n = 5$) and the decomposition rates were calculated similarly as for the pieces of cellulose.

Root biomass was measured from samples collected from the study sites in June 2011 (Sites 2 and 3) and September 2012 (Site 1). Samples ($n = 10$ at Sites 2 and 3, $n = 12$ at Site 1) were taken from the depths of 0–10 cm and 10–20 cm with a soil corer (diam. 8.3 cm). Roots were separated manually, washed and dried at 65 °C. The root biomass was calculated as kg ha$^{-1}$ from samples taken from the top 0–10 and 10–20 cm. For the measurement of the stand volume, a sub-sample plot (100–400 m$^2$) was established inside each study plot. Diameter at breast height ($d_{1.3}$) of all trees was recorded, and height was measured from 18 to 21 randomly chosen sample trees of Scots pine, representing different size classes. The heights of all trees were predicted from the fitted Näslund’s height curve. At Site 2 trees were measured in autumn 2007 and measurements were repeated in autumn 2012. At Site 3 the measurements were made in February 2010 and repeated in autumn 2012. The stem volumes of the trees were computed applying the models of Laasasenaho (1982) and the mean annual growth of 5 growing seasons was calculated. Stem volume growth was not measured in the short-time experiment at Site 1.

For the measurement of the soil CO$_2$ efflux (at Sites 2 and 3) aluminium cylinders 31.5 cm in diam. were inserted into the soil in the middle of May 2011 to a depth of 30 cm to exclude root respiration from the soil CO$_2$ efflux ($n = 10$ at Site 2 and $n = 5$ at Site 3). Above-ground litter was removed from the cylinders, and in order to eliminate autotrophic plant respiration the above-ground parts of the green plants were removed by manual weeding and clipping before the measurements. Thus, the measured CO$_2$ efflux represented the decomposition of soil organic matter and cut roots, similarly for control and ash treatments. The soil CO$_2$ efflux was measured (starting from 6 June 2011) using a closed-chamber system with air circulating in a loop between the chamber and an external infrared gas analyser (IRGA) (EGM-4 CO$_2$ Analyzer, PP-Systems Ltd. UK) equipped with a water vapour equilibrator. During sampling (1–2 min) an aluminium chamber (height 14.9 cm, basal area 779 cm$^2$, equipped with a fan) was installed inside the cylinder into a 2 cm deep groove in the soil.

Gas flux measurements

The N$_2$O and CH$_4$ flux rates from the snow-free soil were measured using a static chamber method with aluminium chambers equipped with a fan (60 × 60 cm, height 30 cm) and aluminium collars (60 × 60 cm, height 30 cm) pre-installed in the soil in the control and ash-fertilized plots ($n = 6$ at Site 1 and $n = 4$ at Sites 2 and 3). After closing the chamber, gas samples of 30 ml were taken with a 60 ml polypropylene syringe (Terumo) at 5, 15, 25 and 35 min intervals from the headspace of the chamber. Samples were injected within 24 hours into pre-evacuated 12 ml vials (Labco Excetainer®) and analyzed for N$_2$O and CH$_4$ with a gas chromatograph (Agilent 6890N, Agilent Technologies, USA) equipped with an autosampler (Gilson, USA) and electron capture (ECD) and flame ionization (FID) detectors. Compressed air containing 0.389 µl l$^{-1}$ of N$_2$O and 1.98 µl l$^{-1}$ CH$_4$ was used for daily calibration.

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The flux rates were calculated from the linear increase or decrease in the gas concentrations in the headspace of the chamber with time. If there were any indications of failures in the gas sampling or gas analysis the results were excluded.

The \( \text{N}_2\text{O} \) and \( \text{CH}_4 \) fluxes from the snow-covered plots, when the snow depth exceeded 10 cm, were determined by measuring gas concentration gradients from the snow 2 cm above the soil surface and from the ambient air and by calculating associated diffusion rates in the snow from the snowpack density (Maljanen et al. 2003). Gas samples (30 ml) were drawn with a stainless steel probe (diam. 3 mm, length 100 cm) from the snow inside the collars installed for the chamber method. Simultaneously, snow samples were collected with a PVC tube (diam. 10.2 cm) for porosity measurements. The intact samples were weighed for calculation of the average porosity of snow using the density of pure ice (0.9168 g cm\(^{-3}\)).

**Gas concentrations in soil**

The \( \text{N}_2\text{O} \) and \( \text{CH}_4 \) concentrations in soil were measured simultaneously with the gas flux measurements. Gas samples of 30 ml were taken with syringes from the pre-installed silicon tubes (diam. 1.0 cm, wall thickness 0.3 cm, length 110 cm, \( V = 86 \text{ cm}^3 \)) inserted horizontally in the peat at depths of 5 and 20 cm (\( n = 2 \)) and also at 40 cm (\( n = 2 \)) at Sites 1 and 2. Samples were treated and analyzed for \( \text{N}_2\text{O} \) and \( \text{CH}_4 \) with a gas chromatograph as described above.

**Laboratory experiments**

In addition to the field measurements, laboratory incubations with peat soils were carried out to study the effect of incubation temperature and ash dose on soil chemical properties and \( \text{N}_2\text{O} \) production rates.

In the first incubation experiment, unfertilized soil from Site 1 and the same granulated wood-ash which was applied in the field were used. 30 g (FW) of soil was weighed in 550 ml incubation flasks with 0 or 1.6 g of granulated wood-ash. The flasks (\( n = 5 \)) were incubated at –6.4, –3.1, +4.1 and +12.5 °C for 10 weeks. The \( \text{N}_2\text{O} \) production rate was measured weekly during the first four weeks and then after six, eight and 10 weeks of incubation. Prior to gas sampling, the flasks were flushed with ambient air, then sealed with a rubber septum and finally 60 ml of ambient air was added through the septum to ensure overpressure for sampling. Gas samples (20 ml) were taken with a needle through the rubber septum 1, 2, 4, and 6 h after closing the flask and \( \text{N}_2\text{O} \) concentrations were analyzed with a gas chromatograph as described above. After the last sampling, 2.5% of acetylene (\( \text{C}_2\text{H}_2 \)) was added to the headspace of the flasks and the \( \text{N}_2\text{O} \) production was measured again with \( \text{C}_2\text{H}_2 \) inhibition. The soil pH, EC and \( \text{NO}_3^- \), \( \text{NH}_4^+ \) and DOC concentrations were measured as described earlier after an incubation period lasting 10 weeks.

In the second incubation experiment, the same peat soils from Site 1 and wood ash were used. 30 g (FW) of soil was weighed in 550 ml incubation flasks and 0, 0.8, 1.6 or 3.2 g of wood ash granules were mixed with the soil (\( n = 5 \)). The flasks were incubated at 15 °C for three weeks and the production rate of \( \text{N}_2\text{O} \) was measured as described above. Soil pH and EC were measured after incubation as described earlier.

**Statistical methods**

The data were tested for normality and homogeneity of variances using the Kolmogorov-Smirnov test and Levene’s test. Differences in environmental variables were tested with one-way ANOVA. The differences in gas flux rates and gas concentrations were studied with non-parametric tests (Mann-Whitney \( U \)-test). Correlations between gas fluxes and gas concentrations in soil were studied with non-parametric Spearman rank correlation test (IBM SPSS ver. 19).

**Results**

**Soil properties and vegetation characteristics**

The average daily air temperature during the
study period varied from –29.2 °C to 25.1 °C (Figs. 1–3). The water table depth during the snow free period varied from 5 to 38 cm at Site 1, from 4 to 37 cm at Site 2 and from 2 to 26 cm at Site 3 (Figs. 1–3 and Table 2).

Ash addition increased the SO$_4^{2-}$ concentrations in soil at Site 1 (one-way ANOVA: $F_{1,11} = 5.75, p = 0.038$) and Site 2 (one-way ANOVA: $F_{1,11} = 10.96, p = 0.008$). The mean soil pH was 0.3 units higher at the old-ash fertilized Sites 2 and 3 but there were no statistical differences. There was no change in pH at Site 1 fertilized two years before soil analysis. The B, Ca, Mg and Mn concentrations analyzed from pooled samples were higher in the ash-fertilized plots than in the control plots but ash fertilization had only minor effects on the soil total P concentrations and electrical conductivity in situ. However, at the old sites (2 and 3), the soil electrical conductivity was higher with ash in the deeper (20–40 cm) soil layers, but not at Site 1 (results not shown). Ash increased the soil total K concentration at the more fertile Sites 1 and 2 but not at the less fertile Site 3 (Table 1). The Cd concentration in peat was 1.6–7 times higher after ash treatment (Table 1). Ash addition had
here negligible effects on the DOC, NH$_4^+$ and NO$_3^-$ concentrations in peat.

Tree growth in 10 years in the ash-fertilized plots was 1.7–3 fold higher than in the control plots at Sites 2 and 3, respectively (Table 1). The increase in the tree growth was not measured in the short term fertilization experiment at Site 1 but changes in the understory vegetation at Site 1 were recorded. The collars for gas flux measurements were originally installed in pairs (one in an ash-fertilized plot and one in a control plot) on similar vegetation surfaces. By the end of the experiment, there were changes in vegetation in the ash-fertilized plots, as the coverage of grasses and herbs e.g. Trientalis europeana, Epilobium angustifolium, Vaccinium myrtillus and Dryopteris carthusiana increased. Also the total coverage of understory vegetation was higher in the ash-fertilized plot at Site 1 (one-way ANOVA: $F_{1,11} = 12.5, p = 0.005$) (Table 2). The effect was not significant at Sites 2 and 3. At Site 2 the mean coverage of grasses and herbs was higher in the control plots, and there were fewer mosses in the ash-fertilized plots. The total root biomass (depth 0–20 cm, < 2 mm) was higher (one-way ANOVA: $F_{1,11} = 6.78, p = 0.018$) in the
ash-fertilized plot than in the control plot at Site 3 but not at Sites 1 and 2 (Table 2). However, in the top 0–10 cm layer the root biomass was higher in the ash-fertilized plots at Site 1 (one-way ANOVA: $F_{1,11} = 5.56, p = 0.028$) and at Site 3 (one-way ANOVA: $F_{1,11} = 6.93, p = 0.017$), but there was no significant difference at Site 2.

The cellulose decomposition rate in the control plots was fastest at Site 1 and the slowest at Site 3. The decomposition rate at Site 2 was between these two extremes (Table 2). Ash fertilization increased cellulose decomposition rates in the top 10 cm at Site 2 (one-way ANOVA: $F_{1,9} = 10.4, p = 0.012$) and in the 10–20 cm layer at Site 3 (one-way ANOVA: $F_{1,9} = 7.5, p = 0.025$) but not at the newly-fertilized Site 1. Green tea decomposed faster than rooibos tea (one-way ANOVA: $F_{2,59} = 262, p < 0.001$) (Table 2). In contrast to cellulose decomposition, there were no statistical differences in the tea decomposition rates among the sites. At Site 3, the decomposition rate of rooibos tea was faster in the ash-fertilized plot (one-way ANOVA: $F_{1,9} = 8.64, p = 0.019$), but at the other sites there were no statistical differences.
Table 2. Average flux rates (FN$_2$O, FCH$_4$) and concentrations of N$_2$O, CH$_4$ in soil air, soil respiration rates, decomposition rates and vegetation characteristics (± SD). C = Control, A = Ash fertilization (5000 kg granulated wood-ash ha$^{-1}$). CD= cellulose decomposition rate, TD = tea decomposition rate, SR = soil respiration rate, RB = root biomass. Asterisks (*) indicate significant difference between ash treatment and control at $p < 0.05$ (Mann-Whitney U-Test or one-way ANOVA), see text for details.

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<th>Site</th>
<th>C</th>
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<td>May–Sep. 2011</td>
<td>580 ± 530</td>
<td>495 ± 510</td>
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<tr>
<td>5 cm</td>
<td>5.0 ± 19</td>
<td>4.9 ± 9.4*</td>
<td>0.58 ± 0.44</td>
<td>0.87 ± 1.1*</td>
<td>0.34 ± 0.22</td>
<td>0.37 ± 0.20*</td>
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<tr>
<td>20 cm</td>
<td>25 ± 60</td>
<td>43 ± 120*</td>
<td>1.0 ± 1.1</td>
<td>4.0 ± 7.6*</td>
<td>0.13 ± 0.09</td>
<td>0.14 ± 0.10</td>
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<tr>
<td>40 cm</td>
<td>41 ± 160</td>
<td>96 ± 240*</td>
<td>9.7 ± 19</td>
<td>11 ± 21</td>
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<td>N$_2$O in soil air, (ppm) May–Sep. 2011</td>
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<tr>
<td>5 cm</td>
<td>1.9 ± 0.16</td>
<td>1.9 ± 0.13</td>
<td>1.7 ± 0.26</td>
<td>1.5 ± 0.40*</td>
<td>1280 ± 6000</td>
<td>3800 ± 13000*</td>
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<tr>
<td>20 cm</td>
<td>3.0 ± 4.7</td>
<td>2.0 ± 0.44*</td>
<td>8.8 ± 0.49</td>
<td>1.3 ± 1.6*</td>
<td>104000 ± 73000</td>
<td>33700 ± 47000*</td>
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<tr>
<td>40 cm</td>
<td>2.0 ± 0.58</td>
<td>2.2 ± 2.2</td>
<td>16.5 ± 74</td>
<td>3.7 ± 12</td>
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<td>CH$_4$ in soil air (ppm) May–Sep. 2011</td>
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<td>3.7 ± 12</td>
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<td>Water table level (cm) May–Sep. 2010</td>
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<td>May–Sep. 2010</td>
<td>–49 ± 18</td>
<td>–49 ± 18</td>
<td>–40 ± 19</td>
<td>–44 ± 21</td>
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<td>CD (loss %) May–Sep. 2011</td>
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<tr>
<td>10 cm</td>
<td>25 ± 17</td>
<td>23.0 ± 6.3</td>
<td>22 ± 2.2</td>
<td>32 ± 6.7*</td>
<td>15 ± 2.8</td>
<td>11 ± 7.1</td>
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<tr>
<td>20 cm</td>
<td>17 ± 9.3</td>
<td>22 ± 9.4</td>
<td>3.3 ± 2.7</td>
<td>7.0 ± 2.8</td>
<td>1.4 ± 1.0</td>
<td>11 ± 7.5*</td>
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<td>TD (loss %) May–Sep. 2011</td>
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<tr>
<td>Green tea, 7 cm</td>
<td>62 ± 4.9</td>
<td>61 ± 4.4</td>
<td>64 ± 5.4</td>
<td>68 ± 8.2</td>
<td>60 ± 3.3</td>
<td>60 ± 3.1</td>
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<tr>
<td>Rooibos tea, 7 cm</td>
<td>44 ± 1.4</td>
<td>45 ± 3.4</td>
<td>45 ± 4.1</td>
<td>47 ± 1.6</td>
<td>42 ± 1.2</td>
<td>44 ± 0.80*</td>
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<td>RB, &lt;2 mm (kg ha$^{-1}$)</td>
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<td>Understory plant coverage (%) in August 2012</td>
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<td>0–10 cm</td>
<td>1800 ± 410</td>
<td>2300 ± 700*</td>
<td>3600 ± 1200</td>
<td>3100 ± 1400</td>
<td>1700 ± 990</td>
<td>2900 ± 1200*</td>
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<tr>
<td>10–20 cm</td>
<td>550 ± 250</td>
<td>730 ± 270</td>
<td>460 ± 280</td>
<td>510 ± 580*</td>
<td>590 ± 370</td>
<td>700 ± 360*</td>
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<tr>
<td>40 cm</td>
<td>2.0 ± 0.58</td>
<td>2.2 ± 2.2</td>
<td>16.5 ± 74</td>
<td>3.7 ± 12</td>
<td>nd</td>
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<td>RB, &lt;2 mm (kg ha$^{-1}$)</td>
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<tr>
<td>Understory plant coverage (%) in August 2012</td>
<td>56.8 ± 15.1</td>
<td>81.5 ± 8.01*</td>
<td>61.3 ± 21.2</td>
<td>46.5 ± 3.70</td>
<td>65.3 ± 16.0</td>
<td>73.3 ± 11.3</td>
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</table>
**In situ gas flux rates**

There were no statistical differences in the N$_2$O and CH$_4$ flux rates *in situ* among the ash fertilized and control plots, but ash fertilization increased soil respiration at Sites 2 and 3 (Mann-Whitney U-test: $U = 3608$, $p < 0.001$ and $U = 309$, $p < 0.001$, respectively) (Table 2). The soil respiration was not measured at Site 1. The N$_2$O emission rates varied largely among the sites and seasons (Figs. 1–3). The N$_2$O emissions from Site 1 were two magnitudes higher than those from Site 2 (Table 2). Site 3 even occasionally showed N$_2$O uptake and the average emissions were always lower than those at Site 2. The highest emissions were measured during the growing season (June–July), and emissions during autumn and winter were lower (Figs. 1–3). During the first winter there was a small N$_2$O emission peak in January when the topsoil was frozen at Sites 1 and 2 (Figs. 1–3). The N$_2$O and CH$_4$ emission dynamics were similar in the control and ash-fertilized plots. There were methane emissions at Site 3 whereas Sites 1 and 2 acted as small net sinks for atmospheric CH$_4$ (Figs. 1–3 and Table 2). The highest methane emissions were measured in the late growing season (Site 3) and the highest net CH$_4$ uptake rates in the early growing season (Sites 1 and 2).

**Soil gas concentrations**

The average N$_2$O concentrations in soil at 5 cm depth were higher at Site 1 than at Sites 2 and 3 (Mann-Whitney U-test: $U = 1195$, $p < 0.001$ and $U = 97.0$, $p < 0.001$, respectively) (Table 2). The N$_2$O concentrations in the deeper (20 and 40 cm) peat profiles were higher at Sites 1 and 2 but were below ambient concentration (0.3 ppm) at Site 3. There was a correlation between the soil N$_2$O concentrations and gas fluxes only at Site 2 where the N$_2$O concentrations at depths of 20 and 40 cm correlated positively but weakly with the N$_2$O fluxes (Spearman rank correlation: $r_S = 0.235$, $p = 0.005$; and $r_S = 0.272$, $p = 0.001$; respectively). The N$_2$O concentrations at 20 and 40 cm depths were higher in the ash-fertilized plots than in the control plots at Site 1 (Mann-Whitney U-test: $U = 1905$, $p = 0.025$ and $U = 1495$, $p < 0.001$) and at 5 and 20 cm depths at Site 2 (Mann-Whitney U-test: $U = 641$, $p = 0.007$ and $U = 417$, $p < 0.001$). However, there were no statistical differences in the measured *in situ* N$_2$O flux rates between the control and ash-fertilized plots.

The CH$_4$ concentrations in the peat (0–40 cm) at Site 1 remained close to the ambient level of 2 ppm, but at Sites 2 and 3 the CH$_4$ concentration increased with depth (Table 2). The CH$_4$ concentration in soil air at 5 and 20 cm depths were lower in ash treated plots at Site 2 (Mann-Whitney U-test: $U = 639$, $p = 0.001$; and $U = 464$, $p < 0.001$; respectively) (Table 2). Very high CH$_4$ concentrations (up to 200 000 ppm) were measured at Site 3 at 20 cm depth, below the water table level. Only at Site 1, the CH$_4$ concentrations (depth 5 cm) correlated weakly with the CH$_4$ flux rates (Spearman rank correlation: $r_S = 0.228$, $p = 0.005$).

**Laboratory incubation experiments**

In the first laboratory incubation experiment using different temperatures with soil from Site 1, the N$_2$O production rates at temperatures below 0 °C were low but the N$_2$O production rates increased with increasing temperature (Fig. 4). The production rates at various incubation temperatures did not change significantly during the incubation time of 10 weeks (data not shown). Ash addition (1.6 g in 30 g of fresh soil, corresponding 5000 kg ha$^{-1}$) decreased the N$_2$O production 70%–80% at temperatures above 0 °C as compared with that in the control (one-way ANOVA: $F_{1,9} = 1323$, $p < 0.001$ and $F_{1,9} = 68.2$, $p < 0.001$, respectively). Additions of 2.5% acetylene into the headspace inhibited 80%–90% of the N$_2$O production (one-way ANOVA: $F_{1,19} = 11.9$, $p = 0.003$; and $F_{1,19} = 11.0$, $p = 0.004$; respectively). At temperatures below 0 °C there were no significant effects. The NO$_3$- and DOC concentrations were lower in ash-fertilized soils when incubated at temperatures above zero (one-way ANOVA: $F_{1,19} = 5.92$, $p = 0.026$; and $F_{1,19} = 9.86$, $p = 0.006$; respectively), whereas the NH$_4^+$ concentration was higher in ash-treated soil (one-way ANOVA: $F_{1,19} = 5.59$, $p = 0.029$) (Fig. 4). In the second experiment, the N$_2$O production also
decreased as a result of the ash treatment and a dose response was evident (Fig. 5). Normal (corresponding 5000 kg ha\(^{-1}\)) ash and double ash doses both significantly reduced the \(\text{N}_2\text{O}\) production (see Fig. 5). The effect of a half-dose (corresponding 2500 kg ha\(^{-1}\)) was not significant. Soil pH did not change with an increasing ash dose but EC did (see Fig. 5).

**Discussion**

The studied peatland forests (unfertilized control sites) had very different annual \(\text{N}_2\text{O}\) emissions, from 0.03 to 2.90 g m\(^{-2}\). The highest \(\text{N}_2\text{O}\) emissions took place at fertile Site 1 (Ptkg), whereas poor Site 3 (Jätkg) had negligible \(\text{N}_2\text{O}\) emissions (Tables 1 and 2). The annual \(\text{N}_2\text{O}\) emission from Site 1 was about 40 times higher than the average emissions from the Finnish Ptkg types, whereas the emissions from Site 2 (Ptkg) were similar to the average emissions (Ojanen et al. 2010). As expected, based on the high water table level at Site 3, the \(\text{N}_2\text{O}\) emissions from Site 3 were negligible. However, there were no significant differences in the water table levels between Sites 1 and 2 which could explain their different \(\text{N}_2\text{O}\) emission rates (Martikainen et al. 1993). Peat humification at the site drained about 50 years ago was higher than at Site 2 drained
20 years ago. This could partly explain the higher N$_2$O emissions from Site 1 (von Arnold et al. 2005, Ojanen et al. 2010). Klemedtsson et al. (2005) suggested that the N$_2$O emissions increased with the decreasing C:N ratio of peat, the threshold ratio being around 20. Above that threshold the emissions are low. Peat at Site 3 had high a C:N ratio (82–87) and low N$_2$O emissions, which supports this conclusion. However, the C:N ratios at Sites 1 and 2 were almost similar (18 and 21) but there was 20 fold differences in their N$_2$O emission rates as well as in the NO$_3^-$ concentrations in their peat. Site 1 has no agricultural history, which could explain this inconsistency (Ojanen et al. 2010). Also the CH$_4$ fluxes differed between the sites. Sites 1 and 2 (Ptkg) were sinks for CH$_4$ annually, whereas Site 3 (Jätkg) with a rather high water table level was a small CH$_4$ source. Site 2 (Ptkg) had a higher net CH$_4$ uptake rate than Site 1 even though this site had slightly lower mean water table level which should support CH$_4$ oxidation in the peat profile. The lower CH$_4$ concentration in the uppermost peat profile of Site 2 also indicated that CH$_4$ produced in deeper peat is effectively oxidized in the topsoil.

Granulated wood-ash fertilization (5000 kg ha$^{-1}$) in the studied peatland forests altered soil chemical properties, increased tree growth and caused some changes in the understory vegetation composition. However, ash treatment did not affect the N$_2$O and CH$_4$ fluxes in situ, but increased the soil respiration and cellulose decomposition rates. Increase in soil pH, higher availability of mineral nutrients and increase in DOC with in situ ash application have been reported in several studies made with loose ash (Demeyer et al. 2001, Saarsalmi et al. 2001, Moilanen et al. 2002, 2012, 2013, Jokinen et al. 2006, Mandre et al. 2010, Hytönen and Aro 2012). Here the changes in soil pH were not statistically significant, the difference in the mean values was only 0.3 pH units. In situ granulated wood-ash application did not significantly change electrical conductivity. However, in the laboratory experiments where ash granules were mixed with peat, EC increased with an increasing ash dose. In contrast to earlier studies with loose ash (Moilanen et al. 2002, Jokinen et al. 2006, Saarsalmi et al. 2012) or self-hardened ash (Norström et al. 2012), we did not find any changes in the soil NO$_3^-$, NH$_4^+$ or DOC concentrations in situ after ash fertilization. However, granulated wood-ash increased the B, Ca, Mg and Mn concentrations measured from pooled samples of soil but less the concentrations of P and K. It is possible that, especially at the nutrient poor sites, growing trees are using the extra K and P. Potassium is also known to leach out easily, especially from poor sites dominated by Sphagnum peat (Demeyer et al. 2001, Piirainen et al. 2013).

There were only minor effects after granulated ash fertilization on the N$_2$O and CH$_4$ fluxes in situ, similarly as obtained in studies with loose ash (Moilanen et al. 2002, Maljanen et al. 2006a, 2006b) or with self-hardened ash (Ernfors et al. 2010, Klemedtsson et al. 2010). However, there were no statistical differences in the seasonal N$_2$O and CH$_4$ flux rates between the control plots and plots treated with granulated ash. No N$_2$O emission peaks were detected
during winter as reported earlier (Maljanen et al. 2010b, Klemedtsson et al. 2010). Also, no significant reduction in the N$_2$O emission rates was found during winter as was found for ash treatments by Klemedtsson et al. (2010). In an earlier study with unfertilized soil at Site 1 (Maljanen et al. 2010a), there was a significant N$_2$O emission peak when soil frost was developing. In the present study, there was only a slight increase in the N$_2$O emission from the fertilized and control plots at Sites 1 and 2 when top soil was freezing during the first winter (Fig. 1). However, some emission peaks may have been missed due to infrequent manual sampling. In contrast to in situ measurements, there was a reduction in the N$_2$O production rates after the ash application in the laboratory experiments. Odłare and Pell (2009) also reported similar decrease in the N$_2$O production as a result of ash addition in laboratory conditions. The reasons for the decrease are not fully understood. It could be associated with the heavy metals in the ash (Holtan-Hartwig et al. 2002). Osmotic effect of the ions derived from ash, or increase in soil pH could enhance activity of the N$_2$O reductase (Bakken et al. 2012). In the laboratory experiments granulated ash was mixed with homogenized peat, whereas in situ ash was spread over the peat surface. Therefore, ash could have changed soil chemistry and biology less in situ. Surprisingly, in the laboratory experiments with various ash doses there was no increase in soil pH. In the control soil, there was a slight decrease in pH during the 10-week incubation. Enhanced nitrification producing H$^+$ ions could be a reason for this (Rice and Herman 2012). The N$_2$O production decreased after the addition of 2.5% acetylene, a concentration inhibiting both ammonium oxidation and the N$_2$O reduction to N$_2$ in denitrification (Klemedtsson et al. 1988). Therefore, ammonium oxidizing nitrifiers could play a role in the N$_2$O production in peat at Site 1.

Respiration and cellulose decomposition were still enhanced by the granulated ash more than 10 years after the fertilization at Sites 2 and 3. This indicates higher decomposition rates in ash-treated peat soil supporting the earlier results with loose ash (Fritze et al. 1994, 1995, Moilanen et al. 2002, 2012) and granulated ash (Rosenberg et al. 2010). In those studies, granulated wood-ash increased microbial activity, decomposition and also C loss in acidic mineral forest soils and in peat soils even though there was only a minor increase in soil pH. The increase in biomass growth (C accumulation in biomass) could compensate for the C loss from peat in forested peatlands (Lohila et al. 2011, Moilanen et al. 2012). The net C-balance of drained and forested boreal peatlands is still uncertain. Lohila et al. (2011) showed by eddy covariance measurements that some forested peatlands can be large net CO$_2$ sinks (on average ~870 g m$^{-2}$ yr$^{-1}$) when carbon accumulated in the tree biomass is included. Based on measurements and modeling, Ojanen et al. (2013) reported that peat soils at nutrient-poor sites are CO$_2$ sinks (~70 ± 30 g m$^{-2}$ yr$^{-1}$) but peat soils at fertile sites are net sources of CO$_2$ (190 ± 70 g m$^{-2}$ yr$^{-1}$). Using the C inventory of peatlands located across central Finland, Simola et al. (2012) found that there is an average net loss of CO$_2$ (550 g m$^{-2}$ yr$^{-1}$) from peat drained for forestry.

During the first year, the sums of N$_2$O and CH$_4$ fluxes calculated as CO$_2$ equivalents (100-year reference period, Solomon et al. 2007) were 525 and 474 g m$^{-2}$ yr$^{-1}$ for the control and ash-fertilized plots at Site 1, respectively, and 36 and 25 g m$^{-2}$ yr$^{-1}$ for the control and ash-fertilized plots at Site 2, respectively. During the second year, the emissions were 863 and 858, 19 and 18, 45 and 48 g m$^{-2}$ yr$^{-1}$ for the control and ash-fertilized plots at Sites 1, 2 and 3, respectively. The high CO$_2$ emission equivalent at Site 1 resulted mainly from the high N$_2$O emissions. At such sites it is not likely that the uptake of CO$_2$-C by biomass could totally compensate for the high greenhouse gas emissions from the peat (as suggested by e.g., Moilanen et al. 2012). However, at Sites 2 and 3 with lower N$_2$O emissions this could be possible. Since the annual net CO$_2$ exchange for these forest ecosystems studied are not known, we cannot calculate the total greenhouse gas balance.

We can conclude that granulated wood-ash application in the three studied peatland forests changed some soil chemical properties, but soil pH did not increase significantly. Granulated wood-ash fertilization altered understory vegetation shortly after fertilization, increased tree growth and decomposition rates but there is no evidence that the GHG emissions increased sig-
significantly. Further studies are needed to explain the decrease in the N$_2$O production caused by ash addition in the laboratory experiments and to differentiate if N$_2$O originates from nitrification or denitrification. Granulated wood-ash fertilization would offer possibilities to reduce N$_2$O emissions and enhance tree stand growth on peatlands. However, to evaluate the overall atmospheric impact, also the effects on organic matter decomposition should be considered.

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References


