

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

Marja Maljanen^{1)*}, Maarit Liimatainen¹⁾, Jyrki Hytönen²⁾ and Pertti J. Martikainen¹⁾

¹⁾ University of Eastern Finland, Department of Environmental Science, P.O. Box 1627, FI-70211 Kuopio, Finland (*corresponding author e-mail: marja.maljanen@uef.fi)

²⁾ Finnish Forest Research Institute, Silmäjärventie 2, FI-69100 Kannus, Finland

Received 16 Apr. 2013, final version received 16 Oct. 2013, accepted 7 Oct. 2013

Maljanen, M., Liimatainen, M., Hytönen, J. & Martikainen, P. J. 2014: The effect of granulated wood-ash fertilization on soil properties and greenhouse gas (GHG) emissions in boreal peatland forests. *Boreal Env. Res.* 19: 295–309.

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, Aronsson 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 (CO₂) and nutrient-rich peat soils can also be sources of nitrous oxide (N₂O), but drained peat soils emit less methane (CH₄) 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 (Väättäinen *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, CO₂ and N₂O 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, N₂O and CH₄ flux rates and soil respiration (CO₂ 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 N₂O 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 m² (Site 1) to 1500 m² (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_2O 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 1		Site 2		Site 3	
	C	A	C	A	C	A
Location	63°54'N, 23°56'E		63°52'N, 23°44'E		63°53'N, 24°26'E	
Dominant tree species	<i>Pinus sylvestris</i> 85% <i>Betula pubescens</i> 15%		<i>Pinus sylvestris</i> 100%		<i>Pinus sylvestris</i> 100%	
Age of trees	52		51		39	
Peatland classification according to Laine <i>et al.</i> (2012)	<i>Vaccinium vitis-idaea</i> type (Ptkg)		<i>Vaccinium vitis-idaea</i> type (Ptkg)		<i>Cladonia</i> type (Jätkg)	
Stand volume ($\text{m}^3 \text{ha}^{-1}$)	296	296	138	110	7.0	17
Increase in stem wood volume in 5 years ($\text{m}^3 \text{ha}^{-1}$)	nd	nd	20	33	3.7	10
pH (H_2O) ^a	3.5 ± 0.2	3.5 ± 0.3	3.8 ± 0.3	4.1 ± 0.5	3.8 ± 0.3	4.1 ± 0.5
EC ($\mu\text{S cm}^{-1}$) ^a	86 ± 18	92 ± 23	35 ± 12	36 ± 14	26 ± 6.6	27 ± 6.9
NO_3^- -N ($\mu\text{g g}^{-1}$) ^a	14 ± 9.4	9.7 ± 4.9	0.9 ± 1.1	1.0 ± 1.7	0.1 ± 0.2	< 0.1
NH_4^+ -N ($\mu\text{g g}^{-1}$) ^a	9.5 ± 10	10 ± 10	7.1 ± 6.3	4.7 ± 4.7	1.6 ± 1.5	1.5 ± 1.4
DOC (mg g^{-1}) ^a	0.6 ± 0.1	0.7 ± 0.3	0.7 ± 0.5	0.5 ± 0.2	0.8 ± 0.4	0.9 ± 0.3
Cl^- ($\mu\text{g g}^{-1}$) ^a	6.2 ± 4.2	8.6 ± 4.6	7.3 ± 3.4	8.4 ± 3.9	13 ± 5.2	12 ± 3.8
SO_4^{2-} ($\mu\text{g g}^{-1}$) ^a	38 ± 24	120 ± 85*	33 ± 13	67 ± 35*	54 ± 59	61 ± 35
Peat depth (cm) ^b	110	98	91	130	> 150	> 150
Bulk density (g cm^{-3}) ^b	0.20	0.18	0.17	0.18	0.05	0.06
Degree of decomposition ^b (H^c)	7–9	7–9	6–8	6–8	1–2	1–2
Organic matter (OM, %) ^b	95	95	91	86	99	98
C (%) ^b	52	51	50	47	46	46
N (%) ^b	2.4	2.5	2.7	2.6	0.56	0.53
C:N ^b	22	21	18	18	82	87
P (mg g^{-1}) ^b	1.1	1.3	1.1	1.2	0.5	0.7
K ($\mu\text{g g}^{-1}$) ^b	300	540	310	490	660	550
B ($\mu\text{g g}^{-1}$) ^b	2.7	5.0	1.5	6.5	0.84	3.9
Ca (mg g^{-1}) ^b	2.4	5.6	1.8	7.0	2.3	7.6
Mg (mg g^{-1}) ^b	0.39	0.69	0.35	1.10	0.74	1.39
Mn ($\mu\text{g g}^{-1}$) ^b	100	270	70	610	130	410
Cd ($\mu\text{g g}^{-1}$) ^b	0.30	0.48	0.23	1.7	0.35	0.59

^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_2SO_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 (5×10 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. The total coverage of plants and the most dominant plant

species were measured inside the collars (60×60 cm) installed for the gas flux measurements.

Gas flux measurements

The N_2O 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_2O 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 $0.389\ \mu l\ l^{-1}$ of N_2O and $1.98\ \mu l\ l^{-1}$ CH_4 was used for daily calibration.

In order to measure 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.

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 N_2O and 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 N_2O and 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 N_2O and 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 N_2O 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. $30 \text{ g}_{(FW)}$ 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 N_2O 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 N_2O concentrations were analyzed with a gas chromatograph as described above. After the last sampling, 2.5% of acetylene (C_2H_2) was added to the headspace of the flasks and the N_2O production was measured again with C_2H_2 inhibition. The soil pH, EC and NO_3^- , 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. $30 \text{ g}_{(FW)}$ 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 N_2O 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

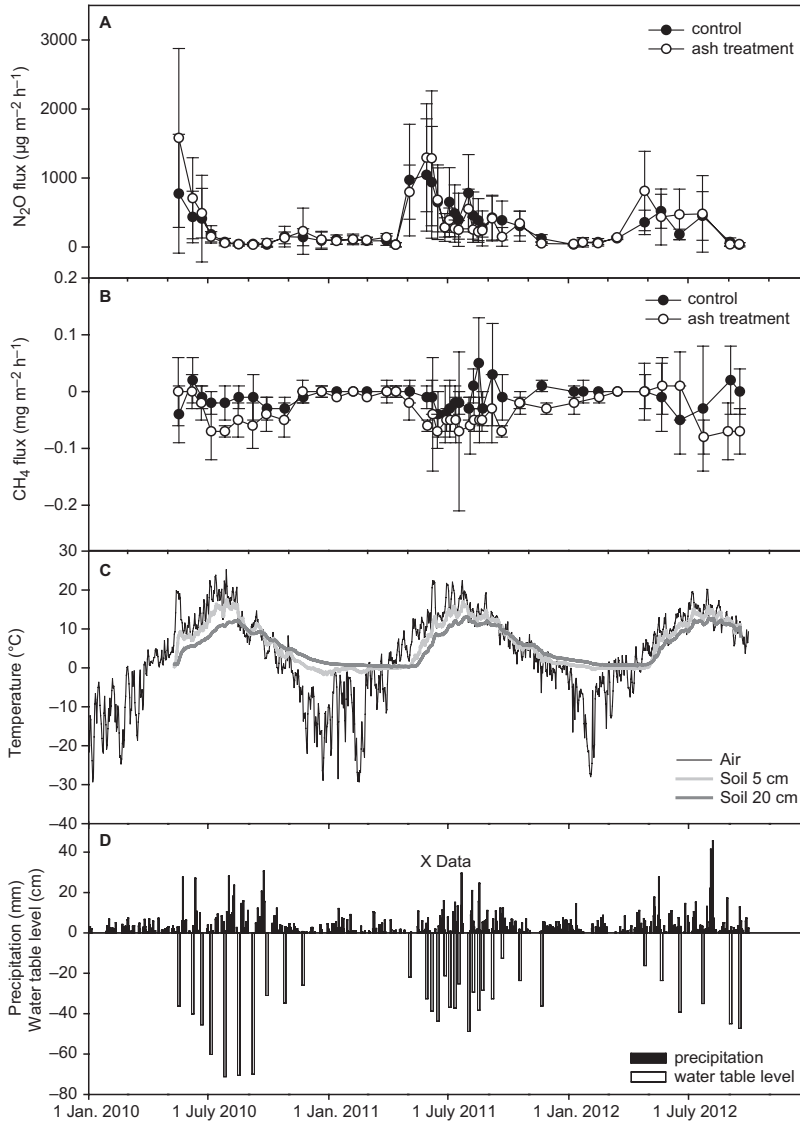


Fig. 1. (A) N_2O and (B) CH_4 flux dynamics at peatland forest Site 1; error bars show standard deviations ($n = 6$). (C) Mean daily temperatures. (D) Daily precipitation and water table level.

study period varied from $-29.2\ ^{\circ}C$ to $25.1\ ^{\circ}C$ (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

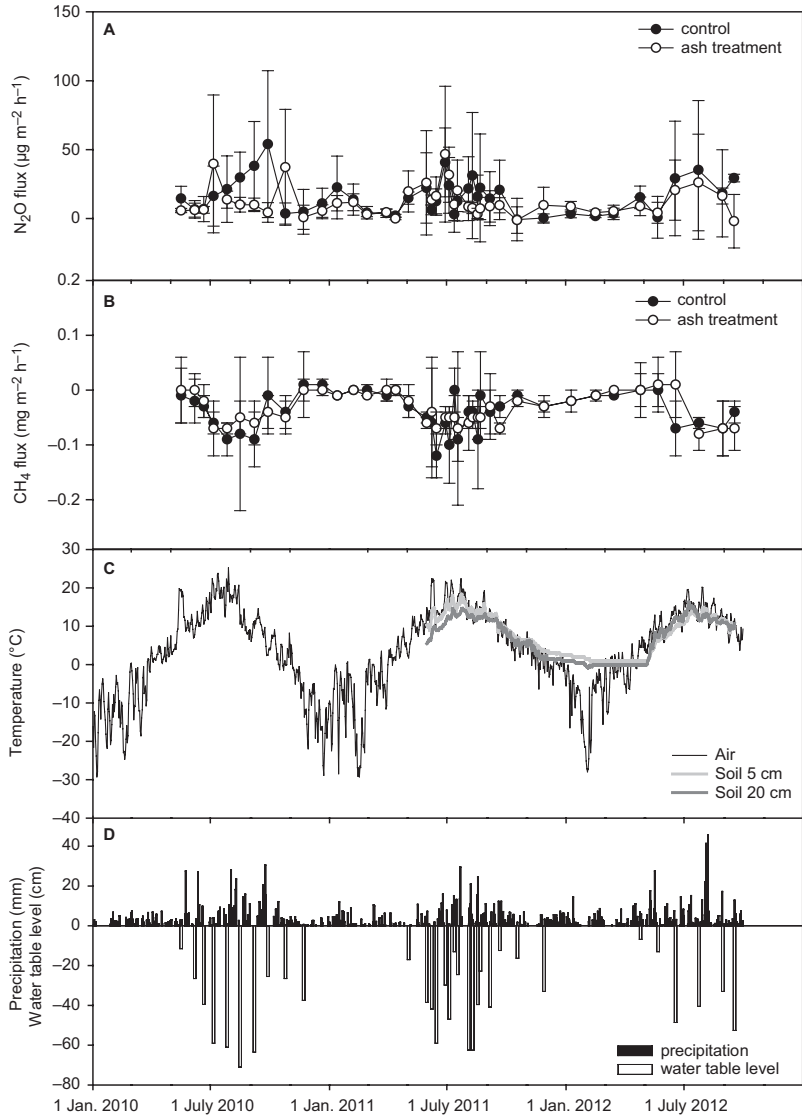


Fig. 2. (A) N₂O and (B) CH₄ flux dynamics at peatland forest Site 2; error bars show standard deviations ($n = 4$). (C) Mean daily temperatures. (D) Daily precipitation and water table level.

here negligible effects on the DOC, NH₄⁺ and NO₃⁻ 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 europæana*, *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

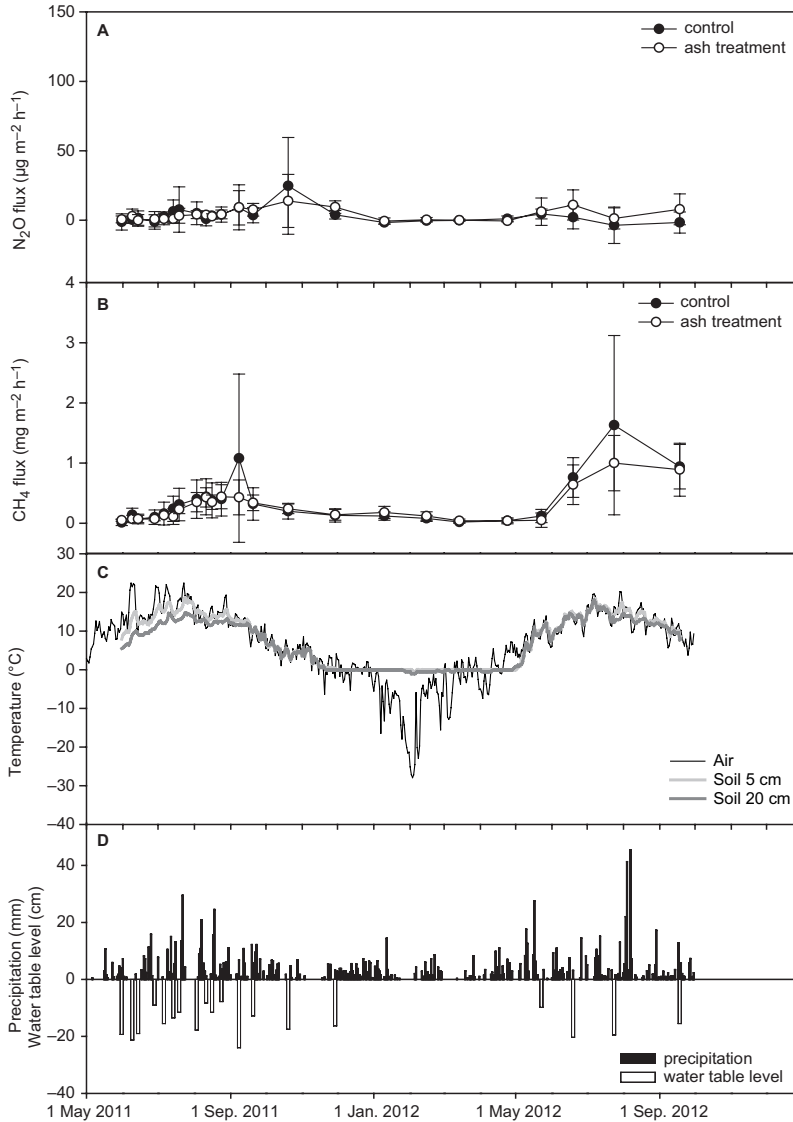


Fig. 3. (A) N_2O and (B) CH_4 flux dynamics at peatland forest Site 3; error bars show standard deviation ($n = 4$). (C) Mean daily temperatures. (D) Daily precipitation and water table level.

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 (FN₂O, FCH₄) and concentrations of N₂O, CH₄ 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.

	Site 1		Site 2		Site 3	
	C	A	C	A	C	A
FN ₂ O (μg m ⁻² h ⁻¹)						
May–Sep. 2010	250 ± 450	390 ± 700	24 ± 28	12 ± 20	nd	nd
May–Sep. 2011	580 ± 530	495 ± 510	19 ± 22	16 ± 21	3.2 ± 7.4	3.1 ± 5.8
Annual (May 2010–May 2011)	1.60 ± 0.69	1.80 ± 0.76	0.14 ± 0.09	0.10 ± 0.07	nd	nd
Annual (May 2011–May 2012)	2.90 ± 1.30	2.90 ± 0.90	0.08 ± 0.07	0.09 ± 0.02	0.03 ± 0.02	0.03 ± 0.01
FCH ₄ (mg m ⁻² h ⁻¹)						
May–Sep. 2010	-0.02 ± 0.03	-0.03 ± 0.03	-0.05 ± 0.07	-0.04 ± 0.04	nd	nd
May–Sep. 2011	-0.01 ± 0.05	-0.02 ± 0.05	-0.05 ± 0.06	-0.05 ± 0.05	0.32 ± 0.47	0.24 ± 0.19
Annual (May 2010–May 2011)	-0.09 ± 0.06	-0.09 ± 0.06	-0.23 ± 0.12	-0.21 ± 0.08	nd	nd
Annual (May 2011–May 2012)	-0.04 ± 0.05	-0.07 ± 0.04	-0.26 ± 0.11	-0.27 ± 0.08	1.65 ± 1.20	1.50 ± 0.50
SR (mg CO ₂ m ⁻² h ⁻¹)	nd	nd	250 ± 110	320 ± 130*	120 ± 50	220 ± 90*
N ₂ O in soil air (ppm)						
May–Sep. 2011	5.0 ± 19	4.9 ± 9.4*	0.58 ± 0.44	0.87 ± 1.1*	0.34 ± 0.22	0.37 ± 0.20*
20 cm	25 ± 60	43 ± 120*	1.0 ± 1.1	4.0 ± 7.6*	0.13 ± 0.09	0.14 ± 0.10
40 cm	41 ± 160	96 ± 240*	9.7 ± 19	11 ± 21	nd	nd
CH ₄ in soil air (ppm)						
May–Sep. 2011	1.9 ± 0.16	1.9 ± 0.13	1.7 ± 0.26	1.5 ± 0.40*	1280 ± 6000	3800 ± 13000*
20 cm	3.0 ± 4.7	2.0 ± 0.44*	8.8 ± 0.49	1.3 ± 1.6*	104000 ± 73000	33700 ± 47000*
40 cm	2.0 ± 0.58	2.2 ± 2.2	16.5 ± 74	3.7 ± 12	nd	nd
Water table level (cm)						
May–Sep. 2010	-49 ± 18	-49 ± 18	-40 ± 19	-44 ± 21	nd	nd
May–Sep. 2011	-33 ± 12	-33 ± 12	-32 ± 18	-36 ± 18	-15 ± 6	-16 ± 6
CD (loss %) May–Sep. 2011						
10 cm	25 ± 17	23.0 ± 6.3	22 ± 2.2	32 ± 6.7*	15 ± 2.8	11 ± 7.1
20 cm	17 ± 9.3	22 ± 9.4	3.3 ± 2.7	7.0 ± 2.8	1.4 ± 1.0	11 ± 7.5*
TD (loss %) May–Sep. 2011						
Green tea, 7 cm	62 ± 4.9	61 ± 4.4	64 ± 5.4	68 ± 8.2	60 ± 3.3	60 ± 3.1
Rooibos tea, 7 cm	44 ± 1.4	45 ± 3.4	45 ± 4.1	47 ± 1.6	42 ± 1.2	44 ± 0.80*
RB, <2 mm (kg ha ⁻¹)						
0–10 cm	1800 ± 410	2300 ± 700*	3600 ± 1200	3100 ± 1400	1700 ± 990	2900 ± 1200*
10–20 cm	550 ± 250	730 ± 270	460 ± 280	510 ± 580*	590 ± 370	700 ± 360*
Understory plant coverage (%)						
in August 2012	56.8 ± 15.1	81.5 ± 8.01*	61.3 ± 21.2	46.5 ± 3.70	65.3 ± 16.0	73.3 ± 11.3

In situ gas flux rates

There were no statistical differences in the N_2O 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_2O emission rates varied largely among the sites and seasons (Figs. 1–3). The N_2O emissions from Site 1 were two magnitudes higher than those from Site 2 (Table 2). Site 3 even occasionally showed N_2O 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_2O emission peak in January when the topsoil was frozen at Sites 1 and 2 (Figs. 1–3). The N_2O 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_2O 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_2O 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_2O concentrations and gas fluxes only at Site 2 where the N_2O concentrations at depths of 20 and 40 cm correlated positively but weakly with the N_2O fluxes (Spearman rank correlation: $r_s = 0.235$, $p = 0.005$; and $r_s = 0.272$, $p = 0.001$; respectively). The N_2O 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_2O 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_2O production rates at temperatures below 0 °C were low but the N_2O 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_2O production 70%–80% at temperatures above 0 °C as compared with that in the control (one-way ANOVA: $F_{1,19} = 1323$, $p < 0.001$ and $F_{1,9} = 68.2$, $p < 0.001$, respectively). Additions of 2.5% acetylene into the headspace inhibited 80%–90% of the N_2O 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_2O production also

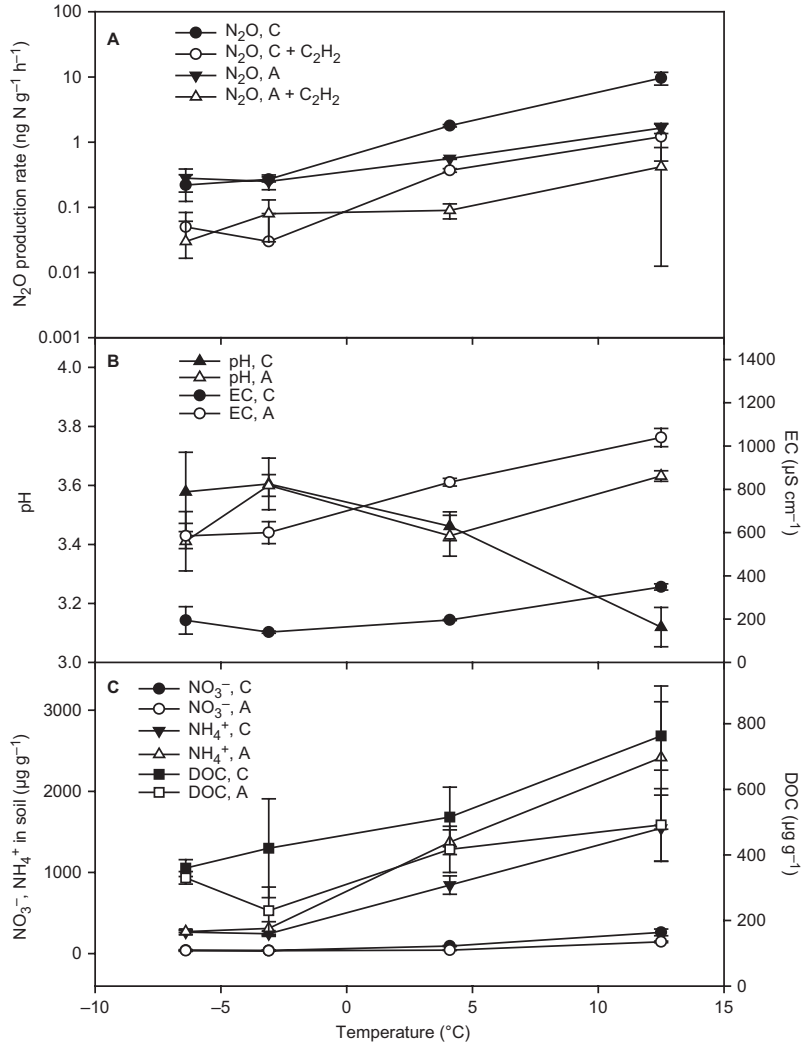


Fig. 4. (A) N₂O production rates after incubation of 10 weeks in laboratory with soil from Site 1 with or without ash addition (1.6 g of ash per 30 g of soil) at different incubation temperatures measured with or without 2.5% acetylene (C₂H₂) inhibition. (B) Soil pH (H₂O) and EC. (C) soil NO₃⁻, NH₄⁺ and DOC contents after incubation. Error bars show standard deviations (*n* = 5).

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 N₂O 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 N₂O emissions, from 0.03 to 2.90 g m⁻². The highest N₂O emis-

sions took place at fertile Site 1 (Ptkg), whereas poor Site 3 (Jätkg) had negligible N₂O emissions (Tables 1 and 2). The annual N₂O 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 N₂O 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 N₂O emission rates (Martikainen *et al.* 1993). Peat humification at the site drained about 50 years ago was higher than at Site 2 drained

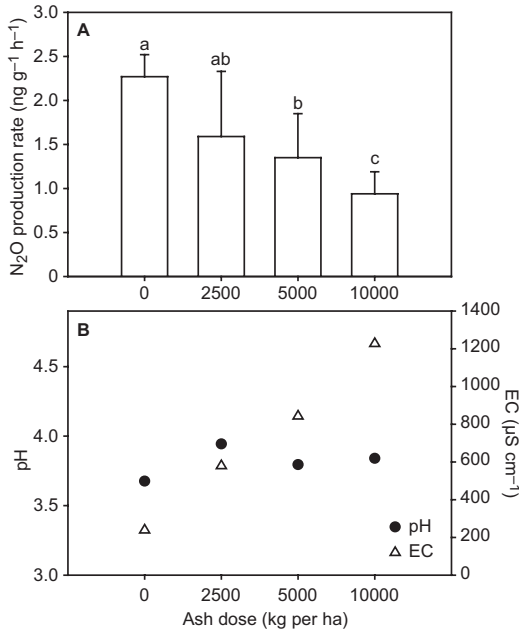


Fig. 5. (A) Mean + SD ($n = 5$) N_2O production rates in laboratory incubation experiment with soil from Site 1 with different ash doses. The differences between the treatments were analyzed by the analysis of variance (one-way ANOVA). The treatments with a common letter do not differ at a statistical significance of $p < 0.05$ in Tukey's test. (B) Soil pH and electrical conductivity (EC).

20 years ago. This could partly explain the higher N_2O emissions from Site 1 (von Arnold *et al.* 2005, Ojanen *et al.* 2010). Klemedtsson *et al.* (2005) suggested that the N_2O 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_2O 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_2O 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_2O 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_2O 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_2O and CH_4 flux rates between the control plots and plots treated with granulated ash. No N_2O emission peaks were detected

during winter as reported earlier (Maljanen *et al.* 2010b, Klemedtsson *et al.* 2010). Also, no significant reduction in the N_2O 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_2O emission peak when soil frost was developing. In the present study, there was only a slight increase in the N_2O 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_2O production rates after the ash application in the laboratory experiments. Odlare and Pell (2009) also reported similar decrease in the N_2O 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_2O 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_2O production decreased after the addition of 2.5% acetylene, a concentration inhibiting both ammonium oxidation and the N_2O reduction to N_2 in denitrification (Klemedtsson *et al.* 1988). Therefore, ammonium oxidizing nitrifiers could play a role in the N_2O 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 $-870 \text{ g m}^{-2} \text{ yr}^{-1}$) 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 ($-70 \pm 30 \text{ g m}^{-2} \text{ yr}^{-1}$) but peat soils at fertile sites are net sources of CO_2 ($190 \pm 70 \text{ g m}^{-2} \text{ yr}^{-1}$). 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 ($550 \text{ g m}^{-2} \text{ yr}^{-1}$) from peat drained for forestry.

During the first year, the sums of N_2O and CH_4 fluxes calculated as CO_2 equivalents (100-year reference period, Solomon *et al.* 2007) were 525 and 474 $\text{g m}^{-2} \text{ yr}^{-1}$ for the control and ash-fertilized plots at Site 1, respectively, and 36 and 25 $\text{g m}^{-2} \text{ yr}^{-1}$ 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 $\text{g m}^{-2} \text{ yr}^{-1}$ 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_2O 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_2O 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-

nificantly. Further studies are needed to explain the decrease in the N_2O production caused by ash addition in the laboratory experiments and to differentiate if N_2O originates from nitrification or denitrification. Granulated wood-ash fertilization would offer possibilities to reduce N_2O 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.

Acknowledgements: Special thanks for assistance in field and laboratory work are due to Seppo Vihanta, Miia Kauppinen, Anni Allosvirta, Katja Vesterinen, Salla Mönkkönen, Matti Pessi and Reetta Kolppanen. The study was funded by the Ministry of Agriculture and Forestry (MYRKLIMA-project), Kone Foundation, North Savo Regional Fund and Finnish Forest Research Institute.

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