A Combined Measurement and Modelling Approach to Assess the Sustainability of Whole-Tree Harvesting—A Swedish Case Study

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Abstract: The demand of renewable energy has increased the interest in whole-tree harvesting. The sustainability of whole-tree harvesting after clear-cutting, from an acidification point of view, depends on two factors: the present acidification status and the further loss of buffering capacity at harvesting. The aims of this study were to investigate the relationship between these two factors at 26 sites along an acidification gradient in Sweden, to divide the sites into risk classes, and to examine the geographical distribution of them in order to provide policy-relevant insights. The present status was represented by the acid neutralizing capacity (ANC) in soil solution, and the loss of buffering capacity was represented by the estimated exceedance of critical biomass harvesting (CBH). The sites were divided into three risk classes combining ANC and exceedance of CBH. ANC and exceedance were negatively correlated, and most sites had either ANC < 0 and exceedance (high risk) or ANC > 0 and no exceedance (low risk). There was a geographical pattern, with the high risk class concentrated to southern Sweden, which was mainly explained by higher historical sulfur deposition and site productivity in the south. The risk classes can be used in the formulation of policies on whole-tree harvesting and wood ash recycling.

Keywords: acidification; forest management; deposition; base cations; site productivity; wood-ash recycling; Sweden

1. Introduction

Emissions of sulfur (S) and nitrogen (N) from the burning of fossil fuels have led to acidic deposition, peaking in the second half of the 20th century [1,2] and resulting in acidified soils and waters in the forest ecosystems of Europe [3]. These emissions have been reduced considerably, but both measurements [4–6] and modelling [7] indicate that the recovery of forest soils, which is required for surface waters to fully recover, is slow compared to the emission reductions. Recovery may slow down more, or even be reversed, if branches, tops, and stumps are harvested, since harvesting is an acidifying process [8,9] which is reinforced if other parts than the stems are harvested [10–13]. Removal of branches and tops means a significantly increased removal of base cations, due to the higher base cation concentrations in those parts [14,15].

The demand for renewable fuel is expected to increase to replace fossil fuel and thus mitigate climate change [16]. In Sweden, the harvesting of branches and tops were notified on 35–41% of the notified final fellings during the five years between 2015 and 2019, whereas stump harvesting was very uncommon [17]. Estimations of the future biomass potential in Sweden have shown that the harvesting of branches and tops has the potential
to increase [18]. It is therefore important to be able to assess the sustainability of whole-tree harvesting in regions with different acidification history and site properties.

The sustainability of whole-tree harvesting at a site, from an acidification point of view, depends on two factors: (1) how the site has been affected by previous acidification and (2) how whole-tree harvesting will affect the buffering capacity of the soil. The effects of anthropogenic acidification on soils are often described by soil solution chemistry below the root zone [4–6,19]. Acid neutralizing capacity (ANC) is an indicator often used to provide a quantitative estimate of the acidification status at a site as well as the quality of the water leaving the root zone [20]. However, it does not give any indications about the effect of whole-tree harvesting. The effect of whole-tree harvesting on the buffering capacity can be estimated using acidity budget calculations, e.g., through the concept of critical biomass harvesting (CBH) [13]. Such estimates can give indications of how the buffering capacity is affected by whole-tree harvesting, but since they only give information about the direction and a rate of the development, and not about the starting point, they do not give a complete assessment of the risks related to whole-tree harvesting. By combining ANC data based on soil solution chemistry measurements with acidity budget calculations, both previous acidification and the effect of whole-tree harvesting on the buffering capacity of the soil can be taken into account.

In this study we perform a refined mapping of the risks of acidification of forest soils caused by whole-tree harvesting, in this article defined as the harvesting of branches and tops but not stumps after clear-cutting, across a strong deposition gradient in Sweden. We achieve this by combining data on the present acidification status, represented by ANC in soil solution, with estimates of the potential effect of whole-tree harvesting on the buffering capacity, represented by exceedance of CBH, at 26 well investigated spruce sites. The aims were to (1) investigate the relationship between ANC in soil solution and exceedance of CBH, (2) identify risk classes for the sustainability of whole-tree harvesting, taking both factors into account, (3) investigate the geographical distribution of the risk classes and (4) discuss the results in relation to policies about whole-tree harvesting and wood ash recycling.

2. Materials and Methods

2.1. Study Sites and Measurements

The study was performed at 26 spruce sites from the Swedish Throughfall Monitoring Network (SWETHRO) [21], most of them active during all five years in the period 2014–2018 (Table 1). The 26 sites are geographically distributed across all three regions of Sweden—South, Central and North (Figure 1a)—to cover the steeply decreasing gradient of atmospheric S deposition from southwest to the north (Figure 1b). The site density is highest in the south of Sweden where deposition is highest, which is reflected in the number of sites in the three regions (Table 1).

The SWETHRO sites are 30 by 30 m squared plots within managed forest stands, where atmospheric deposition and soil solution chemistry are measured continuously. Throughfall deposition, i.e., precipitation that passed through the canopies, is collected monthly using open buckets in the winter, and polyethylene bottles with funnels threaded into the lid in the summer. Ten throughfall collectors are placed on each site, in an “L” shape along two of the borders of the monitoring area. The water samples are merged to one composite sample at the end of each month for chemical analysis at the accredited laboratory at IVL Swedish Environmental Research Institute and analyzed as described in previous studies [5]. The amount of throughfall precipitation per hectare is estimated based on the volume of water and the known diameter of the open bucket/funnel, and the throughfall deposition is derived by multiplying the amount of throughfall precipitation with measured concentrations.
Table 1. Characteristics for the 26 SWETHRO sites included in the study. The sites are organized from north to south.

| Site   | Lat     | Long    | Site Index | Moisture Class | Stoni-ness, % | Data Period | BC Dep OF | No of Lysimeters | Region |
|--------|---------|---------|------------|---------------|---------------|-------------|------------|-------------------|--------|
| BD06   | 66.066  | 21.468  | G18 2      | 4             | 20            | 2016–2018   | Yes        | 5                 | North  |
| AC34   | 65.945  | 16.314  | G17 4      | 3             | 20            | 2014–2018   | No         | 4–5               | North  |
| AC04   | 65.408  | 18.112  | G16 3      | 4             | 20            | 2014–2018   | Yes        | 5                 | North  |
| AC35   | 64.542  | 21.086  | G20 2      | 3             | 20            | 2014–2018   | Yes        | 5–6               | North  |
| Y07    | 62.284  | 16.348  | G20 3      | 3             | 60            | 2014–2018   | No         | 5–6               | North  |
| U06    | 59.941  | 16.524  | G34 3      | 4             | 60            | 2014–2018   | Yes        | 5                 | Central|
| S22    | 59.821  | 12.902  | G32 3      | 3             | 5             | 2014–2018   | Yes        | 5                 | Central|
| S05    | 59.008  | 13.110  | G35 3      | 3             | 5             | 2014–2016   | Yes        | 5                 | Central|
| R09    | 58.625  | 13.777  | G30 3      | 3             | 5             | 2014–2018   | No         | 5                 | South  |
| O35    | 58.441  | 11.731  | G26 3      | 4             | 5             | 2014–2017   | Yes        | 3–5               | South  |
| E21    | 58.156  | 15.435  | G32 3      | 3             | 5             | 2014–2018   | No         | 3–5               | South  |
| P95    | 57.866  | 12.673  | G26 3      | 3             | 15            | 2014–2018   | Yes        | 5                 | South  |
| F22    | 57.839  | 15.000  | G28 3      | 3             | 35            | 2014–2018   | No         | 5                 | South  |
| F12    | 57.825  | 14.394  | G28 3      | 3             | 5             | 2014–2018   | No         | 5                 | South  |
| F23    | 57.509  | 15.341  | G32 3      | 2             | 10            | 2014–2018   | Yes        | 4                 | South  |
| F18    | 57.149  | 13.594  | G26 3      | 3             | 10            | 2014–2018   | No         | 5                 | South  |
| G22    | 57.061  | 14.374  | G28 3      | 4             | 5             | 2014–2018   | Yes        | 5                 | South  |
| N12    | 56.953  | 12.816  | G30 3      | 2             | 50            | 2014–2018   | No         | 4–5               | South  |
| H03    | 56.853  | 16.316  | G28 3      | 3             | 30            | 2014–2018   | Yes        | 5                 | South  |
| N13    | 56.772  | 13.158  | G30 3      | 3             | 20            | 2014–2018   | Yes        | 5                 | South  |
| H22    | 56.638  | 15.629  | G32 3      | 3             | 20            | 2014–2018   | No         | 4                 | South  |
| N19    | 56.355  | 12.993  | G32 3      | 3             | 10            | 2014–2018   | No         | 5                 | South  |
| K13    | 56.270  | 15.453  | G34 3      | 3             | 10            | 2014–2016   | No         | 3–4               | South  |
| L18    | 55.177  | 13.521  | G30 3      | 3             | 10            | 2014–2018   | Yes        | 5                 | South  |
| L15    | 55.620  | 14.094  | G34 3      | 3             | 20            | 2014–2018   | No         | 5                 | South  |
| M16    | 66.066  | 21.468  | G36 3      | 3             | 5             | 2014–2018   | Yes        | 5                 | South  |

1 Site index is directly linked to site productivity (m³ ha⁻¹ y⁻¹) and is a measure of the optimal growth of a stand. The letter “G” means that the tree species is Norway spruce and the number is the projected tree height when the stand is 100 years old. 2 Field assessments in 2020. 3 From the ICP level II database (International Co-operative Programme on Assessment and Monitoring of Air Pollution Effects on Forests, http://www.icp-forests.org (accessed on 19 February 2021)). 4 Assessment based on satellite data and information from a nearby wood-land key habitat. 5 From forest owner/forest manager/Forest Agency. 6 Class 2: Dry (0.15 m³ m⁻³), Class 3: Moderately moist (0.20 m³ m⁻³), Class 4: Moist (0.25 m³ m⁻³). Translation to volumetric water content is based on [22]. 7 Years available in the period 2014–2018. 8 Information on whether there is an open field site close by, where base cation (BC) wet deposition is measured.

For some of the sites, there are also wet deposition measurements on an open field close by, which are used to separate between wet and dry deposition. Wet deposition is measured using a 1.5 m high collector made of polyvinylchloride. A polyethylene plastic bag is placed inside the cylinder and changed at every sampling occasion. In addition, a robust plastic ring with a smooth rim is attached to the top of the cylinder. A net made of polyolefin is placed at the opening of the cylinder during spring, summer and autumn, to prevent debris to enter the plastic bag. During 2016 and 2017, the wet deposition samplers were equipped with “birdrings”, positioned horizontally around the openings of the sampler to prevent birds from sitting on the rim of the sampler.

To be able to quantify dry deposition for the chemical species that interact with the tree crowns, e.g., N, Ca, Mg, and K, string samplers are placed at the open field of some of the sites. The string samplers are placed under a transparent roof made of polycarbonate. The samplers consist of Teflon strings. They are sprayed with deionized water once a month, all year round, and the samples are sent to the laboratory for chemical analysis. During wintertime the string samplers sometimes have to be brought indoors before being sprayed with deionized water [23].
At each of the 26 sites, soil solution samples from a depth of 50 cm in the mineral soil are collected using 3–6 separate suction lysimeters with ceramic cups (P 80) (Table 1). Samples are taken three times a year: before (February–May), during (May–September), and after (September–December) the vegetation period. At the time of the sampling, negative pressure is initiated so that the lysimeter can suck in water, and after two days the water is collected. For each occasion, the water samples from the lysimeters are merged into one composite sample for analysis. If there is no collectible water, the site is still not revisited until the next scheduled sampling occasion. Drier sites therefore have fewer measurements of soil water concentrations.

Starting in 2010, a soil sampling campaign was performed at the SWETHRO sites. Soil samples from 4–5 distinct soil horizons, down to 50 cm in the mineral soil, were collected and sent for chemical analysis of, e.g., total elemental content and grain size distribution. Cylinders (three per soil sample) were used to estimate soil bulk density. Stoniness and moisture were assessed in field at all sites.

### 2.2. Estimation of ANC in Soil Solution

Acid neutralizing capacity (ANC in meq L\(^{-1}\)) in soil solution was calculated for each site and each sampling occasion as:

\[
ANC = [\text{Ca}^{2+}] + [\text{Mg}^{2+}] + [\text{K}^+] + [\text{Na}^+] + [\text{NH}_4^+] - [\text{SO}_4^{2-}] - [\text{Cl}^-] - [\text{NO}_3^-] \tag{1}
\]

where all element concentrations are given in meq L\(^{-1}\). The median ANC for the available samples in 2014–2018 (Table 1) was calculated and used for analyses in this study.
2.3. Estimation of Exceedance of Critical Biomass Harvesting (CBH)

2.3.1. Concept

The concept of critical biomass harvesting (CBH [13]) has been used in Swedish policies since 2018, when a new indicator “Acidification from forestry” was introduced in the Swedish Environmental Objective Framework. CBH builds on the same principle as critical loads (CL), which was a successful concept in reducing acidic deposition [19].

The basis for both CL and CBH calculations is the SMB formula (Equation (2), [24])

\[ S_{\text{dep}} + N_{\text{dep}} + Cl_{\text{dep}} + BC_{\text{harv}} + Alk_{\text{leach}} = BC_{\text{dep}} + BC_{\text{weath}} + N_{\text{imm}} + N_{\text{harv}} + N_{\text{de}} \]  

where \( S \) = deposition, \( BC \) = base cations (Ca, Mg, Na and K), \( N \) = net losses at harvesting, \( Alk \) = Alkalinity, \( leach \) = leaching, \( weath \) = weathering, \( imm \) = immobilization, and \( de \) = denitrification (meq m\(^{-2} \) y\(^{-1} \)).

In CL calculations, the critical load of acidity, i.e., the maximum amount of acid deposition (\( S + N \)) that can be allowed without negative ecosystem effects, is estimated through Equation (3), where all factors except \( S_{\text{dep}} \) and \( N_{\text{dep}} \) have been moved to the right side of the equation and the \( Alk_{\text{leach(crit)}} \) is the critical alkalinity leaching.

To calculate CL at a site, all factors on the right side of the equation except \( Alk_{\text{leach(crit)}} \) are quantified based on site data. A chemical criterion and a critical limit are defined, linking acid deposition to ecosystem effects. They are used to calculate \( Alk_{\text{leach(crit)}} \), which is a measure of the acidification of runoff water. Exceedance is then calculated according to Equation (4), where the actual deposition at the site is compared with the CL.

\[ \text{CL} (S_{\text{dep}} + N_{\text{dep}}) = BC_{\text{dep}} + BC_{\text{weath}} + N_{\text{imm}} + N_{\text{harv}} + N_{\text{de}} - Cl_{\text{dep}} - BC_{\text{harv}} - Alk_{\text{leach(crit)}} \]  

Exceedance = \( S_{\text{dep}} + N_{\text{dep}} - \text{CL} (S_{\text{dep}} + N_{\text{dep}}) \)  

In a national study in Sweden, CBH was defined as the maximum biomass extraction that does not lead to an ANC value less than zero in the water leaving the root zone, and therefore does not lead to export of acidity [13]. Biomass harvesting contributes to the acidity flows by removing BC from the forest ecosystem. In the CBH calculations, the critical removal of BC through harvesting was quantified and compared with the actual biomass harvesting, based on Equation (2) and the same logics as in the CL calculations in Equations (3) and (4), but with a few simplifications. The chemical criterion used was ANC, and since the critical limit was set to 0, the \( Alk_{\text{leach(crit)}} \) factor disappeared. Furthermore, the N cycling was greatly simplified based on three assumptions:

- Only N that leaches is affecting acidity: NO\(_3\)-N leaching is acidifying, one equivalent, and NH\(_4\)-N leaching counteracts acidification, one equivalent, based on theories in [25].
- N leaching is not affected by whole-tree harvesting.
- N accumulating in soil organic matter will not contribute to acidification in the future.

By implementing those assumptions in Equation (1), CBH could be calculated using Equation (5), and exceedance could be calculated using Equation (6).

\[ \text{CBH} (BC_{\text{harv}}) = BC_{\text{weath}} + BC_{\text{dep}} + NH_4^- N_{\text{leach}} - S_{\text{dep}} - Cl_{\text{dep}} - NO_3^- N_{\text{leach}} \]  

Exceedance = \( BC_{\text{harv}} - \text{CBH} (BC_{\text{harv}}) \)  

In this study, the same formulas and assumptions as in the national CBH calculations were applied [13], and calculations were made for the upper 50 cm in the mineral soil, which is assumed to be the root zone for spruce. The calculations assume a steady state, meaning that all fluxes are given as annual averages over an entire forest rotation, varying according to site productivity. Therefore, the acidification history of a site is not taken into account, and the results only show a direction and a rate of the change.
2.3.2. The Weathering Model PROFILE

Whereas deposition, leaching, and removal of BC by harvesting can be based on site measurements, weathering rates were modelled using the PROFILE model [26]. PROFILE is a steady state soil geochemical model, in which weathering rates are calculated using transition state theory. Geochemical properties of the soil, e.g., mineral composition, mineral surface area, and soil moisture, are required as input, along with atmospheric deposition, climate, and biological parameters. An overview of the input data required for PROFILE and for calculations of CBH and its exceedance is given in Table 2, and a detailed description of the data and how they were processed is given in the following paragraphs.

Table 2. Main input data used for weathering modelling (Weath), calculation of critical biomass harvesting (CBH), and exceedance (Exc).

| Parameter          | Use          | Source                                      |
|--------------------|--------------|---------------------------------------------|
| Deposition S, Cl   | Weath, CBH   | Field measurements                          |
| Deposition of N    | Weath        | Based on field measurements                 |
| Deposition of Ca, Mg, K | Weath, CBH | Based on field measurements and modelling |
| Deposition of Na   | Weath, CBH   | Based on Cl deposition                      |
| Leaching           |              |                                             |
| NH$_4$-N$_{leach}$ | CBH          | Based on field measurements and runoff      |
| NO$_3$-N$_{leach}$ | CBH          | Based on field measurements, runoff, and an empirical function |
| Climate            |              |                                             |
| Temperature        | Weath        | Estimated by SMHI $^1$, based on measurements |
| Precipitation      | Weath        | Estimated by SMHI $^1$, based on measurements |
| Runoff             | Weath, CBH   | Estimated by SMHI $^1$, based on measurements |
| Soil properties    |              |                                             |
| Mineral composition| Weath        | Modelled with A 2M $^2$, based on field measurements |
| Specific surface area | Weath  | Estimated based on field measurements       |
| Stoniness          | Weath        | Field observations                          |
| Moisture           | Weath        | Field observations                          |
| Soil bulk density  | Weath        | Estimated based on field measurements       |
| Forest data        |              |                                             |
| Net harvest loss of BC $^3$ | Weath, Exc | Estimated based on site index from sites (Table 1) |
| Net harvest loss of N $^3$ | Weath     | Estimated based on site index from sites (Table 1) |

$^1$ Swedish Meteorological and Hydrological Institute. $^2$ The A2M program [27]. $^3$ Net loss at harvesting of biomass (stems, branches, and tops).

2.3.3. Input Data

Deposition was required both for PROFILE modelling and for calculating CBH and its exceedance. The average deposition of S, Cl, N and BC for the years 2014–2018 was based on SWETHRO measurements (Appendix A). For S and Cl, where the canopy exchange was assumed to be very small, throughfall deposition was used. For N and the base cations Ca, Mg, and K, the canopy exchange is substantial, and thus the throughfall deposition could not be used as it is. For N, wet deposition was measured on all 26 sites. Dry deposition was estimated for ten SWETHRO sites in Sweden, based on measurements and calculations included in the surrogate surface method [23]. These results were generalized to all 26 sites, based on a method where the share of dry to total deposition of inorganic N (NO$_3$-N + NH$_4$-N) could be correlated with geographical position [23]. Wet deposition of Ca, Mg, and K for 2014–2018 was available for 14 of the 26 sites (Table 1). For the sites missing measurements, wet deposition was derived from interpolation using the inverse distance method. For this interpolation, the 14 sites, and additionally 12 sites from the SWETHRO network not included in this study but for which wet deposition data were available, were used. Dry deposition of Ca, Mg, and K was estimated based on the relationship between wet and dry deposition in an extensive modelling study based on data from 1998 [28,29], assuming that the relationship between wet and dry deposition has been
constant over time. Na was assumed to come only from sea salt, and was calculated based on Cl deposition and the sea salt composition.

Climate data was required for the weathering modelling. Temperature, precipitation, and runoff for the years 2014–2018 were obtained from Swedish Meteorological and Hydrological Institute (SMHI) (Appendix A). Temperature and precipitation were extracted from the database PTHBV (data delivered in August 2020). Runoff was derived from SMHI:s water web, with data for 40,000 subcatchments in Sweden.

Leaching of N in the form of NH$_4$-N and NO$_3$-N was required for the CBH calculations, and was based on measurements of concentrations in soil solution at 50 cm depth and runoff (Table 3; Appendix A). Median concentrations from the years 2014–2018 were multiplied with runoff from the sites. For the results to be representative for a forest rotation, NO$_3$-N after clearcutting was added. For that, an empirical function from was used, where NO$_3$-N leaching in the clearcut phase was related to site productivity [30].

**Table 3.** Soil solution chemistry, medians for the period 2014–2018 (meq L$^{-1}$). The sites are arranged from north to south. The last column “n” is the number of samples.

| Site | S  | Cl   | NO$_3$-N | NH$_2$-N | Ca  | Mg  | Na  | K  | ANC  | n  |
|------|----|------|----------|----------|-----|-----|-----|----|------|----|
| BD06 | 0.03 | 0.02 | 0.00     | 0.00     | 0.04 | 0.03 | 0.05 | 0.01 | 0.07  | 6  |
| AC34 | 0.09 | 0.05 | 0.00     | 0.00     | 0.42 | 0.03 | 0.07 | 0.01 | 0.39  | 15 |
| AC04 | 0.03 | 0.02 | 0.00     | 0.00     | 0.03 | 0.02 | 0.05 | 0.01 | 0.06  | 15 |
| AC35 | 0.12 | 0.02 | 0.00     | 0.00     | 0.04 | 0.05 | 0.06 | 0.02 | 0.05  | 10 |
| Y07  | 0.02 | 0.02 | 0.00     | 0.00     | 0.03 | 0.02 | 0.06 | 0.00 | 0.05  | 13 |
| U06  | 0.05 | 0.07 | 0.00     | 0.00     | 0.05 | 0.04 | 0.10 | 0.01 | 0.09  | 2  |
| S22  | 0.08 | 0.12 | 0.00     | 0.00     | 0.07 | 0.02 | 0.12 | 0.00 | −0.003 | 14 |
| S05  | 0.07 | 0.15 | 0.00     | 0.00     | 0.03 | 0.03 | 0.19 | 0.00 | 0.02  | 9  |
| R09  | 0.28 | 0.31 | 0.00     | 0.00     | 0.04 | 0.11 | 0.43 | 0.01 | 0.02  | 14 |
| Q35  | 0.07 | 0.45 | 0.00     | 0.00     | 0.01 | 0.07 | 0.37 | 0.01 | −0.07 | 11 |
| E21  | 0.15 | 0.02 | 0.00     | 0.00     | 0.10 | 0.04 | 0.04 | 0.04 | 0.05  | 9  |
| P95  | 0.11 | 0.34 | 0.00     | 0.00     | 0.03 | 0.08 | 0.40 | 0.01 | 0.04  | 35 |
| F22  | 0.10 | 0.26 | 0.00     | 0.00     | 0.04 | 0.03 | 0.22 | 0.01 | −0.06 | 8  |
| F12  | 0.13 | 0.23 | 0.00     | 0.00     | 0.10 | 0.03 | 0.21 | 0.00 | −0.005 | 9  |
| F23  | 0.28 | 0.48 | 0.00     | 0.00     | 0.07 | 0.08 | 0.39 | 0.01 | −0.20 | 14 |
| F18  | 0.09 | 0.26 | 0.00     | 0.00     | 0.02 | 0.02 | 0.22 | 0.00 | −0.05 | 12 |
| G22  | 0.17 | 0.40 | 0.00     | 0.00     | 0.07 | 0.10 | 0.41 | 0.02 | 0.02  | 12 |
| N12  | 0.10 | 0.37 | 0.00     | 0.00     | 0.02 | 0.07 | 0.46 | 0.02 | −0.03  | 14 |
| H03  | 0.59 | 0.88 | 0.00     | 0.00     | 0.23 | 0.24 | 0.85 | 0.01 | −0.01 | 8  |
| N13  | 0.14 | 0.52 | 0.01     | 0.00     | 0.01 | 0.04 | 0.53 | 0.00 | −0.13 | 12 |
| H22  | 0.12 | 0.13 | 0.00     | 0.00     | 0.04 | 0.04 | 0.14 | 0.00 | −0.01 | 11 |
| N19  | 0.09 | 0.59 | 0.00     | 0.00     | 0.01 | 0.04 | 0.40 | 0.01 | −0.04  | 12 |
| K13  | 0.26 | 0.46 | 0.00     | 0.00     | 0.03 | 0.06 | 0.51 | 0.01 | −0.11  | 18 |
| L18  | 0.19 | 0.78 | 0.17     | 0.00     | 0.02 | 0.08 | 0.76 | 0.01 | −0.29 | 16 |
| L15  | 0.12 | 0.41 | 0.00     | 0.00     | 0.01 | 0.02 | 0.39 | 0.00 | −0.13 | 11 |
| M16  | 0.15 | 0.74 | 0.45     | 0.00     | 0.13 | 0.18 | 0.35 | 0.02 | −0.22  | 11 |

Net harvest losses of N and BC by whole-tree harvesting were required inputs for weathering modelling with the PROFILE model, and net harvest losses of BC were also important in the calculations of exceedance. The net harvest losses were calculated based on site productivity from the sites, derived from site index (Table 1). The site productivity is the growth of a stand under optimal conditions, and to imitate real conditions the site productivity was reduced by 20%, in accordance with earlier mass balance studies [13,31]. All stems and 60% of the branches, accompanied by 75% of their needles, were assumed to be harvested, in accordance with a scenario from the Swedish Forest Agency [32]. The calculations were performed in the same way as in a national CBH study [13], and the methodology along with densities and nutrient concentrations in different tree parts are given there.

Data on soil properties were important inputs to the weathering model PROFILE. Soil data in this study came from soil samplings at the sites, performed between 2010 and 2016. One of the required inputs was mineralogy for all soil layers, which was not
measured, but can be calculated from measured total elemental content, quantified by plasma emission spectrometry analysis (ICP-AES). The total elemental content was used in the A2M program [27], model version 1.41, to calculate all possible mineral modes given a set of allowed minerals. However, for the organic upper layer the total elemental content from the second layer was used since the organic material in the organic layer substantially affects the total elemental content. For each layer at each site, the arithmetic means from A2M were then selected to represent the mineralogy at each layer. In absence of any other information, this was regarded as the best solution as it represents the barycentre of the solution polyhedron spanned by the extreme modes [27]. Mineralogies used for the sites are presented in Appendix B.

Soil bulk density was derived by weighing the dried samples collected with cylinders with known volume. Specific surface area was estimated based on grain size distribution and an empirically derived formula [33]. Stoniness (volume fraction of stones and boulders) and moisture were estimated based on visual assessments on the sites. Moisture was translated from a moisture class to m$^3$ water per m$^3$ soil [22]. Soil input data are described in Appendices C and D.

2.4. Risk Classification

As a basis for the risk classification, factor two, exceedance of CBH was plotted against factor one, ANC in soil solution. The sites were then grouped into three classes, based on their position in the graph (Table 4). Sites where both factors indicated risks related to acidification, i.e., sites where ANC was negative and whole-tree harvesting led to an exceedance of CBH, were placed in Class 1. Sites for which one of the factors indicated a risk but not the other, i.e., either negative ANC and no exceedance, or positive ANC and exceedance, were placed in Class 2. Finally, sites where none of the factors indicated risks related to acidification, i.e., sites for which ANC was positive and whole-tree harvesting did not lead to an exceedance of CBH, were placed in Class 3.

Table 4. Risk classes for whole-tree harvesting sustainability, based on present acidification status (acid neutralizing capacity (ANC) in soil solution) and exceedance of CBH.

| Class | Outcome for the Two Risk Factors | Risk Related to Acidification from Whole-Tree Harvesting |
|-------|----------------------------------|--------------------------------------------------------|
| 1     | ANC < 0 and Exc$_{CBH}$ > 0      | High                                                   |
| 2     | ANC < 0 and Exc$_{CBH}$ < 0 or ANC > 0 and Exc$_{CBH}$ > 0 | Medium                                                |
| 3     | ANC > 0 and Exc$_{CBH}$ < 0      | Low                                                   |

3. Results

3.1. Present Soil Status: ANC in Soil Water (Factor 1)

ANC in soil solution spanned from $-0.29$ meq L$^{-1}$ to $0.39$ meq L$^{-1}$, along a geographical gradient with generally higher ANC towards the north, although several deviations occurred at the regional scale (Figure 2, Table 3). In the northern region, ANC was clearly positive at all five sites. In the central region, ANC was positive at two sites and negative at one. In the southern region only four of the eighteen sites showed a positive ANC.

3.2. Effect of Whole-Tree Harvesting: Exceedance of CBH (Factor 2)

The CBH calculations showed an exceedance of critical harvesting for 21 of the 26 sites (Figure 3, Table 5). Just as for ANC in soil solution, there was a clear geographical gradient, with a generally decreasing exceedance towards the north. In the northern region, there was no exceedance for two of the five sites, and a low exceedance (up to 25 meq m$^{-2}$ y$^{-1}$) for the others. In the central and southern regions, the exceedance was above 25 meq m$^{-2}$ y$^{-1}$ for most of the sites, and at three of the sites in southernmost Sweden the exceedance was over 100 meq m$^{-2}$ y$^{-1}$. In the central and southern regions, three sites stood out with negative exceedance.
Figure 2. ANC in soil solution at a 50 cm depth in the mineral soil as medians for the years 2014–2018, with three sampling occasions per year.

Figure 3. Exceedance of CBH if whole-tree harvesting is applied.
Table 5. CBH and its exceedance on the 26 sites, along with the different components in the calculations (meq m$^{-2}$ y$^{-1}$). The sites are arranged from north to south.

| Site | Weathering | Ca + Mg + K dep. | S dep. | NO$_3$-N Leaching | NH$_4$-N Leaching | BC Removal at Harvesting | CBH | Exc. |
|------|------------|------------------|--------|-------------------|-------------------|-------------------------|------|------|
| BD06 | 52.6       | 5.3              | 4.1    | 1.5               | 0.31              | 19.1                    | 51.9 | −32.7|
| AC34 | 16.5       | 6.2              | 3.1    | 1.4               | 0.59              | 17.7                    | 17.1 | 0.6  |
| AC04 | 16.9       | 6.0              | 3.3    | 1.2               | 0.44              | 16.2                    | 18.0 | −1.8 |
| AC35 | 15.2       | 8.2              | 11.9   | 1.9               | 0.37              | 22.0                    | 8.7  | 13.4 |
| Y07  | 7.7        | 6.7              | 4.8    | 1.9               | 0.47              | 22.0                    | 6.5  | 15.5 |
| U06  | 3.9        | 14.0             | 5.5    | 4.2               | 0.23              | 34.8                    | 6.6  | 28.2 |
| S22  | 15.9       | 10.4             | 12.4   | 8.0               | 0.49              | 60.2                    | 0.9  | 59.3 |
| S05  | 54.1       | 18.4             | 12.5   | 3.6               | 0.33              | 30.7                    | 47.5 | −16.8|
| R09  | 18.4       | 21.1             | 10.2   | 7.7               | 0.24              | 58.5                    | 13.0 | 45.6 |
| O35  | 12.9       | 33.3             | 32.7   | 8.4               | 0.66              | 45.8                    | −22.7| 68.5 |
| E21  | 25.7       | 21.8             | 6.9    | 8.9               | 0.20              | 65.5                    | 28.1 | 37.4 |
| P95  | 19.3       | 30.7             | 28.9   | 5.9               | 0.62              | 45.8                    | −9.5 | 55.2 |
| F22  | 10.8       | 22.3             | 8.8    | 6.8               | 0.25              | 52.1                    | 12.6 | 39.4 |
| F12  | 10.8       | 25.0             | 9.2    | 6.8               | 0.27              | 52.1                    | 12.4 | 39.7 |
| F23  | 13.9       | 39.6             | 11.2   | 8.7               | 0.24              | 65.5                    | 7.7  | 57.7 |
| F18  | 58.1       | 33.7             | 16.4   | 5.9               | 0.51              | 45.8                    | 53.6 | −7.8 |
| G22  | 63.0       | 24.9             | 11.9   | 6.8               | 0.37              | 52.1                    | 61.5 | −9.3 |
| N12  | 5.3        | 44.2             | 43.4   | 7.8               | 0.79              | 58.5                    | −44.2| 102.7|
| H03  | 17.0       | 25.9             | 11.5   | 6.8               | 0.19              | 52.1                    | 18.4 | 33.7 |
| N13  | 22.2       | 52.6             | 32.9   | 11.5              | 0.81              | 58.5                    | 4.6  | 53.9 |
| H22  | 30.2       | 26.1             | 12.5   | 8.7               | 0.25              | 65.5                    | 29.7 | 35.7 |
| N19  | 20.2       | 38.8             | 33.3   | 8.8               | 0.61              | 65.5                    | −5.6 | 71.0 |
| K13  | 24.0       | 28.1             | 19.4   | 9.9               | 0.26              | 73.0                    | 13.3 | 59.7 |
| L18  | 12.2       | 32.9             | 31.4   | 88.6              | 0.52              | 58.5                    | −96.6| 155.1|
| L15  | 4.1        | 34.4             | 37.0   | 9.9               | 0.40              | 73.0                    | −23.4| 96.4 |
| M16  | 4.9        | 34.1             | 50.9   | 135.6             | 0.30              | 80.5                    | −169.0| 249.5|

3.3. Risk Classification Based on the Two Factors

A comparison between ANC in soil solution and the exceedance of CBH for the 26 sites showed that exceedance was negatively and significantly ($p = 0.00025$) correlated with ANC in soil solution (Figure 4a). A slightly weaker correlation was also found between ANC and CBH ($p = 0.0046$).

![Figure 4. The sites divided into risk classes based on ANC in soil solution and exceedance of critical harvesting, shown as a graph (a) and a map (b).](image-url)
The risk classes (Table 4) were unevenly distributed in the country (Figure 4b, Table 6). Risk Class 1 was highly dominating in the southern region, whereas the sites in the northern region belonged to either Class 2 (three sites) or Class 3 (two sites). In the central region, with only three sites, all three classes were represented.

### Table 6. Distribution of the different risk classes among sites in the three regions.

| Region | Number of Sites | Class 1 | Class 2 | Class 3 |
|--------|----------------|---------|---------|---------|
| North  | 0              | 3       | 2       |
| Central| 1              | 1       | 1       |
| South  | 13             | 4       | 1       |

#### 4. Discussion

#### 4.1. Geographical Distribution of Risk Classes

The geographical pattern, with Risk Class 1 highly dominating in the southern region and Risk Class 2 and 3 dominating in the other regions, could be explained by the S deposition gradient with decreasing deposition towards the north (Figure 1b), and the site productivity gradient going in the same direction (represented by site index in Table 1). The historical S deposition, which has been higher than today but always followed a similar gradient [2], could be regarded as the main explanatory factor for the gradient for the first factor in the risk classification: ANC in soil solution. S deposition causes BC to leach from the soils [3], and the effect could be seen in the relationship between S and the base cations Ca, Mg, and K in soil solution, with generally more S compared to BC towards the south (Table 3). For the second factor, exceedance of CBH, site productivity was the most important reason for the gradient. Higher site productivity in the south leads to more removal of base cations at whole-tree harvesting (Table 5), which increases the risk of exceedance of CBH. This is consistent with the conclusions from a study at nine forest sites with different site productivity in Sweden and Scotland where fast growing forests led to more acidification than slow growing forests [8].

The sites in Risk Class 1 had all been exposed to high S deposition and all had high site productivity. The two sites with the lowest ANC, M16 and L18, situated in the southernmost part of Sweden, were the only sites with elevated NO$_3^-$-N concentrations in soil solution. This had a high impact both on ANC in soil solution and exceedance of CBH, and highlights the considerable potential of acidification from N deposition in areas where the N retention capacity is being exceeded [25,34].

Sites in Risk Class 3 appeared in the whole country, except for in the far south. Three of the sites, BD06, S05, and G22, had high weathering rates in comparison with most of the other sites (Table 5), which can partly explain both the high ANC and the negative exceedance. The site productivities for BD06 and S05 were low, which was also the case for the fourth site, AC04, and low site productivities were an important contributing factor for the negative exceedance at those sites.

Additionally, the sites in Risk Class 2 were spread over the country. Several of the sites had values close to 0 for ANC and/or for exceedance of CBH, and for those sites, uncertainties have a great impact in the risk classification. Seven of the eight sites in Class 2 had positive ANC, but the CBH calculations showed exceedance at whole-tree harvesting. Three of these sites, in the northern parts of the southern region, E21, R09, and P95, were situated outside the area with the highest historical S deposition (Figure 1b), but still in the part of southern Sweden where site productivities are generally high, which could explain the combination of positive ANC and exceedance of CBH. In northern Sweden, the exceedances were generally close to 0, and small differences in weathering rates, deposition or site productivity determined if the estimated exceedance was positive or negative. The site AC34 had an exceedance just above 0, but very high ANC. The high ANC...
indicates that there are sources of base cations not accounted for in the CBH calculations, e.g., that weathering rates are underestimated.

One of the eight sites in Class 2, F18, had a slightly negative ANC, but CBH was not exceeded at whole-tree harvesting. Negative exceedance at a site in this part of Sweden, where most sites show exceedance, could be explained by a combination of higher estimated weathering rates and a somewhat lower site productivity than at most surrounding sites.

The low density of sites in the central and northern regions in this study limited the possibilities to perform geographical generalizations. However, earlier studies of ANC in soil solution [21,35] and exceedance of CBH [13] at higher resolution can contribute to the interpretation of the results in this study. The ANC gradient over Sweden has been presented for 2006–2008 [21] and 2017–2019 [35], where ANC on all SWETHRO sites active those years were included (55 sites in 2006–2008 and 48 in 2017–2019). Although the ANC levels were generally slightly lower in 2006–2008 than in 2017–2019, indicating a slight recovery, both of those two studies showed positive ANC in the north (except for one site along the east coast in 2006–2008), ANC around 0 in the central part, and mainly negative ANC in the southern part, just as in the present study.

A high resolution national mapping of exceedance of CBH [13] showed that CBH was generally exceeded in spruce forests in Sweden at whole-tree harvesting, except for the inner part of northern Sweden, but it also showed that there were several exceptions for some areas in central Sweden and also a few areas in southern Sweden where there was no exceedance. This is consistent with the results from this study.

4.2. Policy Implications

At sites in Risk Class 1, from which the water leaving the root zone has no buffering capacity (ANC < 0) and where whole-tree harvesting leads to an exceedance of CBH, whole-tree harvesting is not sustainable from an acidification point of view. The sites in Risk Class 1 were situated in the southern part of Sweden, where recovery can be expected to be slow or non-existing even when whole-tree harvesting is not applied [5]. Whole-tree harvesting will lead to further loss of buffering capacity, which may inhibit recovery from acidification and thereby exacerbate the acidification status of the forest site. Wood ash recycling means that nutrients are returned to the soil, but there is a risk that the lost nutrients are only partly compensated for and/or that there is a time lag before the effect of wood ash recycling appears. Based on this, our assessment is that whole-tree harvesting at sites belonging to Risk Class 1 is not compatible with the Swedish environmental objective about acidification, even if the removal of base cations is compensated for by wood ash recycling.

For Risk Class 2, ANC and/or exceedance of CBH was often close to 0. Due to uncertainties in measurements and calculations, the class affiliation in itself was more uncertain than for Class 1. Sites that have a positive ANC, but an exceedance of CBH at whole-tree harvesting, have a better starting point than those in Risk Class 1, but the exceedance indicates that whole-tree harvesting is not sustainable in the long term; i.e., it will lead to loss of buffering capacity and a decrease in ANC. If ANC is decreasing at sites within this ANC interval, pH can be expected to be substantially affected. This was highlighted in a study on the effects of whole-tree harvesting and wood ash recycling on ANC in surface waters [36]. By compensating for the losses through wood ash recycling, the loss of buffering capacity caused by whole-tree harvesting can be inhibited. Sites in Risk Class 2 that have a negative ANC but no exceedance of CBH at whole-tree harvesting have the potential for recovery, but whole-tree harvesting will slow it down. We suggest that whole-tree harvesting at sites belonging to Risk Class 2 should be accompanied with wood-ash recycling.

In Risk Class 3, ANC was positive and the CBH was not exceeded at whole-tree harvesting. Our assessment is that the risk of negative effects of whole-tree harvesting on the acidification status is small, and that wood-ash recycling is not necessary at those sites.
In Risk Classes 1 and 2, where runoff water is already acidified or where acidification is in progress, there is a risk that nutritional imbalances will arise, which has been indicated in European studies [34,37]. This can, in turn, have a negative effect on tree vitality and tree growth [38,39]. Thus, although the risk assessment in this study focuses on the quality of the runoff water leaving the root zone, with potential effects on surface water, it is also highly relevant for soil acidification and tree nutrition. This study focuses on acidification, but there are other environmental aspects that can place restrictions on sustainable biomass harvesting, e.g., biodiversity [40]. These aspects should be taken into account in an overall risk assessment for sustainable biomass harvesting.

5. Conclusions

We investigated two factors for the assessment of the sustainability of whole-tree harvesting after clear-cutting: present acidification status represented by ANC in soil solution and exceedance of CBH. The two factors were negatively correlated, and the three risk classes, based on the two factors, showed a clear geographical pattern, although some deviations occurred. The gradients of historical S deposition and the site productivity were identified as the main causes of the geographical pattern. The high potential impact of acidification from N was highlighted at the two most extreme sites in the risk classification, where the N retention capacity was exceeded and nitrification caused acidification. The impact of soil weathering could be seen at single sites with high weathering rates, which were assigned in the low risk class, although historical S deposition and site productivity were high. The results strengthen the Swedish recommendations that wood ash recycling should be applied after whole-tree harvesting, except in the areas with low site productivity corresponding mainly to the inner part of northern Sweden. However, by including the present acidification status, we have also identified sites in southern Sweden where we assess that whole-tree harvesting is not compatible with the Swedish acidification objective.

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Data Availability Statement: Input data for weathering modelling and CBH calculations are available in Appendix A (climate and deposition), Appendix B (mineralogy), and Appendices C and D and Table 1 (soil properties). Site productivity is available in Table 1, and nutrient concentrations and tree densities used in the calculations of removal of nutrients are published in papers referred to in the methods section. ANC in soil solution used in the risk classification is presented in Table 3. Deposition and soil solution chemistry presented in this paper is average or median values. The original data can be downloaded from the IVL website, www.ivl.se (accessed on 19 February 2021).

Conflicts of Interest: The authors declare no conflict of interest.
Appendix A

Table A1. Climate data and atmospheric deposition.

| Site | T, °C | Prec, mm | Q, mm | S | NO$_3$-N | NH$_4$-N | Cl | Ca | Mg | K | Na |
|------|-------|----------|-------|---|-----------|---------|----|----|----|---|----|
| BD06 | 1.9   | 694      | 289   | 0.65 | 0.69      | 1.10    | 1.84| 0.59| 0.15| 0.44| 1.02|
| AC34  | 0.3   | 687      | 555   | 0.50 | 0.59      | 0.93    | 4.27| 0.57| 0.17| 0.78| 2.38|
| AC04  | 1.5   | 635      | 411   | 0.52 | 0.57      | 0.90    | 2.08| 0.50| 0.16| 0.85| 1.16|
| AC35  | 4.0   | 709      | 346   | 1.91 | 1.15      | 0.99    | 3.36| 0.79| 0.30| 0.74| 1.87|
| Y07   | 2.9   | 769      | 439   | 0.77 | 1.30      | 1.11    | 3.99| 0.76| 0.20| 0.49| 2.22|
| U06   | 6.9   | 599      | 217   | 0.88 | 2.17      | 2.31    | 4.68| 1.29| 0.44| 1.56| 2.61|
| S22   | 5.6   | 834      | 455   | 2.00 | 3.47      | 3.50    | 13.57| 0.93| 0.40| 0.99| 7.55|
| S05   | 8.0   | 706      | 304   | 2.00 | 3.83      | 3.82    | 22.99| 1.64| 0.75| 1.60| 12.79|
| R09   | 8.4   | 626      | 224   | 1.63 | 3.19      | 4.58    | 22.01| 1.65| 1.04| 1.68| 12.25|
| O35   | 7.9   | 1102     | 619   | 5.24 | 5.41      | 4.45    | 70.83| 2.12| 2.30| 1.49| 39.41|
| E21   | 7.5   | 653      | 191   | 1.10 | 2.96      | 4.30    | 9.75 | 1.75| 0.93| 2.12| 5.43|
| P95   | 7.4   | 1047     | 577   | 4.63 | 5.58      | 4.57    | 63.03| 1.94| 2.07| 1.55| 35.07|
| F22   | 7.0   | 671      | 236   | 1.41 | 3.28      | 4.76    | 12.64| 1.79| 0.93| 2.24| 7.03|
| F12   | 7.0   | 698      | 249   | 1.48 | 3.49      | 5.04    | 19.20| 1.88| 1.24| 2.15| 10.69|
| F23   | 7.3   | 681      | 223   | 1.80 | 3.30      | 4.82    | 15.16| 1.66| 0.62| 2.44| 8.43|
| F18   | 7.7   | 929      | 477   | 2.62 | 5.63      | 5.32    | 41.02| 2.24| 2.04| 2.26| 22.83|
| G22   | 7.7   | 754      | 348   | 1.91 | 3.93      | 5.63    | 20.21| 1.73| 1.13| 2.67| 11.25|
| N12   | 8.0   | 1169     | 734   | 6.95 | 6.55      | 6.19    | 107.88| 2.70| 3.05| 2.20| 60.03|
| H03   | 8.6   | 564      | 180   | 1.85 | 4.23      | 3.89    | 15.87| 2.72| 0.85| 2.09| 8.83|
| N13   | 8.3   | 1097     | 753   | 5.27 | 6.69      | 6.33    | 66.52| 3.03| 3.85| 2.27| 37.01|
| H22   | 8.1   | 696      | 231   | 2.00 | 4.56      | 4.25    | 14.11| 2.31| 1.12| 2.13| 7.85|
| N19   | 8.2   | 1208     | 568   | 5.34 | 5.73      | 8.67    | 57.48| 2.63| 2.39| 2.37| 31.98|
| K13   | 8.8   | 720      | 247   | 3.12 | 5.01      | 4.70    | 24.00| 2.34| 1.35| 2.11| 13.58|
| L18   | 8.6   | 873      | 487   | 5.03 | 5.70      | 8.68    | 55.40| 2.44| 1.77| 2.39| 30.83|
| L15   | 8.7   | 886      | 371   | 5.93 | 6.50      | 9.41    | 38.55| 2.76| 1.65| 2.75| 21.45|
| M16   | 8.7   | 810      | 277   | 8.16 | 7.32      | 10.58   | 54.33| 2.90| 1.49| 2.90| 30.23|

Appendix B

Table A2. Minerals used for each site in the A2M model. The mineralogy is based on previously developed mineral zones of Sweden. All sites additionally include Quartz (Qz), Water (Wa), Hematite (Hem), Rutile (Ru), and Gibbsite (Gibb).

| Site | Minerals 1 |
|------|------------|
| BD06 | Apa Bt Chl2 Ept Hbl II12 Or90 Ab100 An100 CPx Vrm1 |
| AC34 | Apa Cc Chl1 Chl2 Ept Hbl III2 Or100 Mus Ab100 An100 Vrm2 |
| AC04 | Apa Bt Chl2 Ept Hbl II12 Or90 Ab100 An100 CPx Vrm1 |
| AC35 | Apa Bt Chl2 Ept Hbl II12 Or90 Ab100 An100 CPx Vrm1 |
| Y07  | Apa Chl1 Chl2 Ept Hbl II12 Or100 Mus Ab100 An100 Vrm1 Vrm2 |
| U06  | Apa Chl1 Chl2 Ept Hbl II12 Or100 Mus Ab100 An100 Vrm1 Vrm2 |
| S22  | Apa Chl1 Chl2 Ept Hbl III1 Or100 Mus Ab100 An100 Vrm1 Vrm2 |
| S05  | Apa Chl1 Chl2 Ept Hbl III1 Or100 Mus Ab100 An100 Vrm1 Vrm2 |
| R09  | Apa Chl1 Chl2 Ept Hbl III1 Or100 Mus Ab100 An100 Vrm1 Vrm2 |
| O35  | Apa Chl1 Chl2 Ept Hbl III1 Or100 Mus Ab100 An100 Vrm1 Vrm2 |
| E21  | Apa Chl1 Chl2 Ept Hbl II11 Or100 Mus Ab100 An100 Vrm1 Vrm2 |
| P95  | Apa Chl1 Chl2 Ept Hbl III1 Or100 Mus Ab100 An100 Vrm1 Vrm2 |
| F22  | Apa Chl1 Chl2 Ept Hbl III1 Or100 Mus Ab100 An100 Vrm1 Vrm2 |
| F12  | Apa Chl1 Chl2 Ept Hbl III1 Or100 Mus Ab100 An100 Vrm1 Vrm2 |
| F23  | Apa Chl1 Chl2 Ept Hbl III1 Or100 Mus Ab100 An100 Vrm1 Vrm2 |
| F18  | Apa Chl1 Chl2 Ept Hbl III1 Or100 Mus Ab100 An100 Vrm1 Vrm2 |
| G22  | Apa Chl1 Chl2 Ept Hbl III1 Or100 Mus Ab100 An100 Vrm1 Vrm2 |
| N12  | Apa Chl1 Chl2 Ept Hbl III1 Or100 Mus Ab100 An100 Vrm1 Vrm2 |
| N13  | Apa Chl1 Chl2 Ept Hbl III1 Or100 Mus Ab100 An100 Vrm1 Vrm2 |
| H22  | Apa Chl1 Chl2 Ept Hbl III1 Or100 Mus Ab100 An100 Vrm1 Vrm2 |
Table A2. Cont.

| Site  | Minerals 1 |
|-------|------------|
| N19   | Apa Chl1 Chl2 Ept Hbl Ill1 Or100 Mus Ab100 An100 Vrm1 Vrm2 |
| K13   | Apa Chl1 Chl2 Ept Hbl Ill1 Or100 Mus Ab100 An100 Vrm1 Vrm2 |
| L18   | Apa Chl1 Chl2 Ept Hbl Ill1 Or100 Mus Ab100 An100 Vrm1 Vrm2 |
| L15   | Apa Chl1 Chl2 Ept Hbl Ill1 Or100 Mus Ab100 An100 Vrm1 Vrm2 |
| M16   | Apa Chl1 Chl2 Ept Hbl Ill1 Or100 Mus Ab100 An100 Vrm1 Vrm2 |

1 Abbreviations of minerals: Apa: Apatite; Bt: Biotite; Cc: Calcite; Chl1: Chlorite1; Chl2: Chlorite2; Ept: Epidote; Hbl: Hornblende; Ill1: Illite1; Ill2: Illite2; Or100: Orthoclase100; Or90: Orthoclase90; Mus: Muscovite; Ab100: Albite; An100: Anorthite; CPx: Clinopyroxene; Vrm1: Vermiculite1; Vrm2: Vermiculite2.

Table A3. Stoichiometry used for the normative mineralogy.

| Mineral          | Si | Ti | Al | Fe | Mg | Ca | Na | K  | P | H  |
|------------------|----|----|----|----|----|----|----|----|---|----|
| Albite           | 3  | 0  | 1  | 0  | 0  | 0  | 1  | 0  | 0 | 0  |
| Anorthite        | 2  | 0  | 2  | 0  | 0  | 1  | 0  | 0  | 0 | 0  |
| Apatite          | 0  | 0  | 0  | 0  | 0  | 5  | 0  | 3  | 1 |    |
| Biotite          | 218| 9  | 136| 90 | 90 | 0  | 4  | 68 | 0 | 162|
| Calcite          | 0  | 0  | 0  | 0  | 0  | 1  | 0  | 0  | 0 |    |
| Chlorite1        | 138| 1  | 124| 124| 107| 3  | 2  | 0  | 0 | 442|
| Chlorite2        | 87 | 1  | 100| 58 | 103| 0  | 0  | 0  | 0 | 302|
| Clinopyroxene    | 943| 7  | 53 | 107| 433| 390| 20 | 0  | 0 |    |
| Epidote          | 62 | 0  | 48 | 15 | 0  | 40 | 0  | 0  | 0 | 22 |
| Gibbsite         | 0  | 0  | 1  | 0  | 0  | 0  | 0  | 0  | 0 |    |
| Hematite         | 0  | 0  | 1  | 0  | 0  | 0  | 0  | 0  | 0 |    |
| Hornblende        | 10,100| 183 | 3600| 3000| 3500| 2767| 900| 300| 0 | 3133|
| Illite1          | 68 | 0  | 52 | 0  | 0  | 0  | 12 | 0  | 0 | 40 |
| Illite2          | 66 | 0  | 42 | 8  | 4  | 2  | 0  | 13 | 0 | 40 |
| Muscovite        | 60 | 1  | 48 | 6  | 4  | 0  | 1  | 22 | 0 | 47 |
| Orthoclase90     | 30 | 0  | 10 | 0  | 0  | 0  | 1  | 9  | 0 |    |
| Orthoclase100    | 3  | 0  | 1  | 0  | 0  | 0  | 0  | 1  | 0 |    |
| Quartz           | 1  | 0  | 0  | 0  | 0  | 0  | 0  | 0  | 0 |    |
| Rutile           | 0  | 1  | 0  | 0  | 0  | 0  | 0  | 0  | 0 |    |
| Vermiculite1     | 293| 0  | 162| 182| 103| 20 | 0  | 0  | 0 | 804|
| Vermiculite2     | 123| 0  | 68 | 22 | 103| 10 | 0  | 0  | 0 | 490|
| Water            | 0  | 0  | 0  | 0  | 0  | 0  | 0  | 0  | 1 |    |

Appendix C

For all sites used in this study except BD06, clay and silt were not separated in the grain size analysis. Other sites in the SWETHRO database and in other databases had separate analyses of clay and silt, and the average fraction from these soil samples was used for the sites with missing data (Table A4).

Table A4. Average proportions of clay in the clay+silt fraction in three different databases. The overall average was used in this study.

| Database            | Number of Sites | Site Names         | Average (%) | St dev |
|---------------------|-----------------|--------------------|--------------|--------|
| NORDSOIL            | 16              | See [41]           | 12           | 8      |
| SWETHRO             | 9               | L05, LO7, M10, BD06, O35B, M16B, P95B, P95C, P95D | 25           | 15     |
| Whole-tree harvesting experiments | 4            | Tömmersjöheden, Kosta, Lövilden, Lund | 9            | 7      |
| Total               | 29              |                    | 15           | 13     |

[41]. [21]. [42].
Appendix D

Table A5. Layer thickness and density.

| Site | Layer Thickness (cm) | Density (kg m$^{-3}$) |
|------|----------------------|----------------------|
|      | 1 2 3 4 5 1 2 3 4 5 |
| BD06 | 3.9 14.0 36.0 0.0 0.0 200 1378 1591 |
| AC34 | 8.0 10.0 11.0 29.0 0.0 130 890 917 1183 |
| AC04 | 2.3 8.0 9.0 20.0 13.0 106 1024 1116 1205 1519 |
| AC35 | 6.3 11.0 9.0 24.0 6.0 285 1659 1659 1878 1436 |
| Y07  | 1.3 4.0 11.0 25.0 10.0 213 903 891 915 1377 |
| U06  | 3.9 7.0 4.0 25.0 14.0 113 623 1212 1492 1386 |
| S22  | 1.9 10.0 15.0 25.0 0.0 135 834 817 978 |
| S05  | 5.5 12.0 20.0 15.0 3.0 382 1194 1526 1298 1340 |
| R09  | 1.8 6.0 14.0 30.0 0.0 149 901 1352 1513 |
| O35  | 6.4 9.0 7.0 23.0 11.0 172 1186 1202 935 1575 |
| E21  | 1.6 7.0 9.0 30.0 4.0 193 919 986 1313 1326 |
| P95  | 9.5 7.0 37.0 6.0 0.0 123 1267 1063 1420 1420 |
| F22  | 1.8 4.0 24.0 22.0 0.0 108 577 867 1129 |
| F12  | 1.5 8.0 15.0 12.0 10.0 103 658 1073 1538 1239 |
| F23  | 2.0 8.0 10.0 25.0 7.0 131 674 914 1165 1246 |
| F18  | 4.3 5.0 15.0 30.0 0.0 110 1079 1090 1500 |
| G22  | 2.9 5.0 9.0 26.0 10.0 163 1033 1548 1609 1609 |
| N12  | 4.4 5.0 9.0 29.0 7.0 143 606 779 1716 1716 |
| H03  | 6.4 10.0 10.0 20.0 11.0 111 1219 1162 1001 1676 |
| N13  | 4.6 9.0 11.0 19.0 11.0 139 893 910 1082 939 |
| H22  | 2.6 10.0 15.0 25.0 0.0 174 1053 764 1408 |
| N19  | 3.2 10.0 7.0 33.0 0.0 128 713 761 1382 |
| K13  | 4.0 5.0 10.0 35.0 0.0 225 515 886 1382 |
| L18  | 4.8 12.0 10.0 28.0 0.0 277 802 848 1144 1144 |
| L15  | 7.0 13.0 10.0 20.0 7.0 151 1363 1035 897 1171 |
| M16  | 4.1 4.0 10.0 36.0 0.0 195 758 1052 1068 |

Table A6. Specific surface area.

| Site | Specific Surface Area (m$^2$ m$^{-3}$) |
|------|---------------------------------------|
|      | 1 2 3 4 5 |
| BD06 | 28,151 1,059,207 687,921 |
| AC34 | 9598 495,895 471,949 554,705 |
| AC04 | 4821 314,861 334,683 411,358 459,973 |
| AC35 | 6887 256,904 477,979 300,161 438,813 |
| Y07  | 28,327 288,376 197,188 241,860 671,414 |
| U06  | 4302 176,673 391,887 564,446 418,842 |
| S22  | 23,270 315,461 350,423 476,259 |
| S05  | 12,082 545,612 845,384 643,632 773,341 |
| R09  | 10,883 110,442 224,527 261,998 |
| O35  | 6258 326,562 293,151 124,139 216,583 |
| E21  | 45,320 410,031 480,261 553,340 474,955 |
| P95  | 9760 605,526 553,612 451,694 |
| F22  | 3574 160,078 237,043 739,259 |
| F12  | 6175 222,081 218,479 362,367 401,651 |
| F23  | 13,343 245,158 333,636 510,119 515,197 |
| F18  | 4811 383,490 649,066 999,059 |
| G22  | 18,670 310,687 529,126 571,219 611,470 |
| N12  | 13,878 239,824 336,315 121,409 501,299 |
| H03  | 2661 336,315 121,409 501,299 |
| N13  | 7203 499,499 497,650 564,692 507,460 |
Table A6. Cont.

| Site | Specific Surface Area (m² m⁻³) |
|------|--------------------------------|
|      | 1          | 2          | 3          | 4          | 5          |
| H22  | 23,495     | 245,005    | 109,938    | 883,003    |            |
| N19  | 10,033     | 325,829    | 335,206    | 609,040    |            |
| K13  | 33,601     | 147,733    | 286,103    | 461,641    |            |
| L18  | 24,927     | 187,979    | 250,305    | 393,976    |            |
| L15  | 2825       | 101,456    | 523,967    | 68,315     | 75,974     |
| M16  | 13,648     | 203,506    | 227,709    | 169,613    |            |

References

1. Schöpp, W.; Posch, M.; Mylona, S.; Johansson, M. Long-term development of acid deposition (1880–2030) in sensitive freshwater regions in Europe. *Hydrol. Earth Syst. Sci.* 2003, 7, 436–446. [CrossRef]
2. Ferm, M.; Granat, L.; Engardt, M.; Karlsson, G.P.; Daniëls, H.; Karlsson, P.E.; Hansen, K. Wet deposition of ammonium, nitrate and non-sea-salt sulphate in Sweden 1955 through 2017. *Atmos. Environ. X* 2019, 2. [CrossRef]
3. Grünewald, U.; Reuss, J.O.; Johnson, D.W. Acid Deposition and the Acidification of Soils and Waters. Ecological Studies 1982–2008: Slow recovery and high sensitivity to sea-salt episodes. *Sci. Total Environ.* 2013, 444, 271–287. [CrossRef]
4. Pihl-Karlsson, G.; et al. The response of soil solution chemistry in European forests to decreasing acid deposition. *Glob. Chang. Biol.* 2018, 24, 3603–3619. [CrossRef]
5. Akselsson, C.; Hultberg, H.; Karlsson, P.E.; Pihl-Karlsson, G.; Hellsten, S. Acidification trends in south Swedish forest soils 1986–2008: Slow recovery and high sensitivity to sea-salt episodes. *Sci. Total Environ.* 2013, 444, 271–287. [CrossRef]
6. Pannatier, E.G.; Thimonier, A.; Schmitt, M.; Walthert, L.; Waldner, P. A decade of monitoring at Swiss Long-Term Forest Ecosystem Research (LFW) sites: Can we observe trends in atmospheric acid deposition and in soil solution acidity? *Environ. Monit. Assess.* 2011, 174, 3–30. [CrossRef] [PubMed]
7. Belyazid, S.; Westling, O.; Sverdrup, H. Modelling changes in forest soil chemistry at 16 Swedish coniferous forest sites following deposition reduction. *Environ. Pollut.* 2006, 144, 596–609. [CrossRef]
8. Nilsson, S.I.; Miller, H.G.; Miller, J.D. Forest Growth as a Possible Cause of Soil and Water Acidification: An Examination of the Concepts. *Oikos* 1982, 39, 40–49. [CrossRef]
9. Van Breemen, N.; Mulder, J.; Driscoll, C.T. Acidification and alkalinization of soils. *Plant. Soil* 1983, 75, 283–308. [CrossRef]
10. Riek, W.; Russ, A.; Martin, J. Soil acidification and nutrient sustainability of forest ecosystems in the northeastern German lowlands—Results of the national forest soil inventory. *Folia For. Pol. Ser. A* 2012, 54, 187–195.
11. Iwald, J.; Löfgren, S.; Stendahl, J.; Karltn, E. Acidifying effect of removal of tree stumps and logging residues as compared to atmospheric deposition. *For. Ecol. Manag.* 2013, 290, 49–58. [CrossRef]
12. Lucas, R.W.; Holmström, H.; Lämä, T. Intensive forest harvesting and pools of base cations in forest ecosystems: A modeling study using the Heureka decision support system. *For. Ecol. Manag.* 2014, 325, 26–36. [CrossRef]
13. Akselsson, C.; Belyazid, S. Critical biomass harvesting—Applying a new concept for Swedish forest soils. *For. Ecol. Manag.* 2018, 409, 67–73. [CrossRef]
14. Palviainen, M.; Finér, L. Estimation of nutrient removals in stem-only and whole-tree harvesting of Scots pine, Norway spruce, and birch stands with generalized nutrient equations. *Eur. J. For. Res.* 2012, 131, 945–964. [CrossRef]
15. Clarke, N.; Kiær, L.P.; Janne Kjønaas, O.; Bčrcena, T.G.; Vesterdal, L.; Stupak, I.; Finér, L.; Jacobson, S.; Armolaitis, K.; Lazdina, D.; et al. Effects of intensive biomass harvesting on forest soils in the Nordic countries and the UK: A meta-analysis. *For. Ecol. Manag.* 2021, 482, 118877. [CrossRef] [PubMed]
16. Reid, W.V.; Ali, M.K.; Field, C.B. The future of bioenergy. *Glob. Chang. Biol.* 2020, 26, 274–286. [CrossRef] [PubMed]
17. The Swedish Forest Agency’s Statistics Database. Available online: http://pxweb.skogsstyrelsen.se/pxweb/en/ (accessed on 9 February 2021).
18. Börjesson, P.; Hansson, J.; Berndes, G. Future demand for forest-based biomass for energy purposes in Sweden. *For. Ecol. Manag.* 2017, 383, 17–26. [CrossRef]
19. Sverdrup, H.; De Vries, W. Calculating critical loads for acidity with the simple mass balance method. *WaterAir Soil Pollut.* 1994, 72, 20. [CrossRef]
20. Fölster, J.; Bringmark, L.; Lundin, L. Temporal and Spatial Variations in Soil Water Chemistry at Three Acid Forest Sites. *WaterAir Soil Pollut.* 2003, 146, 171–195. [CrossRef]
21. Pihl Karlsson, G.; Akselsson, C.; Hellsten, S.; Karlsson, P.E. Reduced European emissions of S and N—Effects on air concentrations, deposition and soil water chemistry in Swedish forests. *Environ. Pollut.* 2011, 159, 3571–3582. [CrossRef]
22. Warfvinge, P.; Sverdrup, H. Critical Load of Acidity to Swedish Forest Soils. *Methods, Data and Results;* Lund University, Department of Chemical Engineering II: Scania, Sweden, 1995.
23. Karlsson, P.E.; Pihl Karlsson, G.; Hellsten, S.; Akselsson, C.; Ferm, M.; Hultberg, H. Total deposition of inorganic nitrogen to Norway spruce forests—Applying a surrogate surface method across a deposition gradient in Sweden. Atmos. Environ. 2019, 217. [CrossRef]

24. Posch, M.; de Smet, P.; Hettelingh, J.-P.; Downing, R. Calculation and Mapping of Critical Thresholds in Europe. Status Report 1995; Coordination Center of Effects, Rijksinstituut voor Volksgezondheid en Milieu RIVM: Bilthoven, The Netherlands, 1995.

25. Galloway, J.N. Acid deposition: Perspectives in time and space. Water Air Soil Pollut. 1995, 85, 15–24. [CrossRef]

26. Sverdrup, H.; Warfvinge, P. Calculating field weathering rates using a mechanistic geochemical model PROFILE. Appl. Geochem. 1993, 8, 273–283. [CrossRef]

27. Posch, M.; Kurz, D. A2M—A program to compute all possible mineral modes from geochemical analyses. Comput. Geosci. 2007, 33, 563–572. [CrossRef]

28. Persson, C.; Roos, E.; Lövblad, G. Meso-Scale Modelling of Base Cation Deposition in Sweden; Springer: Boston, MA, USA, 2000; pp. 117–125. [CrossRef]

29. Lövblad, G.; Persson, C.; Klein, T.; Ruoho-Airola, T.; Hovmand, M.; Tarasson, L.; Torseth, K.; Moldan, F.; Larssen, T.; Rapp, L. The Deposition of Base Cations in the Nordic Countries; IVL Swedish Environmental Research Institute: Stockholm, Sweden, 2004.

30. Futter, M.N.; Ring, E.; Högbom, L.; Entenmann, S.; Bishop, K.H. Consequences of nitrate leaching following stem-only harvesting of Swedish forests are dependent on spatial scale. Environ. Pollut. 2010, 158, 3552–3559. [CrossRef]

31. Akselsson, C.; Olsson, J.; Belyazid, S.; Capell, R. Can increased weathering rates due to future warming compensate for base cation losses following whole-tree harvesting in spruce forests? Biogeochemistry 2016, 128, 89–105. [CrossRef]

32. Swedish Forest Agency. Skogliga Konsekvensanalys 2008: SKA-VB 08; Swedish Forest Agency: Jönköping, Sweden, 2008.

33. Sverdrup, H.; Warfvinge, P.; Rabenhorst, M.; Janicki, A.; Morgan, R.; Bowman, M. Critical loads and steady-state chemistry for streams in the state of Maryland. Environ. Pollut. 1992, 77, 195–203. [CrossRef]

34. Braun, S.; Tresch, S.; Augustin, S. Soil solution in Swiss forest stands: A 20 year’s time series. PLoS ONE 2020, 15, e0227530. [CrossRef]

35. Pihl Karlsson, G.; Hellsten, S.; Akselsson, C.; Karlsson, P.E. Försurning och Övergödning i Skåne län Resultat från Krondroppsnätet till och med 2018/19; IVL Swedish Environmental Research Institute: Stockholm, Sweden, 2020.

36. Ågren, A.; Löfgren, S. pH sensitivity of Swedish forest streams related to catchment characteristics and geographical location—Implications for forest bioenergy harvest and ash return. For. Ecol. Manag. 2012, 276, 10–23. [CrossRef]

37. Jonard, M.; Fürst, A.; Verstraeten, A.; Thimonier, A.; Timmermann, V.; Potočić, N.; Waldner, P.; Benham, S.; Hansen, K.; Merilä, P; et al. Tree mineral nutrition is deteriorating in Europe. Glob. Chang. Biol. 2015, 21, 418–430. [CrossRef]

38. Jonard, M.; Legout, A.; Nicolas, M.; Dambrine, E.; Nys, C.; Ulrich, E.; van der Perre, R.; Ponette, Q. Deterioration of Norway spruce vitality despite a sharp decline in acid deposition: A long-term integrated perspective. Glob. Chang. Biol. 2012, 18, 711–725. [CrossRef]

39. Schulze, E.D. Air Pollution and Forest Decline in a Spruce (Picea abies) Forest. Science 1989, 244, 776–783. [CrossRef] [PubMed]

40. De Jong, J.; Akselsson, C.; Egnell, G.; Löfgren, S.; Olsson, B.A. Realizing the energy potential of forest biomass in Sweden—How much is environmentally sustainable? For. Ecol. Manag. 2017, 383, 3–16. [CrossRef]

41. Stendahl, J.; Akselsson, C.; Melkerud, P.-A.; Belyazid, S. Pedon-scale silicate weathering: Comparison of the PROFILE model and the depletion method at 16 forest sites in Sweden. Geoderma 2013, 211–212, 65–74. [CrossRef]

42. Brandtberg, P.-O.; Olsson, B.A. Changes in the effects of whole-tree harvesting on soil chemistry during 10years of stand development. For. Ecol. Manag. 2012, 277, 150–162. [CrossRef]