Water chemistry following wood-ash application to a Scots pine stand on a drained peatland in Sweden

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Abstract. The availability of phosphorus and potassium often limit growth of trees on well drained peatlands in the boreal region. Wood ash, which contains phosphorus and potassium, can be used for forest fertilization on peatlands or for nutrient compensation following intensive harvesting. This study was performed in order to investigate the effects on water chemistry of applying wood ash to a Pinus sylvestris L. stand on a drained peatland in southern Sweden. Runoff chemistry was monitored in the main ditch for one year before and three years after the application of self-hardened and crushed wood ash, applied at an average rate of 3.1×10³ kg per hectare. In addition, groundwater was collected from the ash-treated peatland and from an adjacent reference peatland and chemically analysed. Shortly after wood-ash application, the concentrations of boron, calcium, potassium, lithium, magnesium, manganese and sulphate in the ditch water, and the electrical conductivity, all increased. Some variables showed elevated concentrations for a few months only, while pH and the concentrations of boron, potassium, lithium, magnesium, manganese, total phosphorus and silica were significantly (p < 0.05) elevated over the last one and a half or two years of the study, compared to the reference period. The concentrations of iron, total sulphur and sulphate were lower during the last one and a half or two study years than in the reference period. Several trace metals and anions did not appear to be affected. In general, the effects detected in groundwater were consistent with those found in ditch water. Groundwater flow through superficial soil layers seems to have facilitated leaching.

Key words: forest, leaching, peat, water, wood ash.

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Introduction

Logging residues, comprising the tops and branches mostly from final fellings, are increasingly being used for energy production. The harvesting of logging residues increases nutrient removal compared to leaving the logging residues on site (Weetman & Webber, 1972; Mälkönen, 1976), thus increasing acidification and possibly decreasing long-term productivity. On peatlands with a substantial peat layer, a significant proportion of essential nutrients, including phosphorus (P) and potassium (K), are present in the trees, in contrast to mineral soils where the main store is in the soil (Egnell et al., 1998). Therefore, the need for nutrient compensation to sustain long-term productivity at increased harvest intensities is probably greater on peatlands than on mineral soils. The nutrients primarily controlling tree growth in boreal forests often dif-
fer between mineral and peat soils. On peat soils, tree growth is often limited by the availability of P and K (Paavilainen & Päivänen, 1995), whereas on mineral soils, tree growth is generally limited by the availability of nitrogen (N) (Tamm, 1991; Binkley & Högberg, 1997). Provided that the drainage and supply of N is sufficient, stem-wood growth on peat soils generally increases after application of P and K, supplied either in the form of commercial fertiliser or as wood ash (Silfverberg & Hartman, 1999; Moilanen et al., 2004; Moilanen et al., 2005a; Moilanen et al., 2005b).

Apart from increasing tree growth, wood-ash application may affect water chemistry (Aronsson & Ekelund, 2004; Augusto et al., 2008). Wood ash contains most elements present in the fuel (Steenari et al., 1999; Augusto et al., 2008). Elevated concentrations of K⁺, calcium (Ca²⁺), magnesium (Mg²⁺), manganese (Mn), sulphate (SO₄²⁻) and chloride (Cl⁻) in runoff have been reported, with minor exceptions, after wood-ash application to peatlands (Nilsson & Lundin, 1996; Tulonen et al., 2002; Piirainen & Domisch, 2004). The impacts on water chemistry in peatlands are probably affected by the moist conditions, possibly facilitating dissolution of the ash, the high content and leaching of organic matter which may affect adsorption and desorption processes, and the ditch network affecting runoff and leaching.

Effects on water chemistry have been sparsely investigated after wood ash application to peatlands, therefore further investigations are required to provide a more solid basis for evaluating the measure. The present study was initiated to meet this need. Water chemistry was monitored for three years following wood-ash application in a small forest catchment in southern Sweden. The main objective was to evaluate whether wood-ash application to a drained peatland would increase the concentrations and leaching of trace metals and nutrients, both of which are potential unwanted side-effects of the measure.

### Material and methods

#### Study site

The study was performed in two small catchments situated 155 m a.s.l. in southern Sweden (57°11’N, 13°44’E), both containing drained peatland as well as areas with mineral soil. The catchments are referred to as Bredaryd South, the reference catchment, and Bredaryd North, the ash-treated catchment (Figure 1). The annual mean temperature in the area is 6.0°C and the annual precipitation 866 mm (Alexandersson & Eggertsson Karlström, 2001). The experiment was designed as a paired-catchment study, but because of significant windthrow during the course of the study, the method of evaluation had to be modified (see below). Bredaryd North was estimated to cover 4.9 to 7.0 hectares, including 3.3 hectares of drained peatland. The range in catchment area reflects the uncertainty in identifying the water divide. This uncertainty was related to the unknown drainage capacity of the most eastern ditch (running from south to north), and to the northern water divide, which was probably located on the peatland between the two main ditches. Therefore, a minimum and a maximum estimate are presented.

The tree stands on both peatlands were predominantly Scots pine (*Pinus sylvestris* L.) with a mean height of 15 m, about 750 stems ha⁻¹ and a stem volume including bark of approximately 130 m³ ha⁻¹. The forests were thinned and ditched in the late 1980s. Ditches about 0.5 to 1 m deep were dug at intervals of 20 to 30 m. The peatlands are ombrotrophic, dominated by *Sphagnum* peat (Ernfors et al., 2010). The depth of the peat layer exceeded one metre, measured at nine locations in each peatland. The field layer contained *Eriophorum vaginatum* L., *Vaccinium vitis-idea* L., *V. myrtillus* L. and *V. uliginósum* L. The bottom layer contained *Pleurozium schreberi* Willd. and *Dicranum* spp. (Ernfors et al., 2010).

In January, 2005, a hurricane caused significant windthrow at Bredaryd South, in
areas with mineral soil. This affected the ditch-water chemistry. Therefore, Bredaryd South could no longer serve as reference for the ditch-water chemistry. However, the Bredaryd South peatland could still be used as reference for the groundwater study, since no significant windthrow occurred on the peatland. The hurricane caused some windthrow at Bredaryd North as well, but the extent and location of the windthrown trees were unlikely to affect the ditch-water chemistry significantly (Figure 1).

Wood-ash application
At Bredaryd North, self-hardened and crushed wood ash was spread using a centrifugal fertiliser spreader pulled by a small tractor. The chemical properties of the ash are presented by Ernfors et al. (2010). The ash was applied on 2.4 ha of the peatland, corresponding to 73% of the peatland area and 34–49% of the entire catchment (Figure 1). A 5-metre zone was left untreated along the main ditch. No measures were taken to avoid spreading over the side ditches. About 25% of the area was treated on 25 Oct., 2004 and the remaining area two days later. The split application was due to technical problems with the tractor. At the time of application, the mean groundwater level was about 9 cm below the soil surface. The ash dose was $3.1 \times 10^3 \pm 2.1 \times 10^3$ (SD) ha$^{-1}$ (dry mass, 105°C for 24h), determined by sampling ash along two transects with 0.25 m$^2$ collection frames ($n = 2$ transects $\times$ 8 positions per transect $\times$ 3 frames per position). The applied dose could be considered either a low fertiliser dose or a high dose to compensating for the increased nutrient removal resulting from whole-tree harvesting.

The ash applied was a mixture of biofuel ash from two thermal power stations in southern Sweden. At one of the stations, the fuel consisted of 75% bark, 20% wood chips and 5% sawdust, and the ash was a mixture of bottom ash (60%) and fly ash (40%). The ash from the other station was fly ash resulting from the burning of wood pellets. Both ashes were mixed with water and left outdoors to harden naturally for up to about six months before subsequent crushing and sieving (10 mm mesh size). The applied load of some elements was calculated based on the element concen-
tration of the ash (Ernfors et al., 2010) and the mean application rate of 3.1x10⁻³ kg ha⁻¹. The applied loads were 37 kg sodium (Na⁺) ha⁻¹, 1.8x10² kg K⁺ ha⁻¹, 74 kg Mg²⁺ ha⁻¹, 6.2x10² kg Ca²⁺ ha⁻¹, 29 kg Mn ha⁻¹, 40 kg P ha⁻¹, 43 kg sulphur (S) ha⁻¹, 3.5x10² kg silica (Si) ha⁻¹, 78 kg Al ha⁻¹, 50 kg iron (Fe) ha⁻¹, 7.4 kg zinc (Zn) ha⁻¹, 1.0 kg arsenic (As) ha⁻¹, 0.043 kg cadmium (Cd) ha⁻¹, < 0.03 kg cobalt (Co) ha⁻¹, 0.23 kg chromium (Cr) ha⁻¹, 0.37 kg copper (Cu) ha⁻¹, 0.024 kg molybdenum (Mo) ha⁻¹, 0.11 kg nickel (Ni) ha⁻¹, 0.42 kg lead (Pb) ha⁻¹ and 0.096 kg vanadium (V) ha⁻¹.

**Water sampling**

At Bredaryd North, ditch-water samples were collected from the main ditch at the catchment outlet (Figure 1). The sampling commenced in late Oct., 2003, and ended in Dec., 2007. The sampling interval was generally one to two weeks. The reference period prior to wood-ash application was one year and comprised 29 samplings. The post-treatment period extended over more than three years and comprised 111 samplings. Data from the first year following application were reported by Sikström et al. (2006) and effects on forest floor greenhouse gas emissions by Ernfors et al. (2010).

Groundwater samples were collected by piezometers installed in the peatlands of Bredaryd North and Bredaryd South. Piezometer nests, and groundwater tubes to measure the groundwater level, were installed at nine positions in each peatland, hereafter referred to as N1 to N9 at Bredaryd North and S1 to S9 at Bredaryd South (Figure 1). Each piezometer nest consisted of three piezometers with the intakes at depths of 30–40 cm, 60–70 cm or 90–100 cm. The nests were located approximately halfway between two adjacent side ditches. The piezometers were made of uncleansed PVC-pipes (φ_inner = 17 mm), sealed at the bottom with a plastic plug, with 65–80 holes (φ = 1.5–2 mm) distributed over a 10 cm interval. The piezometers were emptied and allowed to refill for one to two days before sampling. Groundwater samples were collected before ash application (20 Dec., 2003), two months after application (17 Dec., 2004), and approximately two and three years after application (3 Dec., 2006 and 15 Dec., 2007). For each sampling depth in each catchment, three composite samples were created in the laboratory by mixing equal volumes from three piezometer nests located in lines parallel to the main ditch. At Bredaryd North, composite samples were created from piezometer nests N1, N6 and N7 (corresponding to S7–S9 at Bredaryd South); N2, N5 and N8 (S4–S6 at Bredaryd South) and N3, N4 and N9 (S1–S3 at Bredaryd South) (Figure 1).

**Chemical analysis**

All water samples were collected in 100 ml acid-washed plastic flasks and frozen immediately after sampling. The samples were sent by mail to the laboratory for chemical analyses. The electrical conductivity was measured with a conductivity meter, equipped with a PCM 121 conductivity electrode. The pH was measured potentiometrically at 20 ± 2 °C using a combination pH electrode. Aluminium, As, boron (B), Ca²⁺, Cd, Co, Cr, Cu, Fe, K⁺, Li, Mg²⁺, Mn, Mo, Na⁺, Ni, total P (TP), Pb, total sulphur (TS), selenium (Se), Si, V and Zn were determined using inductively coupled plasma–mass spectrometry. As the water samples from May to Dec., 2007 had to be analysed at two laboratories, the sample volume was not sufficient for all analyses. Therefore, data on the concentrations of Al, Ca²⁺, Cr, Fe, K⁺, Li, Mg²⁺, Mn, Na⁺, TS, Se, Si, Zn and TP are missing for this period, both for ditch water and groundwater. Fluoride (F⁻), Cl⁻, nitrite-N (NO₂⁻-N), NO₃⁻-N, bromide (Br⁻), phosphate-P (PO₄³⁻-P) and SO₄²⁻-S were determined by anion chromatography. Ammonium (NH₄⁺-N) was determined by flow injection analysis (FIA). Total N (TN) was also determined by FIA, after conversion to NO₃⁻ using a peroxodisulphate alkaline oxidising reagent under pressurised boiling. Organic N was calculated as...
TN less the sum of NH$_4^+$-N, NO$_2^-$-N and NO$_3^-$-N. Since a major portion of the concentrations of NO$_2^-$-N and NO$_3^-$-N were below the chemical detection limit and were set equal to this limit, the calculated concentrations of organic N were underestimated. Total organic C (TOC) was determined with a TOC analyser, as the difference between total C (TC) and inorganic C (IC). When calculating charge concentrations, the charge was assumed to be +2 for Al, Fe and Mn and the H$^+$ concentration was assumed to equal 10$^{-pH}$. The total positive charge was calculated as $\sum$(Na$^+$+K$^+$+Mg$^{2+}$+Ca$^{2+}$+Al$^{3+}$+Fe$^{3+}$+Mn$^{2+}$+NH$_4^+$+H$^+$) and the total negative charge as $\sum$(Cl$^-$+SO$_4^{2-}$+PO$_4^{3-}$+F$^-$+OH$^-$). For Br$^-$, NO$_2^-$-N, NO$_3^-$-N, PO$_4^{3-}$ and F$^-$, more than 70% of the concentrations were below the detection limit. Only PO$_4^{3-}$ and F$^-$ were included in the total negative charge.

**Leaching rate**

The leaching rate of different elements from Bredaryd North was calculated by multiplying the daily concentration in the main ditch by the calculated daily discharge. The daily concentration was linearly interpolated between samplings and the daily discharge was calculated using an application of the HBV model. The HBV model is a semi-distributed, conceptual runoff model, originally developed for hydrological forecasting (Bergström, 1976; Lindstrom et al., 1997). One application of the model is in calculating discharge from locations lacking recorded discharge data (Johansson, 1992). At larger spatial scales, it has been found that the large temporal and spatial variations in discharge can be better captured by discharge modelling, using recorded values of precipitation and temperature, than by extrapolation of recorded discharge from neighbouring stations (B. Johansson, Swedish Meteorological and Hydrological Institute, pers. comm., 2011). This is because that the network for monitoring precipitation is denser than that for discharge in Sweden. The HBV model is applied for several discharge stations in a region, and calibrated to find a set of model coefficients which are representative for all stations within the region. These parameters are then used for simulating the discharge in ungagged catchments, with the assumption that the model error at a single ungagged point is of the same magnitude as at the recording discharge stations in the region, for which model results can be compared with recorded discharge.

The simulations for Bredaryd North were made using the same parameter values used for the calculations of annual discharge in the rivers in the western Jönköping and southern Halland counties, Sweden. Daily precipitation was measured about 5 km from the Bredaryd site using a rain gauge from the Swedish Meteorological and Hydrological Institute (SMHI) mounted 1.5 m above the ground. Temperature data was from the SMHI station at Hagshult (Alexandersson & Eggertsson Karlstrom, 2001). Monthly mean values for potential evapotranspiration from Borås and Hagshult (Eriksson, 1981) were used to calculate actual evapotranspiration. The discharge rates were calculated for 1 October 2003 to 31 December 2007. Given the uncertainty in the identification of the water divide, the daily discharge rates, and hence the leaching rates, were calculated for an estimated minimum and maximum size of the catchment, i.e. 4.9 ha and 7.0 ha respectively.

**Statistical analysis**

The chemistry of the ditch–water samples from Bredaryd North, i.e. electrical conductivity, pH and element concentrations, was statistically analysed to identify short-term and long-term effects of ash application. First, a randomisation approach was used to determine possible long-term effects. For the variables without a long-term effect, but with a clear, graphically indicated, short-term effect, a non-parametric smoothing technique was used to determine the approximate duration of the short-
term effect. Locally weighted linear regres-
sion smoothers were used (PROC GAM in
SAS® software).

Randomised intervention analysis (RIA)
was used to identify long-term effects
(Carpenter et al., 1989). In RIA, the com-
pared differences between medians are
compared to differences between medi-
ans from the data series, resampled ran-
domly from the original data series. The
error distributions of the differences are
derived from these resampled series. In the
current approach, a resampling technique
was used that accounts for seasonal varia-
tion (for details see Löfgren et al., 2009).
RIA was also used to test whether the level
of discharge during the post-treatment period
was similar to the level during the refer-
ce period. Since daily values were mod-
elled for discharge, block bootstrapping
was used in order to retain the serial corre-
lation in the resampled data. This approach
is described by Sørensen et al. (2009).

Shortly after the wood-ash application,
several chemical variables in the ditch
water showed a distinct, graphically indi-
cated, short-term effect that lasted a few
months. Hence, to determine the long-term
effects, the data from the first year after ash
application were omitted. Then the median
concentration prior to application was com-
pared with the corresponding median for
the data from the second and third post-
treatment years. Variables with more than
16% of the values below the chemical detec-
tion limit were not analysed by this method.
Concentrations equal to or below the detection
limit were set to be equal to the detection
limit. This is not likely to have affected
the results, since RIA was based on differ-
ces between medians and the analysis
was not conducted for variables with more
than 16% of the concentrations below the
detection limit.

The duration of the short-term effects,
that were graphically indicated, was esti-
mated for the variables which the RIA indi-
cated had no long-term effects; it was there-
fore assumed that the effects lasted less
than one year. First, the data series for each
measured chemical variable was individ-
ually smoothed using a locally weighted
linear regression smoother (LOESS;
Cleveland, 1979) to smooth out day-to-day
variation. The same smoothing parameter
was used for all data series. The smoothed
curve for the reference year and the sec-
ond and third year after ash application
was used to establish the maximum level
of the normal variation in concentration.
The duration of the short-term effects was
estimated as the period during which the
smoothed curve for the first post-treatment
year exceeded the maximum level of nor-
mal variation. Substituting concentrations
below the detection limit with the value of
this limit has a clear effect on the estimate.
In this study, however, the smoother was
used only to determine the length of the
short-term increase in concentration, the
magnitude of which was far above the level
of the detection limit. The non-parametric
smoothing technique was performed for
electrical conductivity and the concentra-
tions of Al, Ca²⁺, Na⁺, Pb, TOC, V and total
positive and negative charges.

Linear correlations between different
chemical variables were calculated by means
of the Pearson product-moment correlation
using the CORR procedure in SAS ver. 9.2
(http://support.sas.com). Concentrations
below or equal to the detection limit were
set to be equal to the detection limit. In RIA
and correlation analyses, statistical signifi-
cance was defined as \( p < 0.05 \)

Effects on the groundwater chemistry
were evaluated, in graphical form, by eye.
In the groundwater samples, all observa-
tions of Al, As, Ca²⁺, Cl⁻, Cr, Cu, Fe, K⁺, Li,
Mg²⁺, Na⁺, Ni, Pb, Si, TN, V, Zn, TC and TOC
were above the limit of detection, while for
NH₄⁺-N and Mn one observation for each
variable was below the limit. For B, Cd, Co,
TP, TS, Se and SO₄²⁻-S, 10–25% of the obser-
vations were below the limit of detection
and for Mo and IC about 30–40%. Nearly all
observations of Br⁻, F⁻, NO₃⁻-N, NO₂⁻-N and
PO₄³⁻-P were below the limit of detection.
Results

Precipitation and runoff
During the reference year, the annual precipitation in the area was 29% higher than normal (Table 1, cf. Alexandersson & Eggertsson Karlström, 2001). In the first and second year following ash application, the annual precipitation was close to normal (+7% and -3% respectively), while, in the third year, the annual precipitation was 71% higher than normal. Monthly precipitation varied between 15 mm and 215 mm during the study. About 80 mm of precipitation fell during the first month following wood-ash application.

The calculated annual runoff varied between 290 and 610 mm, with the highest runoff in the third year after treatment (Table 1). Monthly runoff varied between 7 mm and 95 mm, with 49 mm during the first month following wood-ash application. According to RIA, the calculated daily discharge showed no change in level during the post-treatment period compared to the reference period (p = 0.6).

Ditch-water chemistry
In the ditch-water samples, the concentrations of Al, As, B, Ca^{2+}, Cl, Co, Cr, Fe, Li, Mg^{2+}, Mn, Na^+, Pb, V, Zn, TN, TC and TOC were all above the detection limit. Less than 3% of the concentrations of Cd, K^+, Ni, Si and SO_{4}^{2-} were equal to or below the detection limit. The corresponding figures were 5-10% for Cu, NH_{4}^{+}-N and TP, 15-16% for TS and Se, 29% for Mo and 70% or more for Br, F, IC, NO_{3}^{-}-N, NO_{2}^{-}-N and PO_{4}^{3-}-P.

During the reference year, H^+ and Na^+ accounted for, on average, 40% and 31%, respectively, of the total positive charge (\Sigma (Na^{+}+K^{+}+Mg^{2+}+Ca^{2+}+Al^{3+}+Fe^{2+}+Mn^{2+}+NH_{4}^{+}+H^{+})), whereas Cl^- accounted for approximately 71% of the total negative charge (\Sigma (Cl^{-}+SO_{4}^{2-}+PO_{4}^{3-}+F+OH^-)). The positive charge during the reference year was, on average, 2.4 times the negative charge. Organic N was the dominant N species. On average, SO_{4}^{2-} accounted for 34% of TS. The median TOC concentration was 75 mg l^{-1} and the median pH 3.7 (Table 2).

Effects on ditch-water chemistry were apparent when the data was presented graphically, within one to two weeks of

<table>
<thead>
<tr>
<th>Variable</th>
<th>Reference year</th>
<th>Year 1</th>
<th>Year 2</th>
<th>Year 3</th>
</tr>
</thead>
<tbody>
<tr>
<td>Precipitation (mm)</td>
<td>1000</td>
<td>770</td>
<td>760</td>
<td>1100</td>
</tr>
<tr>
<td>Runoff (mm)</td>
<td>400</td>
<td>360</td>
<td>290</td>
<td>610</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Variable</th>
<th>Leaching rate (kg yr^{-1})</th>
</tr>
</thead>
<tbody>
<tr>
<td>H^+</td>
<td>4.7 ± 0.84</td>
</tr>
<tr>
<td>B</td>
<td>0.077 ± 0.014</td>
</tr>
<tr>
<td>Fe</td>
<td>8.9 ± 1.6</td>
</tr>
<tr>
<td>K^+</td>
<td>8.7 ± 1.5</td>
</tr>
<tr>
<td>Li</td>
<td>0.0082 ± 0.0015</td>
</tr>
<tr>
<td>Mg^{2+}</td>
<td>15 ± 2.8</td>
</tr>
<tr>
<td>Mn</td>
<td>0.16 ± 0.028</td>
</tr>
<tr>
<td>TP</td>
<td>1.2 ± 0.21</td>
</tr>
<tr>
<td>SO_{4}^{2-}-S</td>
<td>13 ± 2.3</td>
</tr>
<tr>
<td>TS</td>
<td>38 ± 6.8</td>
</tr>
<tr>
<td>Si</td>
<td>7.8 ± 1.4</td>
</tr>
</tbody>
</table>

ash application. The concentration of total positive and total negative charges then increased sharply (Figure 2), as did the electrical conductivity. The total positive charge and the electrical conductivity remained elevated for about three months, and the total negative charge was elevated for nearly six months according to the non-parametric smoothing technique. The initial increase in positive charge resulted mainly from elevations in the concentrations of Ca\textsuperscript{2+}, K\textsuperscript{+}, Mg\textsuperscript{2+} and possibly Na\textsuperscript{+} and H\textsuperscript{+} (Figures 3–4). Elevated concentrations of B, Li, Mn and Zn were also indicated by the graphs (Figures 5–6). The short-term increase in Ca\textsuperscript{2+} concentration lasted three months, according to the non-parametric smoothing technique, whereas pH and the concentrations of B, K\textsuperscript{+}, Li, Mg\textsuperscript{2+}, Mn and Si showed a long-term increase accord-

Table 2. Approximate p-values according to RIA, based on differences in median concentrations, pH or electrical conductivity in ditch water before and after treatment, and median concentrations in ditch water during the reference year, the first year and the 2\textsuperscript{nd} and 3\textsuperscript{rd} years following ash application. na: not analysed, bd: many observations below detection limit.

<table>
<thead>
<tr>
<th>Variable</th>
<th>p-value</th>
<th>Increase (+) or decrease (-) in concentration or pH</th>
<th>Unit</th>
<th>Medians in ditch water</th>
<th>Reference year</th>
<th>Year 1</th>
<th>Years 2 and 3</th>
</tr>
</thead>
<tbody>
<tr>
<td>Σcations\textsuperscript{a}</td>
<td>0.26</td>
<td></td>
<td>mmol l\textsuperscript{-1}</td>
<td>0.45</td>
<td>0.60</td>
<td>0.41\textsuperscript{b}</td>
<td></td>
</tr>
<tr>
<td>Σanions\textsuperscript{c}</td>
<td>0.16</td>
<td></td>
<td>mmol l\textsuperscript{-1}</td>
<td>0.19</td>
<td>0.39</td>
<td>0.24</td>
<td></td>
</tr>
<tr>
<td>pH</td>
<td>0.000</td>
<td>(+)</td>
<td>mmol l\textsuperscript{-1}</td>
<td>3.7</td>
<td>3.7</td>
<td>4.4</td>
<td></td>
</tr>
<tr>
<td>Electrical conductivity</td>
<td>0.096</td>
<td>µS cm\textsuperscript{-1}</td>
<td>99</td>
<td>1.2×10\textsuperscript{2}</td>
<td>95</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Al</td>
<td>0.50</td>
<td></td>
<td>mg l\textsuperscript{-1}</td>
<td>0.17</td>
<td>0.14</td>
<td>0.18\textsuperscript{b}</td>
<td></td>
</tr>
<tr>
<td>As</td>
<td>0.33</td>
<td></td>
<td>µg l\textsuperscript{-1}</td>
<td>1.0</td>
<td>0.9</td>
<td>1.0</td>
<td></td>
</tr>
<tr>
<td>B</td>
<td>&lt; 0.001</td>
<td>(+)</td>
<td>µg l\textsuperscript{-1}</td>
<td>2.9</td>
<td>13</td>
<td>12</td>
<td></td>
</tr>
<tr>
<td>Ca\textsuperscript{2+}</td>
<td>0.57</td>
<td></td>
<td>mmol l\textsuperscript{-1}</td>
<td>0.031</td>
<td>0.053</td>
<td>0.033\textsuperscript{b}</td>
<td></td>
</tr>
<tr>
<td>Cd\textsuperscript{d}</td>
<td>na</td>
<td></td>
<td>µg l\textsuperscript{-1}</td>
<td>0.1</td>
<td>0.1</td>
<td>0.1</td>
<td></td>
</tr>
<tr>
<td>Cl\textsuperscript{−}</td>
<td>0.058</td>
<td></td>
<td>mmol l\textsuperscript{-1}</td>
<td>0.15</td>
<td>0.22</td>
<td>0.18</td>
<td></td>
</tr>
<tr>
<td>Co</td>
<td>0.15</td>
<td></td>
<td>µg l\textsuperscript{-1}</td>
<td>0.2</td>
<td>0.2</td>
<td>0.2</td>
<td></td>
</tr>
<tr>
<td>Cr</td>
<td>0.57</td>
<td></td>
<td>µg l\textsuperscript{-1}</td>
<td>1.0</td>
<td>1.0</td>
<td>0.9\textsuperscript{b}</td>
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</tr>
<tr>
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<td></td>
<td>µg l\textsuperscript{-1}</td>
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<td>0.8</td>
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<td></td>
</tr>
<tr>
<td>Fe</td>
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<td>(−)</td>
<td>mg l\textsuperscript{-1}</td>
<td>0.35</td>
<td>0.31</td>
<td>0.20\textsuperscript{b}</td>
<td></td>
</tr>
<tr>
<td>K\textsuperscript{+}</td>
<td>&lt; 0.001</td>
<td>(+)</td>
<td>mmol l\textsuperscript{-1}</td>
<td>0.0053</td>
<td>0.073</td>
<td>0.046\textsuperscript{b}</td>
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</tr>
<tr>
<td>Li</td>
<td>&lt; 0.001</td>
<td>(+)</td>
<td>µg l\textsuperscript{-1}</td>
<td>0.3</td>
<td>1.2</td>
<td>1.0\textsuperscript{b}</td>
<td></td>
</tr>
<tr>
<td>Mg\textsuperscript{2+}</td>
<td>0.038</td>
<td>(+)</td>
<td>mmol l\textsuperscript{-1}</td>
<td>0.048</td>
<td>0.071</td>
<td>0.059\textsuperscript{b}</td>
<td></td>
</tr>
<tr>
<td>Mn</td>
<td>&lt; 0.001</td>
<td>(+)</td>
<td>µg l\textsuperscript{-1}</td>
<td>6.1</td>
<td>29</td>
<td>20\textsuperscript{b}</td>
<td></td>
</tr>
<tr>
<td>Mo</td>
<td>na</td>
<td></td>
<td>µg l\textsuperscript{-1}</td>
<td>0.1</td>
<td>0.2</td>
<td>0.1</td>
<td></td>
</tr>
<tr>
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<td>0.38</td>
<td></td>
<td>mmol l\textsuperscript{-1}</td>
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<td>0.15</td>
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<tr>
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<td></td>
<td>µg l\textsuperscript{-1}</td>
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<td>0.40</td>
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<td></td>
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<tr>
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<td></td>
<td>µg l\textsuperscript{-1}</td>
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<td>5.4</td>
<td></td>
</tr>
<tr>
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<td>na</td>
<td></td>
<td>µg l\textsuperscript{-1}</td>
<td>0.3</td>
<td>0.3</td>
<td>0.3</td>
<td></td>
</tr>
<tr>
<td>Si</td>
<td>0.020</td>
<td>(+)</td>
<td>mg l\textsuperscript{-1}</td>
<td>0.26</td>
<td>0.24</td>
<td>0.42\textsuperscript{b}</td>
<td></td>
</tr>
<tr>
<td>V</td>
<td>0.38</td>
<td></td>
<td>µg l\textsuperscript{-1}</td>
<td>1.1</td>
<td>0.80</td>
<td>1.0</td>
<td></td>
</tr>
<tr>
<td>Zn</td>
<td>0.72</td>
<td></td>
<td>µg l\textsuperscript{-1}</td>
<td>11</td>
<td>16</td>
<td>11\textsuperscript{b}</td>
<td></td>
</tr>
<tr>
<td>TP</td>
<td>0.030</td>
<td>(+)</td>
<td>µg l\textsuperscript{-1}</td>
<td>35</td>
<td>95</td>
<td>91\textsuperscript{b}</td>
<td></td>
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<tr>
<td>SO\textsubscript{4}\textsuperscript{2−}</td>
<td>0.030</td>
<td>(−)</td>
<td>mmol l\textsuperscript{-1}</td>
<td>0.029</td>
<td>0.11</td>
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</tr>
<tr>
<td>TS</td>
<td>0.012</td>
<td>(−)</td>
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<td>1.5</td>
<td>2.4</td>
<td>0.69\textsuperscript{b}</td>
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<td>NH\textsubscript{4}\textsuperscript{−}</td>
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<td>0.1</td>
<td>0.23</td>
<td>0.19</td>
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<tr>
<td>TN</td>
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<td>1.5</td>
<td>1.8</td>
<td>1.6</td>
<td></td>
</tr>
<tr>
<td>Org N</td>
<td>na</td>
<td></td>
<td>mg l\textsuperscript{-1}</td>
<td>1.2</td>
<td>1.1</td>
<td>1.2</td>
<td></td>
</tr>
<tr>
<td>TOC</td>
<td>0.93</td>
<td></td>
<td>mg l\textsuperscript{-1}</td>
<td>57</td>
<td>57</td>
<td>75</td>
<td></td>
</tr>
</tbody>
</table>

\textsuperscript{a} Σ(Na\textsuperscript{+}+K\textsuperscript{+}+Mg\textsuperscript{2+}+Ca\textsuperscript{2+}+Al\textsuperscript{3+}+Fe\textsuperscript{2+}+Mn\textsuperscript{2+}+NH\textsubscript{4}\textsuperscript{+}+H\textsuperscript{+}),

\textsuperscript{b} Data were available for the 2\textsuperscript{nd} year and until early May of year 3,

\textsuperscript{c} Σ(Cl\textsuperscript{−}+SO\textsubscript{4}\textsuperscript{2−}+PO\textsubscript{4}\textsuperscript{3−}+F\textsuperscript{−}+OH\textsuperscript{−}),

\textsuperscript{d} RIA failed probably because of too little variation in concentrations.
Figure 2. Total positive charge (solid line) and total negative charge (dashed line) concentrations of ditch water from Bredaryd North. The vertical dashed line indicates the date of ash application.

Figure 3. Concentrations of Na\(^+\) (open circles), K\(^+\) (filled circles), Mg\(^{2+}\) (solid line) and Ca\(^{2+}\) (dashed line) in ditch water. The vertical dashed line indicates the date of ash application.

Figure 4. pH of ditch water (filled circles) and calculated daily discharge, according to an application of the HBV model (solid line), assuming a catchment area of 7.0 ha. The vertical dashed line indicates the date of ash application.
Figure 5. Concentrations of B (filled circles) and Li (dashed line) in ditch water. The vertical dashed line indicates the date of ash application.

Figure 6. Concentrations of Mn (filled circles) and Zn (dashed line) in ditch water. On one occasion in 2003, the Zn concentration was 200 µg l⁻¹. The vertical dashed line indicates the date of ash application.

Figure 7. Concentrations of TOC (dashed line) and Pb (filled circles) in ditch water. The vertical dashed line indicates the date of ash application.
These elements and pH were therefore not analysed for short-term effects. The annual leaching rates of B, K, Li and Mn appeared elevated in all three study years following ash application (Table 1). There were no significant long-term effects of ash application on the concentrations of Al, As, Ca\(^{2+}\), Co, Cr, Mo, Na\(^+\), Ni, Se, V and Zn (Table 2). According to RIA, the Fe concentration decreased, and an increase and a decrease was indicated for Pb and Cu, respectively (Table 2). Several metals, notably Al, As, Fe, Pb, and V, were correlated with the TOC concentration (cf. Figure 7); the Pearson’s correlation coefficient was 0.83 for Al, 0.67 for As, 0.65 for Fe, 0.89 for Pb, and 0.79 for V \(p < 0.0001\) for the post-treatment data. During the reference period, pH was negatively correlated with the TOC concentration (Pearson correlation coefficient -0.75, \(p < 0.0001\)), but after ash application, no such correlation was detected \(p = 0.53\).

The initial increase in negative charge was caused mostly by an elevation in the SO\(_4^{2-}\)-S concentration (Figure 8). The peak in TS following ash application was almost entirely made up by SO\(_4^{2-}\)-S. In the long term, the concentrations of TS and SO\(_4^{2-}\)-S were lowered compared to the levels during the reference year according to RIA, and an increase was indicated for Cl\(^-\) (Table 2). Most of the Br\(^-\) and F\(^-\) concentrations were \(\leq 0.2\) mg l\(^{-1}\), but both variables displayed slightly higher concentrations during one period following ash application. The Br\(^-\) concentration was higher for about six months during the first post-treatment year reaching at most 0.56 mg l\(^{-1}\). The F\(^-\) concentration exceeded 0.2 mg l\(^{-1}\) for about a year during the latter half of 2006 and the first part of 2007, the highest concentration recorded was 0.71 mg l\(^{-1}\).

The concentrations of NH\(_4^+\)-N, TP and PO\(_4^{3-}\)-P displayed a number of peaks following ash application, which sometimes occurred simultaneously (Figures 9–10). The IC concentration showed a similar temporal pattern, but peaks of the same magnitude occurred during the reference year as well. Organic N was generally the dominant N species after ash application, but the peaks in NH\(_4^+\)-N were of the same magnitude as that for organic N (Figure 9). There was no significant long-term effect on the concentration of NH\(_4^+\)-N or TN (Table 2). The NO\(_3^-\)-N and NO\(_2^-\)-N concentrations were \(\leq 0.11\) mg l\(^{-1}\) and \(\leq 0.26\) mg l\(^{-1}\), respectively, throughout the study. The peaks in TP concentration were largely made up of PO\(_4^{3-}\)-P (Figure 10). There was a long-term increase in the TP concentration (Table 2) and the leaching rate of TP appeared elevated in all three study years following ash application (Table 1).

Figure 8. Concentrations of Cl\(^-\) (filled circles) and SO\(_4^{2-}\)-S (solid line) in ditch water. The vertical dashed line indicates the date of ash application.
Groundwater chemistry
The mean groundwater level (gwl) varied between depths of 3 and 57 cm at Bredaryd North (see also Ernfors et al. (2010)). The mean level at Bredaryd South exhibited the same temporal pattern as at Bredaryd North (gwlSouth = 0.998 × gwlNorth − 4.28, R² = 0.97, gwl expressed as a negative value in cm). At Bredaryd North, the mean groundwater level (SD) at groundwater sampling was 15 (7.5) cm in 2003, 11 (3.3) cm in 2004 (measured two days before sampling), 8.7 (6.3) cm in 2006 and 11 (5.3) cm in 2007.

Before ash application, the chemical variables studied generally had similar concentrations in the groundwater collected from the two peatlands. Following ash application, four typical response patterns were distinguished. 1) Some variables appeared unaffected by the ash application, including the concentrations of Al, As, Cu, Fe, NH₄⁺-N, Pb, Se, Si, TN, V, Zn and possibly Ni. Furthermore, nearly all concentrations of Br⁻, NO₂⁻-N, NO₃⁻-N, F⁻ and PO₄³⁻-P were below the detection limit, indicating that these variables were not significantly affected by the ash application. 2) Some variables exhibited an increase in concentration two months after ash application, but not two or three years later. The concentrations of Ca²⁺, Cl⁻, Co, Mg²⁺, Mn, Na⁺, SO₄²⁻, TS, TP and possibly Cd dis-
played such a pattern (Table 3), while the TOC concentration tended to decrease at a depth of 30–40 cm. For Cd, an elevation was also suggested in two samples collected at a depth of 60–70 cm three years after ash application. 3) The Mo, and possibly Cr, concentration at a depth of 30–40 cm tended to be elevated in the samples collected two years after ash application, but not after two months or three years (data available only for Mo). 4) Boron, K⁺ and Li showed an increase in concentration both two months and more than two years (and three years for B) after ash application (Table 3). Data on K⁺ and Li concentrations were not available for the third year after application.

Data on groundwater pH, available mainly for the second and the third year, showed no effect of wood ash application.

When treatment effects were indicated, the highest concentrations were generally observed in the samples collected at a depth of 30–40 cm. For some elements, especially B, K⁺ and Li, the concentration in groundwater at a depth of 30–40 cm was similar to the concentration in ditch water on all sampling occasions following ash application (Table 3).

High concentrations of Cu, Cr, Ni and Zn were found in a few groundwater samples collected in 2004. These concentrations were one to two orders of magnitude higher than the concentrations in 2003 and 2006. The high concentrations of all four elements were found in the same samples, which originated from both Bredaryd North and Bredaryd South.

Table 3. Mean concentrations of B (µg l⁻¹), Ca (µmol l⁻¹), K (µmol l⁻¹) and TP (µg l⁻¹) in groundwater samples collected at depths of 30–40 cm, 60–70 cm and 90–100 cm at Bredaryd South and North, and concentration in ditch water (Ditch). All groundwater samples were taken in December of each year and the ditch water was sampled simultaneously or within two days. For 2007, however, an average of concentration in the ditch-water samples taken on 9 and 23 Dec. is presented. The means were calculated using the concentrations in three composite samples taken per soil depth and peatland. Standard deviations are shown within brackets.

<table>
<thead>
<tr>
<th>Sampling depth (cm)</th>
<th>2003a</th>
<th>2004b</th>
<th>2006b</th>
<th>2007b</th>
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<tr>
<td></td>
<td>South</td>
<td>North</td>
<td>South</td>
<td>North</td>
</tr>
<tr>
<td>B</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>30–40</td>
<td>0.57</td>
<td>1.8</td>
<td>1.5</td>
<td>14</td>
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<tr>
<td>60–70</td>
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<td>0.67</td>
<td>0.87</td>
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<td>0.3</td>
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<td>Ca</td>
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<td>2.5</td>
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<td>3.8</td>
<td>7.7</td>
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<td>4.8</td>
</tr>
<tr>
<td>Ditch</td>
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<td>1.4×10²</td>
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<tr>
<td>Ditch</td>
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Table. Before ash application
b After ash application
Discussion

Excess leaching of trace metals and nutrients is a possible adverse effect of ash application, affecting downstream ecosystems and reducing the efficacy of nutrient application. At Bredaryd North, the ditch-water chemistry changed distinctly following ash application. The chemical variables that were affected in ditch water generally responded in a similar way in groundwater, especially in the short term but for many variables also over the longer term. Several factors could have contributed to the distinct initial response. A large proportion of the peatland was treated with ash and this area was located immediately upstream of the sampling point. Ash was spread over the side ditches and the groundwater table was shallow at the time of application, which may have facilitated leaching.

The concentrations of several trace metals in ditch water and groundwater were not significantly affected by the ash application, supporting the findings of Nieminen et al. (2005). The high concentrations of Cu, Cr, Ni and Zn that were found in a few groundwater samples probably resulted from contamination, since these samples were collected from both the reference and the ash-treated peatlands. There was an indication of elevated Cd and Co concentrations in some of the groundwater samples. In ditch water, the concentrations of As, Al, Fe, Pb and V were correlated with the TOC concentration, and the correlation coefficients indicated that significant proportions of these elements were bound to organic compounds. Elements that are associated with organic compounds could, therefore, be affected if ash application changes the leaching rate of the organic compounds. However, at Bredaryd North, no significant effect on the TOC concentration was detected. The decrease which occurred soon after ash application could reflect seasonal variation (Figure 7). The lack of response in the concentration of several metals which were present in the ash indicates that these metals either had a low solubility or were retained in the peatland during the period of this study.

Increased concentrations of K\(^{+}\), Ca\(^{2+}\), Mg\(^{2+}\), Mn and SO\(_4\)\(^{2-}\) were recorded in the present study. Similar results, with minor exceptions, have previously been reported after ash application to peatlands (Nilsson & Lundin, 1996; Tulonen et al., 2002; Piirainen & Domisch, 2004). At Bredaryd North, B concentration increased in ditch water and the same response was found in groundwater. Ernfors et al. (2010) report elevated B concentrations in one-year-old needles collected two and a half years following wood-ash application, and Nieminen et al. (2005) found, in a field study, that Na, K, B and S were readily released from wood ash. A rapid release of Na and K from wood ash was also reported from a laboratory experiment (Steenari et al., 1999), and was indicated in a soil solution study after wood-ash application to a mineral soil site (Ring et al., 2006). Thus, it is likely that the elevated concentrations of K\(^{+}\), B and SO\(_4\)\(^{2-}\) at Bredaryd North originated from the ash and not from exchange reactions with the soil. The decreases in the concentration of Fe, TS and SO\(_4\)\(^{2-}\) during the last two years of the study could not be explained.

Although Na\(^{+}\) and Cl\(^{-}\) have been found to leach readily from wood ash (Steenari et al., 1999), the Na\(^{+}\) and Cl\(^{-}\) concentrations responded weakly to the ash application at Bredaryd North. One explanation could be that the study site was located in an area where the deposition of Na\(^{+}\) and Cl\(^{-}\) is rather high (Lövblad et al., 2000). The throughfall deposition at a nearby spruce forest was, on average, 28 kg Cl\(^{-}\) ha\(^{-1}\) yr\(^{-1}\) in 1988/89 to 2005/06 (data from the Swedish Environmental Research Institute, www.ivl.se). Thus, when additional Na\(^{+}\) and Cl\(^{-}\) were applied in the wood ash, its potential effects on the Na\(^{+}\) and Cl\(^{-}\) concentration in water could have been masked by high background levels.

The K load applied was 4.5 times the P
load, while the estimated leaching rate of K⁺ was up to 27 times higher than for P following application (Table 1). This could help explain the more prolonged effect on the P concentration in needles, compared to the K concentration, observed following fertilisation with P and K of Scots pine stands deficient in these elements. In Scots pine needles, the P concentration is generally elevated for more than 30 years whereas the K concentration is elevated for 10–20 years following fertilisation with P and K (Silfverberg & Moilanen, 2008).

Other studies have shown or suggested that the P contained in ash typically has limited solubility (Tulonen et al., 2002; Nieminen et al., 2005, 2007). At Bredaryd North, the TP concentration in ditch water increased according to RIA and elevated concentrations were observed in groundwater two months after application. There was a period when the PO₄³⁻-P concentration in ditch water exceeded the TP concentration, possibly indicating an error, or reflecting the variation, in the chemical analyses. The TP, PO₄³⁻-P and NH₄⁺-N concentrations displayed a number of peaks following ash application. The temporal pattern of these variables was similar and appeared to be related to the discharge rate (cf. Figures 9–10).

There is uncertainty associated with the calculated leaching; this can be attributed to the interpolation of concentrations between samplings, the estimate of the catchment area, the calculated discharge rates, and the short reference period. The discharge rates calculated using the HBV model appear reasonable when compared with recorded data from other stations in the region and with one previous model simulation for a neighbouring river. On average, the simulated monthly mean discharge has been found to differ less than 10% from recorded monthly mean discharge at most of the stations (Johansson, 1992; M. Gardelin, pers. comm., 2011). For a single month, however, the difference could be larger, especially for small catchments. Although the annual runoff varied substantially between years, there was no significant difference in calculated daily discharge levels between the reference and post-treatment periods according to RIA. Since wood-ash application can increase tree growth and thereby evapotranspiration, runoff may decline. At a forest peatland nearby the Bredaryd catchments, the groundwater level was lowered, compared to the reference plots, in plots that had received wood ash five years earlier, possibly because tree growth had increased (Ernfors et al., 2010).

The pH of the ditch water increased significantly. Before ash application, dissolved organic compounds, possibly organic acids, seem to have had an important influence on pH, as suggested by the negative correlation between pH and the TOC concentration. After ash application, no significant correlation with TOC was detected, suggesting that pH was controlled by other factors. One possible explanation for the increase in pH could be that significant runoff seems to have taken place through the upper soil layer, where alkaline substances could have been released from the ash and transported to the main ditch. Tulonen et al. (2002) reported a slight increase in the pH of surface water following ash application to peatland forests in Finland.

The hydraulic conductivity of peat typically decreases with increasing soil depth (Paavilainen & Päivänen, 1995). Groundwater flow through superficial soil layers could therefore have been important, since the groundwater level tended to be rather shallow at Bredaryd North. Indeed, the depth gradients of the B, K⁺ and Li concentrations in groundwater indicated superficial flow. The concentrations of these elements in ditch water, at the time of groundwater sampling, were similar to the concentrations in groundwater at a depth of 30–40 cm (Table 3).

Although the original experimental design was spoiled by windthrow, the temporal pattern, the consistent response in ditch water and groundwater chemistry,
the statistical analyses and the corresponding results compared with previous studies indicate that the reported effects were largely caused by the ash application and were not an expression of normal variation. However, the Bredaryd study represents one case and, from a strictly statistical viewpoint, the conclusions of the study are not general, since the study was unreplicated.

The present study shows that wood-ash application to a drained peatland affects several chemical variables in ditch water for a period ranging from a few months up to at least three years. Groundwater flow through superficial soil layers appears to have facilitated leaching.

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