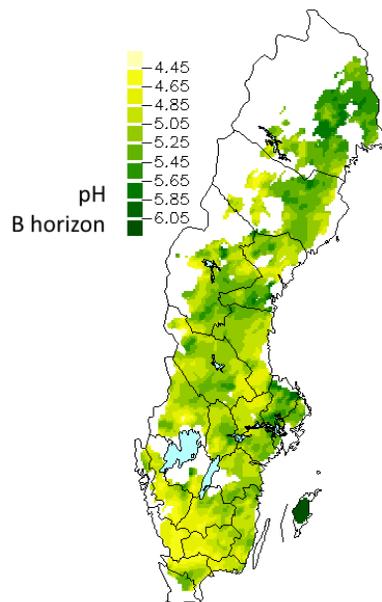# Acidification of Swedish Forest Soils

Evaluation of Data from the Swedish Forest Soil Inventory

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Licentiate Thesis
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Cover: Map of Sweden showing pH in the B horizon of forest soils.
    Source: The Swedish Forest Soil Inventory
    (http://www-markinfo.slu.se/sve/kem/cnph/phbgril1.html)

Acidification of Swedish Forest Soils
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Abstract

Acidification of forest soils in Sweden has negatively affected nutrient status and caused toxic effects on biota. The aim of this thesis was to investigate potential impacts of two drivers of acidification of forest soils in Sweden: (1) tree growth with subsequent harvest and (2) deposition of sulphur and nitrogen compounds.

The magnitude of acidification by tree growth was calculated using estimates of stem wood, logging residue and stump harvesting from a national forestry impact assessment and data of cations and anions in different tree parts. Acidification by tree growth was defined as the net cation uptake.

The magnitude of acidification by deposition was estimated based on deposition data from four catchments throughout Sweden. Acidification by deposition was defined as the negative value of the acid neutralizing capacity in deposition.

The acidifying effect of tree harvesting decreases in the order Norway spruce>birch>Scots pine. Harvesting of logging residues causes more soil acidification than harvesting of stumps, despite the fact that the biomass of stumps is larger, due to the higher contents of base cations in needles and branches than in stem wood. Harvesting of Scots pine and birch causes soil acidification at a magnitude equal to recent deposition levels while harvesting of Norway spruce causes more acidification than what is caused by deposition.

To study the acidifying effects on the soil of tree growth during a forest generation and deposition, data from the Swedish Forest Soil Inventory – a large-scale inventory throughout Sweden – from the O (n=1990), B (n=1902) and C (n=1477) horizons were used. Four variables were chosen as indicators of soil acidity: pH-H_2O (pH), base saturation (BS), exchangeable calcium (Ca) and exchangeable aluminium (Al) from each horizon. The variables were analysed with respect to tree age and data of sulphur deposition, using linear and multiple regression. For tree age, a relative age concept was used to account for different tree growth rates in different parts of Sweden.

Tree growth during a forest generation and deposition of acidifying substances both affect acidity related variables in Swedish forest soils. Tree growth causes decreased pH, BS and Ca and increased Al in the O and to some extent the B horizon in the beginning of the trees' life cycle. Deposition causes decreased pH, BS and Ca and increased Al in the O, B and C horizons throughout the deposition gradient in Sweden. The effect of deposition is most evidently expressed in deep soil layers where biological acidification have less influence.

Keywords: soil acidification, tree growth during a forest generation, deposition, Sweden

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Dedication

To my family

Det är skönare lyss till en sträng, som brast, än att aldrig spänna en båge.
  Verner von Heidenstam

Contents

List of Publications

This thesis is based on the work contained in the following papers, referred to by Roman numerals in the text:

I   Iwald, J., Löfgren, S., Stendahl, J. & Karltun, E. (2013). Acidifying effect of removal of tree stumps and logging residues as compared to atmospheric deposition. *Forest Ecology and Management,* 290, pp. 49-58.

II Iwald, J., Stendahl, J., Löfgren, S., von Brömssen, C. & Karltun, E. (2016). Spatial variations of acid-base properties in Swedish forest soils – the impact of tree growth, temperature and atmospheric deposition. *Manuscript*.

Paper I is reproduced with the permission of the publishers.

The contribution of Johan Iwald to the papers included in this thesis was as follows:

I   Planned the study together with the co-authors, performed data analysis and wrote the manuscript with assistance from the co-authors.

II   Planned the study together with the co-authors, performed data analysis and wrote the manuscript together with the co-authors.

Abbreviations

ANC   Acid neutralizing capacity
BS   Base saturation
EMEP   European Monitoring and Evaluation Programme
lnAl   ln(exchangeable aluminium)
lnCa   ln(exchangeable calcium)
RA   Relative tree age
SDep   Sulphur deposition
SFSI   Swedish Forest Soil Inventory
TSum   Temperature sum

1 Introduction

1.1 Acidification of forest soils

Acidification of forest soils, a natural process as well as one of the major environmental problems in Europe and several other parts of the world, can be defined as "a permanent change in the acid neutralizing capacity (ANC) of the inorganic soil fraction (ΔANC<0) resulting from an irreversible H^+ flux. This irreversible H^+ flux can be caused either by direct proton addition, by different mobility of components of the ANC or by a permanent change in redox conditions." (van Breemen *et al.*, 1983). Acidification of forest soils occurs, mainly, in two ways: by trees as they grow and by emissions of sulphuric and nitric oxides (SO_2 and NO_x) that are converted to acids in the atmosphere and brought to forests as either dry or wet deposition. Acidification by tree growth is caused by the excess uptake by trees of cations over anions and the consequent release of hydrogen ions (van Breemen *et al.*, 1983; Nilsson *et al.*, 1982). Acidification by deposition is caused mainly by 1) combustion of fossil fuels leading to emissions of sulphuric (SO_2) and nitric (NO_x) oxides and 2) use of nitrogen-based fertilizer in agriculture leading to ammonia (NH_3) emissions (Rice & Herman, 2012). In addition to acidification by tree growth and acidification by deposition, some forest soils can be acidified when they are drained and transferred from a reduced to an oxidized state (van Breemen *et al.*, 1983). Acidification of forest soils by deposition has led to nutrient depletion and aluminium (Al) toxicity for primarily aquatic organisms over large areas in Europe and North America (Driscoll *et al.*, 2001).

1.2 Processes and trends related to biological acidification and forest growth

Acidification by tree growth is a natural phenomenon, caused by the fact that tree roots take up a higher charge equivalent of cations (calcium Ca^{2+}, magnesium Mg^{2+}, potassium K^+ and sodium Na^+) than anions (dihydrogen phosphate $H_2PO_4^-$, sulphate SO_4^{2-} and chloride Cl^-). The excess cation uptake is compensated for by a return of hydrogen ions (H^+) from the tree to the soil. Tree growth acidifies mainly the upper soil layers, i.e. the root zone, in contrast to deposition that acidifies the whole soil profile due to the presence of mobile anions (van Breemen *et al.*, 1983; Nilsson *et al.*, 1982). Acidification by tree growth has been observed by a number of authors: Brand *et al.* (1986) found a decrease in pH between the years 1938 to 1984 in soils on abandoned farmland afforested with pine and spruce in Ontario, Canada, and Tamm and Hallbäcken

(1986) found a decrease in pH between the years 1927 to 1982/84 in soils with deciduous and coniferous forest in southwestern Sweden. The latter authors found pH decrease in all soil horizons (A, B and C) and explained pH decrease in the A horizon by tree growth and pH decrease in the B and C horizons by acid deposition. A decrease in extractable calcium ions (Ca^{2+}) – the main base cation on the exchange complex that is buffering soil acidity – from the 1930s until 1984 in forest soils in the Adirondacks, NY, has also been observed (Johnson *et al.*, 1994); this was attributed to Ca^{2+} uptake by trees.

1.3 Processes and trends related to acidification by deposition

With industrial expansion and increased use of fossil fuels after World War II, emissions of sulphur (S) and nitrogen (N), and consequent deposition, largely increased. In Europe, S deposition reached its peak in the 1970s and N deposition around 1990 (Schöpp *et al.*, 2003). During the period 1990-2014, however, emissions in Europe decreased by 84% for SO_x, 55% for NO_x and 27% for NH_3 according to the European Monitoring and Evaluation Programme database (EMEP, 2016). In North America, the development has been similar: precipitation in eastern US became increasingly more acidic from the 1950s to the 1970s (Likens & Butler, 1981). From 1990 until 2013, total S deposition in eastern US decreased by 66% and N deposition decreased by 24% (Beachley *et al.*, 2016). However, while emissions of S and N are on the decline in Europe and North America, they have increased since 2000 in other parts of the world like Asia and Africa (Vet *et al.*, 2014).

Concerning acidification by deposition in Sweden, the development has been similar to Europe as a whole. In Sweden, a country that receives most of its deposition from emissions generated outside its borders, pH values in precipitation gradually decreased after World War II (Odén, 1968), which become known to the general public through a newspaper article in 1967 (Odén, 1967). After the European countries agreed upon the Convention on Long-Range Transboundary Air Pollution (CLRTAP) (UNECE, 1979), S emissions were cut down considerably while N emissions did not decrease as much. In Sweden from 1990 until 2014, emissions decreased by 77% for SO_x, 50% for NO_x and 2% for NH_3 (EMEP, 2016). According to a study of deposition and throughfall at more than 50 sites across Sweden (Karlsson *et al.*, 2011), non-marine SO_4 deposition decreased by 22-67 % between the time periods 1996-1999 and 2005-2008; the largest decreases were found in the southwestern part of the country that had the highest deposition pressure. There was, however, no general trend concerning changes in N deposition between the two periods.

When the strong acids H_2SO_4 and HNO_3, created from SO_2 and NO_x emissions, reach the ground as wet deposition, they can leach through the soil profile; the hydrogen ions can exchange with base cations Ca^{2+}, Mg^{2+}, K^+ and Na^+ and increase the solubility of aluminium, an element that is toxic to many plants and animals (Sparling & Lowe, 1996). As a result, deposition has caused lower pH and base cation supplies as well as increased amounts of inorganic Al in forest soils (Driscoll *et al.*, 2001; Likens *et al.*, 1996; Falkengren-Grerup, 1987). Falkengren *et al.* (1987) found decreases of exchangeable Na, K, Mg, Ca, zinc (Zn) and manganese (Mn) in soils in southern Sweden during 1949-1985 that were attributed to acidic deposition. The fact that these losses of exchangeable cations occurred down to a soil depth of 1 meter, below the rooting depth of trees and plants, indicated that they could not have been caused by root uptake.

Decreases of base cation concentrations in soil solution due to deposition have been reported in several studies: for example, in Norway during 1986-1997 (Moffat *et al.*, 2002), in Switzerland during 1987-1997 (Blaser *et al.*, 1999), in Sweden during 1990-1994 (Fölster *et al.*, 2003) and in the Czech republic during 1994-2005 (Oulehle *et al.*, 2006). In the Norwegian study base cation concentrations increased in the soil organic layer, indicating a partial recovery from acidification, while they decreased at 15 and 40 cm depth. In the Swedish study results could be interpreted both as continued acidification and beginning of recovery, with decrease in base cation but also in SO_4 concentrations, leading to increased pH and ANC at some sites. A weaker decline in SO_4 concentrations in soil solution than in SO_4 deposition indicated that SO_4 desorption was a factor delaying recovery. Sulphate desorption was also mentioned as a key factor controlling recovery in the Czech study, along with decrease in Ca deposition. In that study, pH in soil solution at 30 cm depth remained unchanged – which may be attributable to SO_4 desorption – while Ca as well as Al concentrations decreased.

In a study of soil water chemistry at 67 forest sites in Sweden during 1996-2008 (Karlsson *et al.*, 2011) there were decreases in SO_4-S (attributed to reduced deposition) and decreases in Ca (attributed to decreased anion flow). The pH and ANC values increased and Al decreased on several sites, indicating recovery from acidification, but there were also sites with opposite trends. Overall the findings suggested recovery from acidification, albeit a recovery that was slow and variable between sites. There was a clear gradient in ANC of the soil solution following the gradient in S deposition throughout Sweden: soils in the southwest had negative ANC values during 2005-2008 indicating that they were still acidified and only in the beginning of the recovery process, soils in the southeast often had ANC around zero and soils in northern Sweden had mostly positive

ANC and high pH, indicating that they had been little affected by deposition. Another study of soil solution chemistry, covering nine sites in southwest Sweden during the period 1986-2008 (Akselsson *et al.*, 2013), revealed that some recovery had occurred in terms of increased pH and decreased Al but also that Ca levels had decreased. The decrease in Ca was explained by reduced net charge of the soil solution, leading to reduced release of positive ions from the exchangeable pool. According to the Gaines-Thomas cation exchange equilibrium (Gaines & Thomas, 1953) a decrease in ionic strength, as when deposition goes down, leads to a higher ratio of cations with high valence (e.g. Al^{3+} and Ca^{2+}) on soil exchange sites and a lower ratio of these cations in soil solution. The sites in the study by Akselsson *et al.* (2013) were sensitive to transient sea salt episodes that could cause re-acidification of the soil solution due to large input of the mobile Cl^- ion.

A gradient in soil pH, similar to the ANC gradient found by Karlsson *et al.*, (2011), was found in a study of data for the years 1993-1998 from a large-scale forest soil inventory throughout Sweden (Karltun *et al.*, 2003). When pH was plotted with respect to latitude, there were positive correlations between pH in the B and C horizons and latitude; the correlation was most significant in the C horizon. In the O horizon there was no correlation between pH and latitude. The authors concluded that the positive correlations in the B and C horizons, with lower pH values in southern Sweden and higher values in the north, were caused by the deposition gradient throughout Sweden with deposition being highest in the south and decreasing northward. The acidifying effect of deposition had been most pronounced in the deep soil layers, where the effect of biological activity is small. While some recovery from acidification has been observed in soil solution (see the previous paragraphs), not much recovery has been detected in the solid, exchangeable phase of Swedish forest soils. However, a study of 27 sites in northeast US and southeast Canada (Lawrence *et al.*, 2015) showed some recovery from acidification by deposition in the exchangeable phase from the 1980s/1990s to the 2000s; there were increases of pH, Ca and base saturation and decrease of Al in the O horizons, but decrease of Ca and base saturation and increase of Al in the B horizons. The deposition levels at the American sites (4-10 kg S ha^{-1} yr^{-1}) were similar to deposition levels in northern Sweden (2-10 kg S ha^{-1} yr^{-1}).

Acidification of soils and waters are intimately connected. After leaching through the soil profile the mobile anions SO_4^{2-} and nitrate (NO_3^-) continue out into surface waters, thereby lowering the ANC of streams and lakes (Driscoll *et al.*, 2001). When S and N emissions increased after World War II, it was initially the soils that were acidified; acidification of waters began only when base saturation in soils had decreased considerably (Sverdrup *et al.*, 2005).

Conversely, when S emissions declined from the 1970s the recovery of surface waters began first, before the recovery of soils. According to Löfgren *et al.* (2011) there is a difference between recharge and discharge areas in catchments; during the historic period of high SO_4^{2-} deposition it was the discharge areas, not the recharge areas, that buffered against acidification into surface waters, due to their high organic matter content and cation exchange capacity as well as their ability to accumulate aluminium. When deposition has been declining, on the other hand, it has been the discharge areas that buffer against recovery.

From the 1990s, recovery of streams and lakes has been observed in Sweden (Fölster & Wilander, 2002) as well as in other parts of Europe and North America (Davies *et al.*, 2005; Stoddard *et al.*, 1999). A study of 36 Swedish lakes during 1987-2012 (Futter *et al.*, 2014) revealed slow recovery: strong acid anions declined and chloride, derived from sea salt, was often the dominant anion at the end of the period. Base cations also declined but less rapidly, leading to increased ANC of lake water. The decline in SO_4^{2-} deposition led to increased concentrations of dissolved organic carbon (DOC) and in the end of the period, organic acidity was often comparable with inorganic acidity. According to another study of four catchments in different parts of Sweden during 1996-2009 (Löfgren *et al.*, 2011), there were increases in ANC and decreases in ionic strength in stream water in some places, explained by reductions in SO_4^{2-} deposition. There were reductions of inorganic Al in stream water in some of the catchments, which was attributed by the authors to increased pH in soil water and groundwater. In soil water SO_4^{2-} concentrations declined, ANC and pH increased at the catchments in southern Sweden, ionic strength decreased at some of the sites and there were some reductions of inorganic Al. There was a difference between the catchment in northern Sweden and the three catchments in the south in that the northern catchment was much less acidified – but on the other hand, the recovery rate was slower in the north. ANC values were generally negative in southern Sweden while they were positive in the north. In the southern catchments, desorption of sulphate and mineralization of organic S may have affected recovery from acidification. In the catchment near the western coast of Sweden, chloride deposition originating from sea salt partly compensated for the decrease in sulphate deposition and contributed to maintaining high ionic strength levels.

1.3.1 The role of nitrogen deposition

While human-induced S emissions occur as sulphuric oxides (SO_2), N emissions can occur either as nitrous oxides (NO_x) originating mainly from fossil fuel burning (Rice & Herman, 2012) or as ammonia (NH_3) originating mainly from agriculture and animal husbandry (Boxman *et al.*, 2008). In the atmosphere,

nitrous oxides can be converted to nitric acid (HNO_3) that acidifies the soil when deposited while ammonia can be protonated to ammonium (NH_4^+), a process that neutralizes acidity in the atmosphere. In the soil, however, an NH_4^+ ion can either be taken up by a tree root, whereby a hydrogen ion is released in return, or converted to NO_3^- through nitrification, whereby two hydrogen ions are produced. Normally, the uptake of the NO_3^- ion by the tree neutralizes one of the H^+ that was formed through nitrification. Thus, deposition of NH_4^+ and HNO_3 both result in formation of a stoichiometrically equivalent amount of H^+ in the soil. Therefore, N deposition is acidifying both when it occurs as HNO_3 and NH_4^+.

Tree uptake of NH_4^+ is acidifying while uptake of NO_3^- counteracts acidification. As trees take up nitrogen both as NH_4^+ and NO_3^-, the effect on soil acidity may be small. In N-limited forests, all N is generally recycled well within the system and no N leaching occurs. A small portion of the NO_3^-, though, is converted to N_2O or N_2 through denitrification, particularly in wet soils (Klemedtsson *et al.*, 1997). According to a study of soil water, groundwater and stream water in four catchments throughout Sweden during 1996-2009 (Löfgren *et al.*, 2011), inorganic N deposition was almost completely retained within soils and living biomass or lost to the atmosphere. In areas receiving high N deposition, however, ecosystems may not be able to retain all N. A study of Swedish forest soils (Akselsson *et al.*, 2010) revealed that there is a risk for N leaching in the south of Sweden, where deposition has been high. Clear-cutting may also induce nitrogen leaching. A review of the effects of air pollution and forest management on N leaching from temperate forests (Gundersen *et al.*, 2006) concluded that there are three main pathways to N leaching: increased N input (e.g. through deposition), reduced plant uptake (e.g. after logging) and enhanced N mineralization (due to liming, ditching and climate change). N leaching occurs when N deposition and N mineralization exceed plant demand. The elevated N deposition levels in Europe explain about half of the variability in N leaching. After clear-cut, NO_3^- losses peak after 2-3 years and return to pre-cut levels after 3-5 years. At a site in the Czech Republic that had been subject to high deposition (Oulehle *et al.*, 2006), leaching of N ceased in 2003-2005; a reason was thought to be that when S deposition, and subsequent Al concentrations in soils, were reduced, more N could be retained in the ecosystem through biotic immobilization by microbes and fungi.

1.4 Future trends in soils and waters

Whereas deposition of S and N are now on the decline, forest growth and harvest have increased in Sweden during the post-World War II years. In the time period

1956-2009, forest growth increased from $80 \cdot 10^6$ m^3 to $120 \cdot 10^6$ m^3 and forest harvest increased from $50 \cdot 10^6$ m^3 to $90 \cdot 10^6$ m^3 (Christiansen, 2013). In addition, the use of logging residues (branches, tree tops and needles) as biofuel has increased a lot since the 1980s and during the last ten years, also stumps have been harvested as biofuel (Christiansen, 2013). Nitrogen deposition, that enhances tree growth, can contribute to soil acidification indirectly through increased tree growth and uptake of base cations, in addition to the direct effect on soil chemistry. In a modelling study of 16 coniferous forest sites in Sweden (Belyazid *et al.*, 2006), it was predicted that high N deposition in the future will be one reason for continued acidification of forest soils, due to its enhancement of tree growth. Increased tree growth and more intensive harvesting can also affect open water chemistry: in a modelling study of 3000 Swedish lakes (Moldan *et al.*, 2013) the acidification peak was around 1985 with widespread recovery afterwards, but future reductions in soil base cation pools due to intensive tree harvesting could lead to re-acidification of sensitive lakes. A modelling study (Zetterberg *et al.*, 2014) concluded that whole-tree harvesting could lead to large depletion of soil Ca^{2+} supply and a negative trend for stream-water ANC. According to another study (Löfgren *et al.*, 2016), however, the impact of whole-tree harvesting is expected to be most pronounced in the recharge areas; the pH effect on streams is expected to be small due to high buffering capacity of riparian zones and low pH-sensitivity of headwater streams.

With the contradicting trends concerning deposition and tree growth in mind, a potential development in the future is that forest soils in Sweden will be less acidified by deposition while they are becoming increasingly acidified due to biomass growth and extraction. With this in regard, there is a need to evaluate the extent of forest soil acidification in Sweden; the magnitude and effects of different kinds of acidification and possible trends for the future. This is the scope of this thesis.

2 Aim

The main objective of this thesis was to investigate the effects of tree growth and deposition on acidification of forest soils in Sweden. Most other studies on forest soil acidification have used a limited number of sites. This study, in contrast, encompasses a large data set spanning over a broad range of Swedish soil and forest types.

The specific objectives of this thesis were to investigate the following questions:

What is the magnitude of acidification by tree harvesting and by deposition respectively, in Sweden? (Paper I)

What is the effect of tree growth during a forest generation and deposition, respectively, on the acid-base status of forest soils in Sweden? (Paper II)

In Paper II, the following hypotheses were tested:

In *soils acidified by tree growth* the excess uptake of cations over anions will result in a net H^+ input to the soil, resulting in increased soil acidity in the soil layers from which the nutrient uptake takes place, *i.e.* primarily the O horizon and the upper part of the mineral soil, but the acidity will not penetrate to deeper layers since no mobile anions are released together with the acidity.

In *soils acidified by deposition of sulphur and nitrogen*, acidity is percolated through the O horizon into the mineral soil due to the availability of mobile anions, gradually exhausting the buffer systems. In areas receiving high S and N deposition, also the deeper soil layers will be acidified.

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3 Materials and methods

3.1 Calculation of acidification by tree growth and harvest (Paper I)

The area studied is the country of Sweden and the trees considered are Scots pine (*Pinus sylvestris* L.), Norway spruce (*Picea abies* (L.) Karst.) and birch (*Betula spp.*). For estimates concerning logging, including logging residue and stump harvesting, results from a national forestry impact assessment were used (Claesson, 2008). In this report, the state of the Swedish forests during the period 2010–2109 is predicted based on the state in 2004 as described by the Swedish National Forest Inventory (SLU, 2012). Several different scenarios concerning harvesting of biomass for energy (logging residues and stumps) are described in the report; in the scenario used in this study, 27% of the logged areas are exempted from slash and stump harvesting due to ecological, technical and/or economic restrictions; where harvested, 40% of logging residues and stumps are left on the ground and no birch stumps are harvested. In the present study, acidification by tree harvesting is defined as net cation uptake (charge equivalents) in harvested tree parts (stem wood, logging residues and stumps) adapted from van Breemen *et al.* (1983):

$$(Ca^{2+} + Mg^{2+} + K^+ + Na^+) - (H_2PO_4^- + SO_4^{2-} + Cl^-)$$

Calculations of biological acidification caused by extraction of tree biomass were first made for each of the 21 different counties in Sweden. This was done using county-level data of standing volume for each tree species (SLU, 2012), projections of yearly logging, including harvesting of logging residues and stumps, for each species during 2010-2109 (Claesson, 2008) and data of nutrient contents in various tree parts such as stem wood, bark, branches and needles (Eriksson (pers. comm.), 2012; Werkelin *et al.*, 2010; Werkelin, 2006; Werkelin *et al.*, 2005; Hägermark *et al.*, 2002; Alriksson & Eriksson, 1998; Nilsson & Wiklund, 1995; Eriksson & Rosen, 1994; Nilsson & Wiklund, 1994; Mälkönen & Saarsalmi, 1982; Mälkönen, 1978; Björkroth & Rosén, 1977). County-level data were then merged into data for three regions, representing a gradient in deposition: southwest Sweden, mid & southeast Sweden, and northern Sweden (Figure 1). Calculation of acidification by tree harvesting was made per unit area but for logging residues and stumps, it was also made per unit heating value of these tree parts as biofuel, using the relation that 1000 kg dry matter gives a heating value of 4.9 MWh (Claesson, 2008).
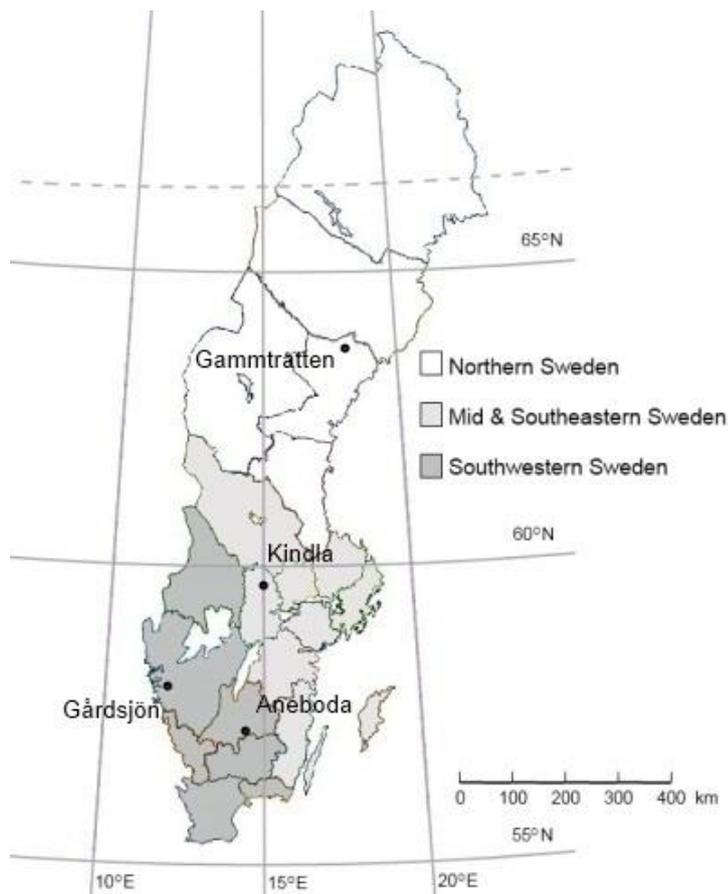
Figure 1 Map of Sweden (Maps of World, 2012; Wikipedia, 2012) with
three regions "Southwest", "Mid & Southeast" and "North". The names
on the map are the monitoring sites from where acid deposition data were
obtained.

3.2 Estimation of acidification by deposition (Paper I)

Estimation of acidification by deposition was made, separately for each of the
three regions Southwest, Mid & Southeast and North, using deposition data from
four monitoring sites located in different parts of Sweden (Löfgren *et al.*, 2011)
(Figure 1). For each of these sites, annual deposition data for anions Cl^-, NO_3^-
and SO_4^{2-} and cations Ca^{2+}, Mg^{2+}, K^+, Na^+ and NH_4^+ were available for the years

1996-2009. Acidification by deposition was defined as the negative value of ANC in deposition. ANC in this study is defined as (Cosby *et al.*, 2001):

$$(Ca^{2+} + Mg^{2+} + K^+ + Na^+ + NH_4^+) - (SO_4^{2-} + NO_3^- + Cl^-)$$

For each of the four sites, a range of acidification by deposition (lowest to highest value) was obtained for the period 1996-2009. To compare with biological acidification in the three regions Southwest, Mid & Southeast and North, we used the range of acidification by deposition in the combinations of sites Aneboda–Gårdsjön for Southwest, Kindla–Aneboda for Mid & Southeast and Kindla–Gammtratten for North.

3.3 Selection of soil data for analysis (Paper II)

In this study, soil data from the Swedish Forest Soil Inventory (SFSI) (SLU, 2016) were used. The SFSI collects information on soil conditions and soil properties from ca 20000 objectively located sampling plots distributed across the country. All land types, except agricultural land, urban land and high mountain areas are included. The SFSI started in 1983 and all plots were visited during the first inventory period 1983-1987. After that, one tenth of the plots were revisited each year during the inventory periods 1993-2002 and 2003-2012. A fourth inventory period 2013-2022 is underway. During the inventory, humus samples were taken from ca 50 % of the plots and mineral soil samples from ca 25% of the plots.

Soil data from the second inventory period 1993-2002 were used. A systematic, grid-based sample plot design was used; a selected sample of plots from the inventory can be considered an un-biased sample of the strata selected for study. A stratification of the dataset based on soil parent material type and texture was made: only till soils and soils developed in poorly sorted sediments were included while rocky and clayey soils, as well as peat soils, were omitted from the data set. Data from calcareous soils (pH-CaCl$_2$ > 6.5 in the C horizon; 1% of all soil samples) were also omitted due to their high acid neutralizing capacity. Data on soil properties from the O, B and C horizons were analysed. The O horizon is sampled as the entire mor or moder layer, the B horizon as the top 5 cm of the B horizon directly underlying an E or A horizon and the C horizon at 55-65 cm below the mineral soil surface. After selection, there were 1990, 1902 and 1477 plots from the O, B and C horizons, respectively, representing 55% of the Swedish forest area and 64% of all forest land with mineral soils. Omitted categories represent 27% (highly sorted sediments), 8% (stony), 3% (clayey) and 8% (peat soils). The lower amount of plots with B and

C horizon samples reflects that limited soil depth sometimes prevent these layers from being sampled.

3.4 Analysis of soil acidity variables versus tree growth and deposition (Paper II)

Four soil chemical variables were used as indicators of soil acidity: pH-H_2O (pH), effective base saturation (BS), exchangeable calcium extracted in 1M NH_4Ac at pH=7 (Ca) and exchangeable aluminium extracted in 1M KCl (Al). Two of these, pH and BS, are so called intensity variables less prone to vary with soil properties like texture and cation exchange capacity. The other two, Ca and Al, are capacity variables and they may vary considerably depending on various soil properties. Ca and Al were chosen because they are the major exchangeable cations in Swedish forest soils.

To investigate acidification by tree growth during a forest generation, the acidity variables pH, BS, Ca and Al were analysed, using linear regression, with respect to variation with tree age. Relative age (RA) was used in order to avoid differences between different parts of Sweden due to the climate gradient and subsequent differences in tree growth. RA is defined by Georgiadis (2011) as

$$RA = \frac{stand\ age}{least\ cutting\ age * 1.1}$$

where least cutting age is defined as the lowest age at which the stand could legally be clear-felled (Swedish Forest Agency, 2011) and ranges from 45 years on fertile soils in the south of Sweden up to 100 years on poor soils in the north. Analysis using RA ensures a more equal comparison with respect to actual stand development and avoids biased representation of geographical areas in certain age groups.

To investigate acidification by deposition, linear regression of the acidity variables was done with respect to sulphur deposition data (SDep) from EMEP (2013). The geographical coordinates of the SFSI plots were matched with corresponding EMEP coordinates. For each SFSI plot, SDep according to EMEP during the period 1980-1992 (annual average), immediately preceding the soil sampling period 1993-2002, was used as an estimate of acidification by deposition. As acidification by deposition is a gradual process, causing soil depletion of base cations, increase of S and N and mobilisation of Al (Driscoll *et al.*, 2001), soil acidity status should not depend so much on deposition the current year but rather on accumulated deposition during a number of preceding years. In addition to SDep, data from EMEP also include oxidized N (NO_3^-) and reduced N (NH_4^+) deposition; for this study, however, only data for SDep were

used as the gradients for the three kinds of deposition were highly correlated in Sweden during that period (r = 0.99 for SDep vs NO_3^-, SDep vs NH_4^+ and NO_3^- vs NH_4^+). The SDep data that were used do not include S from sea salt sources. The influence of chloride deposition (data available from the SWETHRO network (Karlsson *et al.*, 2011), r=0.69 for SDep vs Cl), which is particularly evident on the western coast of Sweden (Akselsson *et al.*, 2013), was not assessed in this study.

The analyses of the acidity variables pH, BS, Ca and Al with respect to RA and SDep were done, separately for the O, B and C horizons, in two ways: plots and multiple regression analysis. The software SAS version 9.4 (SAS Institute Inc., Cary, NC, USA) was used. The residuals in the analyses were checked for normality and it was found, that if logarithmically transformed values of Ca and Al were used, the residuals became more normally distributed. Thus, the natural logarithms of Ca (lnCa) and Al (lnAl) were used in the statistical analyses.

In the plots of the acidity variables versus RA and SDep, data from all soil samples in each horizon were aggregated into 10 equally sized groups according to the value of the independent variable (RA or SDep). For each of these grouped averages of RA or SDep, average value and 95% confidence intervals for the acidity variable are shown. As it was found that the relationships of pH, BS, lnCa and lnAl with RA were non-linear, a quadratic term (RA^2) was included in the regression line as well as in the rest of the analyses.

Multiple regression analysis of pH, BS, lnCa and lnAl versus RA, RA^2, SDep and the interactions RA*SDep and RA^2*SDep was carried out using the regression model:

$$v = a + b * RA + c * RA^2 + d * SDep + e * RA * SDep + f * RA^2 * SDep + g$$

where *v* is one of the acidity variables (pH, BS, lnCa or lnAl) for a soil horizon (O, B or C), "*a*" is the intercept, *b*, *c, d, e* and *f* are parameter estimates for each term and horizon and *g* is an independently and normally distributed error term. In order to be able to compare acidification in different parts of Sweden, analyses were made for the whole country as well as for two regions "North" and "South", divided at latitude 61.4° N (equal number of observations in the two groups). Considering the similarity between the deposition and temperature gradients in Sweden (r=0.89 for SDep vs temperature sum in the SFSI data set), we also separated the effects of deposition and temperature on soil acidity by doing multiple regression of the acidity variables versus SDep, temperature sum (TSum) and the interaction SDep*TSum, according to the model:

$$v = a + b * SDep + c * TSum + d * SDep * TSum + e$$

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where *v* is one of the acidity variables (pH, BS, lnCa or lnAl) for a soil horizon (O, B or C), "*a*" is the intercept, *b*, *c* and *d* are parameter estimates for each term and horizon and *e* is an independently and normally distributed error term. Data of temperature sum according to Odin *et al.* (1983) were used. As the magnitude is very different for the explaining variables RA, SDep and TSum ($0<RA<2$, $0<SDep<800$ and $0<TSum<1600$), all explaining variables were normalised with mean=0 and variance=1 prior to multiple regression analysis.

4 Results

4.1 Magnitude of acidification by tree harvesting and by deposition (Paper I)

Estimated net cation extraction by harvesting of Scots pine, Norway spruce and birch, compared to acidification by deposition, is given in Figure 2. Acidification by tree harvesting is largest in southwestern Sweden and decreases further north, due to the warmer climate in the south. Harvesting of Norway spruce causes more acidification per unit area than harvesting of Scots pine. The acidifying effect of birch harvesting is in between that of Norway spruce and Scots pine. Looking at the acidifying effect of harvesting various tree parts, stem wood harvesting yields the highest acid input per unit area to the soil followed by logging residue harvesting and then stump harvesting. Comparing harvesting of logging residues and stumps, the former gives a higher acid input per unit area but it also gives a higher acid input per unit heating value, when used as biofuel (Table 1), as the higher nutrient content of logging residues gives a higher acidifying effect compared to stumps for the same amount of harvested biomass. Comparing tree harvesting with acidity in deposition during 1996-2009 (Figure 2), whole-tree + stump harvesting of Scots pine yields acidification equal to that of acid deposition while whole-tree + stump harvesting of Norway spruce acidifies the soil more than deposition. Whole-tree harvesting of birch causes acid input comparable with that of deposition in northern Sweden and higher acid input than deposition in the south.

Future (2010-2109) projections of tree harvesting, compared to recent (1996-2009) acid deposition levels, are presented in Figure 3. In the figure, lower and upper boundary for acidity in deposition are presented as green lines; the very low lower boundary for deposition in southwestern Sweden is due to occasional input of base cations from sea salt on the western coast (Akselsson *et al.*, 2013; Köhler *et al.*, 2011). In a "business as usual" scenario (Claesson, 2008), tree growth and harvesting are expected to increase considerably during the following century. Norway spruce is expected to increase the most while Scots pine and birch will not increase as much. Increased tree harvesting will yield increased biological acidification, which is particularly evident for Norway spruce; harvesting of this tree species, including harvesting of logging residues and stumps, will cause considerably more acid input to the soil than current acidity in deposition. Harvesting of Scots pine (stem wood, logging residues and stumps) and birch (logging residues and stumps) is projected to yield acidification to the soil comparable with acidification by deposition 1996-2009.
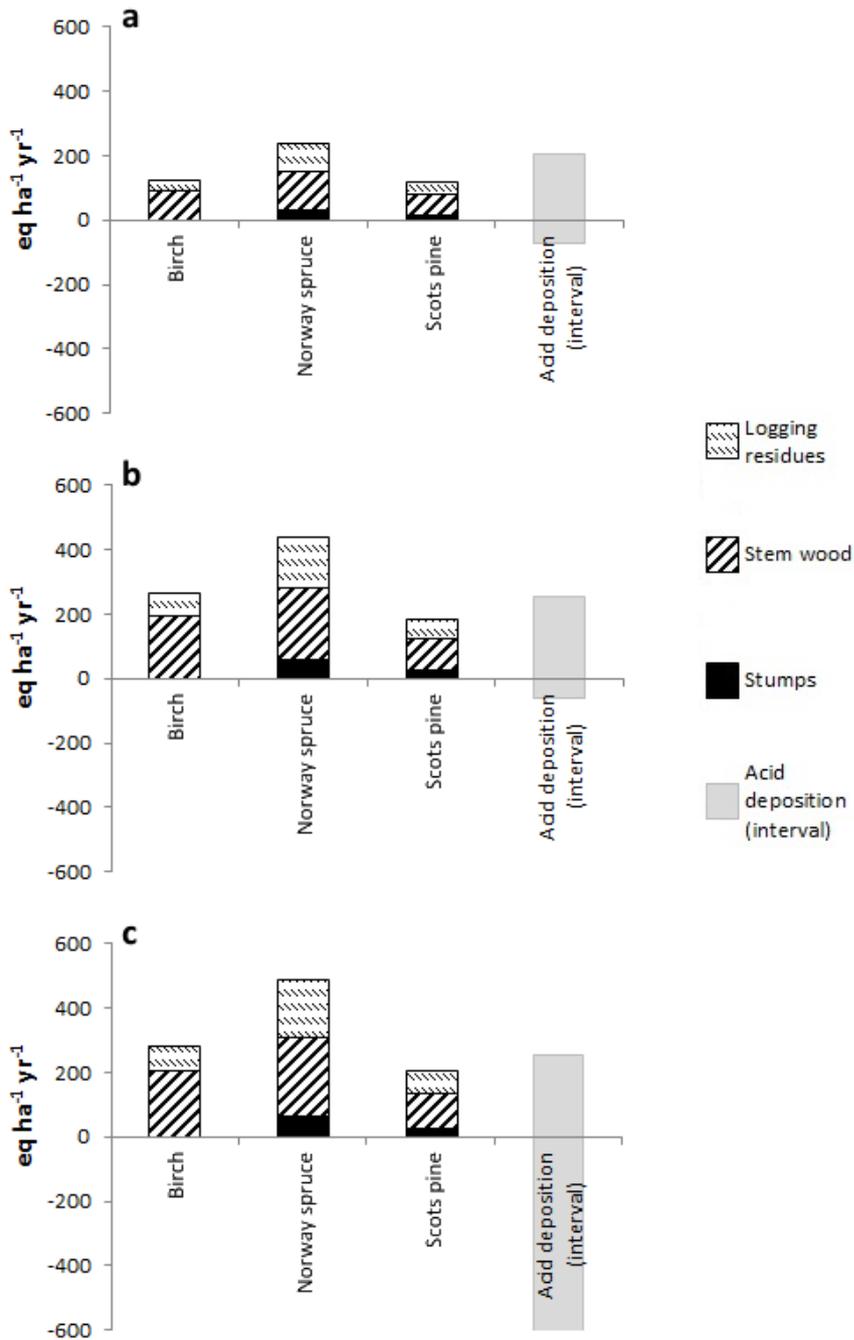
Figure 2 Estimated net cation extraction (equivalents per hectare and year) by harvesting of tree parts in three regions in Sweden during 2010-2019. For comparison, range of acidity in deposition (defined as negative value of ANC in deposition during 1996-2009 for reference sites) is presented.

a – north Sweden, b – mid & southeast Sweden, c – southwest Sweden
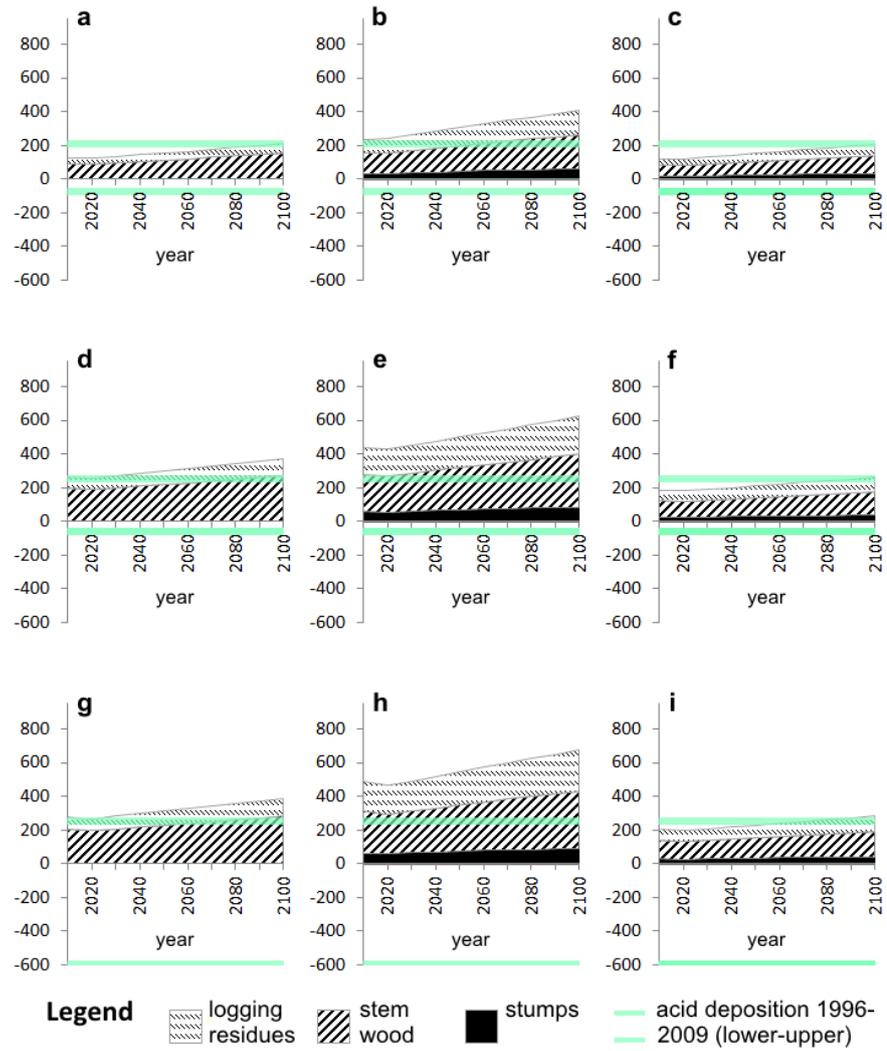
Figure 3 Predictions of acidification by tree growth (equivalents per hectare and year) through harvesting of stumps, stem wood (including bark) and logging residues during the period 2010-2109. For comparison, current max and min values of acidity in deposition (acidity defined as negative value of ANC in deposition during 1996-2009 for reference sites) are presented as green lines.
a b c – north Sweden, d e f – mid & southeast Sweden, g h i – southwest Sweden
a d g – birch, b e h – Norway spruce, c f i – Scots pine

Table 1 Acid input to the soil from harvesting stumps and logging residues relative to the heating value of these tree parts as biofuel (equivalents H^+ MWh^{-1})

	Stumps	Logging residues	Stumps and logging residues
Birch	27	51	37
Norway spruce	18	66	37
Scots pine	12	39	23

4.2 Effects on soil acidity of tree growth and deposition (Paper II)

Plots of the acidity variables versus RA and SDep are presented in Figure 4. The plot of pH versus RA (Figure 4a) shows that pH in the O and B horizons decrease with RA at least up to the least cutting age (RA≈1). The decrease in the O horizon is particularly large in the younger stands. In the C horizon there is virtually no change in pH with RA. This plot supports the hypothesis that tree growth results in increased acidity in the O horizon and the upper part of the mineral soil, but not in the deep soil layers. In the BS versus RA plot (Figure 4c), BS in the O horizon declines rapidly in the beginning but recovers with increasing RA. In the B horizon there is a slower decline of BS with RA (for RA≤1) and in the C horizon there is no significant decline. The curves for lnCa vs RA (Figure 4e) are similar to those of BS vs RA. LnAl versus RA for the O horizon (Figure 4g) looks opposite to lnCa vs RA; it increases sharply in the beginning but peaks at a somewhat later RA≈0.6 than for the lnCa minimum (RA≈0.4). In the B horizon, lnAl vs RA is non-significant and in the C horizon there is no clear trend for lnAl vs RA.

Plots of the acidity variables with respect to SDep (Figures 4b, d, f and h) show much stronger and more consistent trends than plots with respect to RA. With the exception of pH in the O horizon, adjusted r^2 values are always higher for relations with SDep than with RA. In the plots with respect to RA, the acidity variables generally show the largest variation in the O horizon and adjusted r^2 values are lower in the C horizon. In the plots with respect to SDep, on the other hand, the response of the acidity variable is fairly equal in all soil horizons, both in terms of adjusted r^2 values and shape of the curve. The pH, BS and lnCa vs SDep plots (Figures 4b, d and f) all show negative trends with SDep in all soil horizons. Conversely, lnAl vs SDep (Figure 4h) show consistent positive trends in all horizons. The strong correlations of acidity variables with SDep in all soil horizons support the hypothesis that when soils are acidified by deposition, acidity is percolated through the O horizon into the mineral soil and also deeper soil layers are acidified.

In the plots, r^2 values and significance levels are for the grouped average values whereas in the multiple regression analyses (Tables 2 and 3), they are for

the individual soil samples. The multiple regression analysis of pH, BS, lnCa and lnAl versus RA, RA^2, SDep and the interactions RA*SDep and RA^2*SDep (Table 2), verifies that the acidity of Swedish forest soils is more strongly related to deposition than to tree growth. All acidity variables in all horizons are significantly correlated with SDep; in most cases $p<0.001$. RA and RA^2, on the other hand, are significant only for pH in the O and B horizons and for lnAl in the O horizon. The only variable that is more strongly correlated with RA than with SDep is pH in the O horizon. The coefficient of determination (adjusted r-square value) increases down the soil profile and is highest in the C horizon for all acidity variables with the exception of lnCa. The acidity variables seem to be correlated with RA and RA^2 independently of SDep and vice versa, as the interactions RA*SDep and RA^2*SDep are non-significant for all variables except pH and lnCa in the B horizon. RA and SDep are not correlated with each other ($r = -0.040$, $p>0.05$).

In a multiple regression analysis similar to Table 2, where Sweden is divided into a southern and a northern part at latitude $61.4°$ N (equal number of observations), the correlation of the acidity variables with SDep is often stronger in the southern part of the country than in the north (data not shown). RA in the O horizon, on the other hand, is more significant in the northern part than in the south. As in Table 2, SDep is more significant than RA throughout; the only exception is pH in the O horizon, northern Sweden.

Table 3 shows the results of the multiple regression analysis versus SDep, temperature sum (TSum) and the interaction SDep*TSum. In Sweden, the deposition and temperature gradients are similar ($r=0.89$ for SDep vs TSum) but deposition and temperature may have different effects on soil acidity. In order to be able to separate the effects of deposition and temperature, TSum was included in the analysis alongside with SDep. In Table 3, the correlation with SDep is generally negative, the correlation with TSum is positive and the correlation with the interaction SDep*TSum is also positive. For lnAl in the O horizon, though, the correlation with SDep is positive and the correlation with SDep*TSum is negative. The correlation with SDep is significant throughout except for lnAl in the B horizon; the correlation with TSum is significant for lnCa in all horizons, BS in the C horizon and lnAl in the O horizon; and the interaction SDep*TSum is significant in all of the analyses except pH and lnAl in the B horizon.
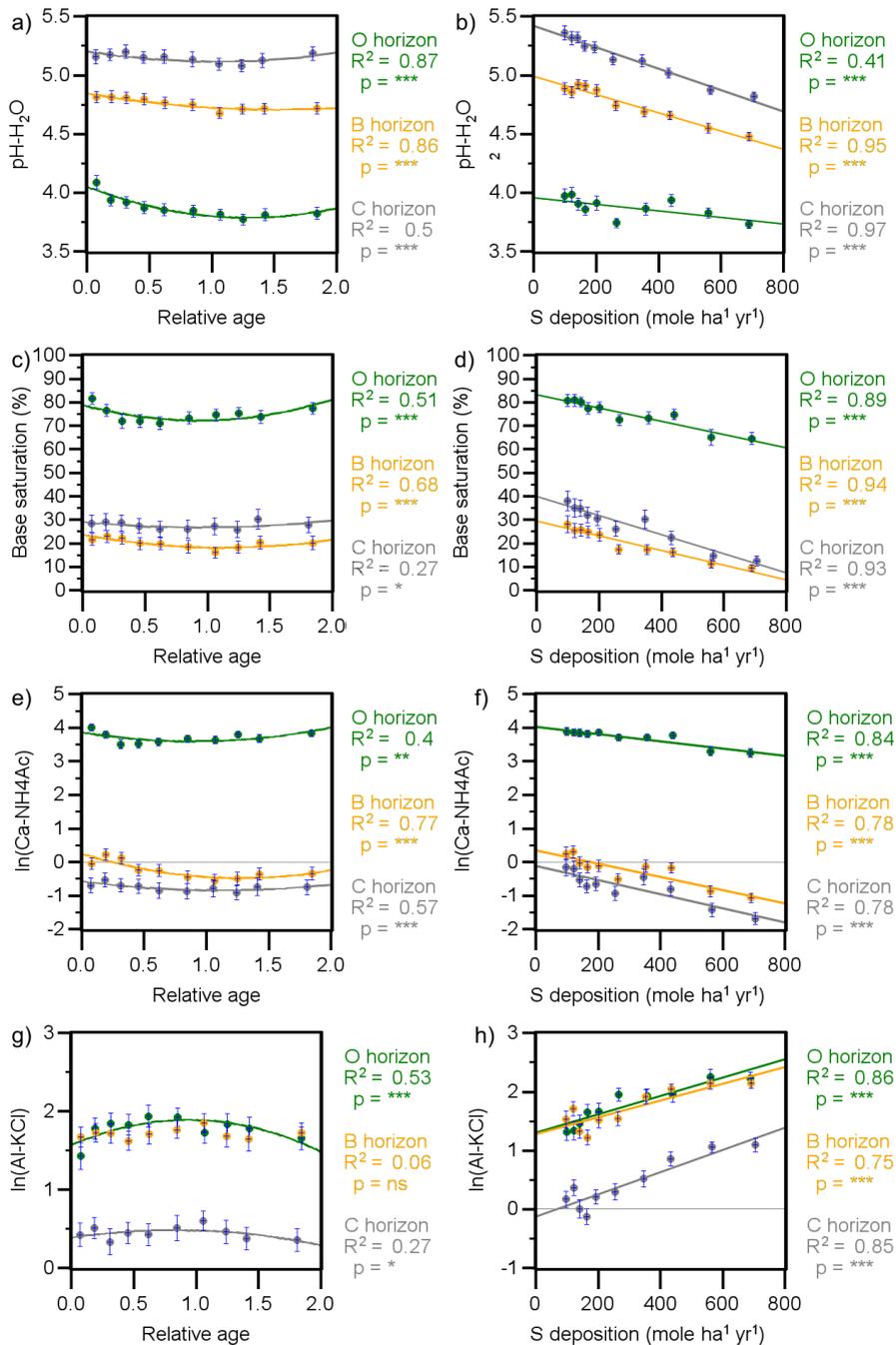
Figure 4 Plots of pH (a-b), BS (c-d), lnCa (e-f) and lnAl (g-h) versus RA RA^2 (left) and SDep (right), respectively, with r^2 values and significance levels, for the O (green), B (orange) and C (grey) horizons. Dots are grouped average values and error bars indicate 95% confidence intervals. When there is a significant correlation with RA and RA^2 or SDep, it is marked with a line. Significance levels: * - p<0.05, ** - p<0.01, *** - p<0.001, ns – not significant.

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Table 2 Multiple regression analysis of pH, BS, lnCa and lnAl versus RA^2, RA, SDep and the interactions RA*SDep and RA^2*SDep. All explaining variables were normalized with mean=0 and variance=1 prior to analysis. Significance levels: * - p<0.05, ** - p<0.01, *** - p<0.001, ns – not significant.

Dependent Variable	Horizon	n	Adj. r^2	Intercept	Estimates					Significance levels				
					RA^2	RA	SDep	RA* SDep	RA^2* SDep	RA^2	RA	SDep	RA* SDep	RA^2* SDep
pH-H$_2$O	O	1990	0.06	3.87	0.11	-0.18	-0.07	0.04	-0.01	**	***	**	ns	ns
	B	1902	0.22	4.76	0.07	-0.14	-0.21	0.15	-0.09	*	***	***	**	*
	C	1477	0.26	5.15	0.03	-0.05	-0.20	0.07	-0.04	ns	ns	***	ns	ns
Base saturation	O	1990	0.08	74.78	3.23	-3.38	-5.14	0.18	0.11	ns	ns	***	ns	ns
	B	1902	0.11	19.97	0.73	-3.29	-7.03	0.97	1.09	ns	ns	***	ns	ns
	C	1477	0.13	27.67	-1.66	-0.16	-8.85	0.41	1.68	ns	ns	***	ns	ns
lnCa	O	1989	0.06	3.70	0.06	-0.09	-0.19	-0.09	0.12	ns	ns	***	ns	ns
	B	1900	0.11	-0.24	0.02	-0.22	-0.27	-0.36	0.32	ns	ns	***	ns	*
	C	1476	0.08	-0.76	-0.14	0.06	-0.34	-0.27	0.26	ns	ns	***	ns	ns
lnAl	O	1969	0.09	1.78	-0.24	0.22	0.32	-0.16	0.16	**	*	***	ns	ns
	B	1896	0.10	1.71	-0.03	0.11	0.38	-0.23	0.10	ns	ns	***	ns	ns
	C	1466	0.18	0.44	0.01	0.06	0.47	-0.15	0.03	ns	ns	***	ns	ns

Table 3 Multiple regression analyses of pH / BS / lnCa / lnAl versus SDep, temperature sum (TSum) and the interaction SDep*TSum. The explaining variables SDep, TSum and SDep*TSum were normalized with mean=0 and variance=1 prior to analysis. Significance levels: * - p<0.05, ** - p<0.01, *** - p<0.001, ns – not significant.

Dependent variable	Horizon	n	Estimates				Adj. r²	Significance levels		
			Intercept	SDep	TSum	SDep*TSum		SDep	TSum	SDep*TSum
pH-H₂O	O	1990	3.87	-0.41	0.03	0.34	0.03	***	ns	***
	B	1902	4.76	-0.17	0.01	0.01	0.20	**	ns	ns
	C	1477	5.15	-0.39	0.01	0.21	0.27	***	ns	**
Base saturation	O	1990	74.78	-22.68	0.51	17.24	0.09	***	ns	***
	B	1902	19.97	-25.19	1.29	18.28	0.12	***	ns	***
	C	1477	27.67	-30.82	5.18	18.59	0.15	***	***	***
lnCa	O	1989	3.70	-0.74	0.17	0.40	0.07	***	***	*
	B	1900	-0.24	-2.34	0.33	1.70	0.14	***	***	***
	C	1476	-0.76	-2.78	0.47	1.99	0.15	***	***	***
lnAl	O	1969	1.78	1.30	0.16	-1.16	0.10	***	**	***
	B	1896	1.71	-0.01	0.00	0.29	0.10	ns	ns	ns
	C	1466	0.45	-0.41	-0.03	0.82	0.19	*	ns	***

5 Discussion

5.1 Magnitude of acidification by tree harvesting and by deposition (Paper I)

The magnitude of acidification by harvesting of various tree parts is outlined in Figure 2. Harvesting of logging residues and stumps, in addition to stem wood, leads to higher nutrient exports from the soil. This is in line with other studies: A modelling study of the long-term effect of whole-tree harvesting (harvesting of branches, tree tops and needles in addition to stem wood) on soil and stream water acidity in three Swedish forested catchments (Zetterberg *et al.*, 2014) predicted that whole-tree harvesting led to depletion of soil exchangeable Ca and decrease of stream water ANC. A field study of soil exchangeable Ca after conventional and whole-tree harvest, respectively, in Sweden (Zetterberg *et al.*, 2016) concluded that Ca pools decreased at all sites and treatments during 40 years after harvest, but that the pools were lower at the whole-tree harvested plots. Over time, however, the difference in Ca diminished between the two harvesting regimes. In the current study, the net cation extraction by harvesting of tree parts (Figure 2) is considerably higher for Norway spruce than for birch or Scots pine and the reason is that harvested Norway spruce biomass contains more base cations than Scots pine or birch biomass. Norway spruce needles contain more Ca (Werkelin *et al.*, 2005) and the stands generally have higher biomass per unit area than Scots pine stands. Birch leaves are rich in base cations (Werkelin *et al.*, 2005), but as only branches, not leaves, are harvested as part of birch logging residues, the net cation extraction is not that large.

From a nutrient point of view, when harvesting logging residues from Norway spruce or Scots pine a possibility to decrease losses is to leave the needles on-site, as they contain large amounts of base cations but constitute only a minor part of the tree biomass. In a modelling study evaluating the environmental impact of different forest harvesting scenarios in Finland (Aherne *et al.*, 2012), only the scenarios where needles were left on-site (stem-only harvesting and stem-branch harvesting) were considered sustainable without additional nutrient inputs. In a meta-analysis of published data worldwide on harvesting forest residues (Achat *et al.*, 2015), one of the conclusions were that harvesting foliage induces a small gain in biomass harvest but large nutrient losses, due to the low mass of foliage and its high nutrient concentrations. In Sweden, it has been recommended by the Swedish Forest Agency (2005) to leave the needles on-site as evenly distributed as possible; needles should be harvested only on nitrogen-rich soils in order to minimize the risk for N leaching. Another factor that has influence on nutrient export is the recovery rate, i.e. the

fraction of logging residues that are harvested during logging. According to a review of whole-tree harvesting in boreal and temperate forests in North America and Europe (Thiffault *et al.*, 2014), the recovery rate varies from 4 to 89 % with an average of 52 %. The sites studied in Sweden and Finland had high recovery rates (72 %) attributed to strong support for the use of forest biomass for energy during a long time which, in turn, has created a comparatively strong market and good economic value for forest biomass. A high recovery rate is desirable from an economic point of view; however, high nutrient export increases the risk for soil acidification.

In the current study, logging residues have considerably higher base cation contents than stumps, which is attributable to the higher contents of base cations in branches and needles compared to stem wood (Werkelin *et al.*, 2005). Even if needles are left on-site, branches contain more base cations per mass unit than stump wood. Stump harvesting, therefore, seems like a better alternative than logging residue harvesting from a strict nutrient balance and acidification perspective. This is, however, assuming that stumps have similar contents of cations and anions as stem wood. There are concerns that stump harvesting may lead to increased CO_2 emissions from the soil due to soil disturbance; however, recent studies in Sweden (Egnell *et al.*, 2015; Strömgren *et al.*, 2013) and Finland (Karlsson & Tamminen, 2013) have found that stump harvesting has little or no effect on the soil C pools.

When comparing acidification by tree harvesting with acidification by deposition, it seems that, particularly, harvesting of Norway spruce may pose a problem concerning soil acidity and nutrient sustainability in the future. This is evident in future projections of the two kinds of acidification (Figure 3), where Norway spruce harvesting is expected to lead to substantially more acidification than current acidification by deposition. According to a study of base cation budgets in Swedish forest soils (Akselsson *et al.*, 2007) there is a need for compensatory fertilization, e.g. ash return, after whole-tree harvesting in most of Sweden. Ash return is also recommended by the Swedish Forest Agency on locations where significant amounts of forest residues are harvested, in order to mitigate the acidifying effects of biomass extraction (Hjerpe, 2008). According to a comparative study of stem-only and whole-tree harvesting at four sites in Sweden (Brandtberg & Olsson, 2012), whole-tree harvesting causes more nutrient depletion and soil acidification but the effects do not always persist over time; at the sites studied, differences between harvesting regimes in calcium and manganese levels in the forest floor and topsoil diminished during the first 25 years after harvest. After 37-38 years, the differences between stem-only and whole-tree harvested plots had continued to diminish although levels of

exchangeable Ca were still lower in the whole-tree harvested plots (Zetterberg *et al.*, 2016).

Base cations are lost from a forest ecosystem due to harvest and leaching; input sources are deposition and mineral weathering. As weathering is a slow process and difficult to measure directly, weathering estimates are often impaired by large uncertainties. A synthesis of seven different methods to estimate weathering rates at a forested catchment in northern Sweden (Klaminder *et al.*, 2011) concluded, that none of the methods could be used to predict weathering with enough precision to assess whether or not certain forestry practices are sustainable. In another study, where base cation budgets were calculated to estimate weathering rates at a site in southwestern Sweden (Simonsson *et al.*, 2015), uncertainties in calculated weathering rates were often greater than the calculated weathering rates themselves. A conclusion was, though, that forest management on soils with low weathering rates is highly dependent on internal base cation cycling in the stand in order to be sustainable. Overall, it is today not possible to say exactly to what extent weathering will be able to compensate for base cation losses through harvesting. Continued monitoring is necessary to determine the sustainability of forestry in this regard.

In a mass balance study like this there are, of course, also uncertainties. Data of cation and anion contents in tree parts, used for calculating acidification by tree harvesting, are from various sources in Sweden and Finland (with a climate similar to Sweden). Use of other sources would have yielded different data, which may have affected magnitude of acidification by tree harvesting as well as differences between tree species. The ratio of needles in logging residues used in the calculations also matters, as needles have considerably higher nutrient contents than branches. A modelling study of the long-term effects of whole-tree harvesting on soil Ca supply and stream water ANC (Zetterberg *et al.*, 2014) concluded that tree biomass Ca^{2+} concentrations was the single most important factor determining modelled outcome. The assumption that stump wood has cation and anion contents similar to stem wood has not been tested; the stump itself may be similar to the stem in that respect but the accompanying roots may have higher cation and anion contents.

5.2   Effects on soil acidity of tree growth and deposition (Paper II)

The decrease of pH, BS and exchangeable Ca in the O horizon with relative age found in this study can be attributed to tree uptake of base cations and release of hydrogen ions. It supports the hypothesis that tree growth causes a net H^+ input to the upper soil horizons. Similar pH declines have been found in other studies

in Canada (Shrestha & Chen, 2010; Brais *et al.*, 1995), the US (Matlack, 2009) and Sweden (Zetterberg *et al.*, 2016; Brandtberg & Olsson, 2012; Tamm & Hallbäcken, 1986). In the current study, the change towards more acidic conditions is particularly large early in the forest rotation, which is difficult to explain from a nutrient uptake perspective as young trees cannot accumulate as much biomass as older trees. A previous study (Nilsson *et al.*, 1982) concluded that rates of hydrogen ion production reach a maximum early in the life of a forest (at around 20 years of age), which is in line with the findings here. A reason for the high levels of base cations in the O horizon at RA zero (Figures 4c and e) may be that residues from previous logging enrich the soil. A study of soil exchangeable calcium after conventional and whole-tree harvesting (Zetterberg *et al.*, 2016) concluded that after clear-cutting, soil Ca^{2+} pools partly recovered during a period of 15-20 years due to decomposition of residues, stumps and roots. At the same time, however, nutrient uptake by the next tree generation caused these Ca^{2+} pools to decline. The decline of lnCa and BS in the O horizon at low RA values seen in Figures 4c and e may, therefore, be a combined effect of decomposition of residues and tree uptake of base cations. In the beginning of the stand development, proportionally more biomass is allocated to the needle and bark biomass pools while the most rapid stem biomass accumulation comes later. As needles and bark contain more base cations per mass unit than stem wood (Werkelin *et al.*, 2005), this may contribute to, but not fully explain, the rapid loss of base cations from the O horizon in the beginning of the trees' life cycle. Leaching of Ca after tree harvesting, accompanied by anions SO_4^{2-}, Cl^-, NO_3^-, bicarbonate (HCO_3^-) and organic anions ($RCOO^-$) (Zetterberg *et al.*, 2014) may be an additional reason for loss of base cations. For higher RA values, there is an increase of BS and lnCa in the O horizon; this may be caused by the fact that these trees grow very slowly and there is litterfall, adding base cations to the forest floor.

Correlations of the acidity variables with RA and/or RA^2 are much weaker than correlations with SDep (Table 2); this implies that the acidity of Swedish forest soils is still to a larger extent a result of the historic deposition of acidity than a result of tree growth by the current forest generation. It is important to keep in mind, however, that soil data in this study are from the period 1993-2002 and that deposition, at least for sulphur, has declined considerably since then. This implies that the additional effect of deposition on soil acidity is not as large today as the results in this study suggest. In the plots of the acidity variables with respect to RA and SDep (Figures 4a-h) regression lines vs SDep are linear, pointing at a dose-response relationship; regression lines vs RA are more curvilinear which is why the RA^2 term is used. The non-linear shape of regression lines in relation to RA, which is most pronounced in the O horizon,

may be a result of acidification by tree growth for lower RA values and return of alkalinity by litterfall for higher RA values. The only variable for which the influence of RA^2 and RA is stronger than the influence of SDep is pH in the O horizon. Tree roots have a large influence on soil acidity in the O horizon (Nilsson *et al.*, 1982) while deposition seems to have more influence in the deeper horizons according to the adjusted r^2 values in Table 2. Projecting into the future, when deposition levels may continue to decline but biomass growth will probably increase, particularly for Norway spruce (compare Paper I), the effects of historic deposition may decrease and the effects of biomass growth and harvest grow stronger. As loss of base cations from the soil during the decades of acid rain will take a long time to replenish (Moldan *et al.*, 2013; Belyazid *et al.*, 2006; Likens *et al.*, 1996), it is likely that the effects of deposition during the latter part of the 1900s will remain to some extent in the soil for a very long time. Exchangeable base cations do not seem to be much affected by tree growth, as RA and RA^2 are non-significant for BS and lnCa in all horizons in the multiple regression analysis (Table 2). In plots 4c and 4e RA and RA^2 are significant for BS and lnCa, but the plots are for grouped average values and the tables for individual sampling points – the former giving more significant results and higher r^2 values. The weak relationship between base cations in the soil and tree growth during a forest generation may become stronger in the future if deposition declines further and harvest intensity increases (Zetterberg *et al.*, 2014; Moldan *et al.*, 2013); however, base cation stores in the soil may remain low for a long time due to the historic effects of deposition and earlier biomass harvests, thereby hampering any effect tree growth may have on the base cation pools during a forest generation.

In the multiple regression analysis (Table 2), the effect of SDep increases down the soil profile which is indicated by higher adjusted r^2 values in the C horizon than in the O horizon. This is probably due to the fact that deposition provides both acidity and mobile anions (SO_4^{2-}, NO_3^-) which enables leaching of acidity through the soil profile and into ground and surface waters (Driscoll *et al.*, 2001). It supports the hypothesis that deposition of S and N can acidify the whole soil profile due to the presence of mobile anions. RA and RA^2, on the other hand, are significant only in the O (pH and lnAl) and B (pH) horizons; this supports the finding by Nilsson *et al.* (1982) that tree growth primarily acidifies the root zone.

The more significant results for SDep found in the south of Sweden than in the north can be attributed to the higher accumulated deposition in southern Sweden. The fact that RA is significant in northern Sweden but not in the south for pH in the O horizon, however, indicates that the acidifying effect of tree growth is relatively stronger in northern Sweden, where deposition has had less

influence. In the southern part of Sweden, the influence of deposition overshadows that of the growing tree stands.

The acidity variables are, generally, negatively correlated with SDep and positively correlated with TSum (Table 3). The positive effect of TSum on soil exchangeable Ca and base saturation may be caused by the fact that higher temperature enhances mineralization and weathering of Ca and other base cations. The interaction SDep*TSum is mostly significant and opposite to SDep (positive when SDep is negative). This causes the product SDep*TSum to be a counteracting factor for the acidifying effect of SDep; it reduces the slope of the line for the acidity variables versus SDep. This indicates that in the south of Sweden, pH, BS and lnCa do not decrease as much by a certain increase in SDep as in the north – the decrease with deposition is steeper in northern Sweden. If this is the case, it could be attributed to the much lower historic deposition in northern Sweden; soils in the north are at an earlier stage of acidification and thus respond faster to deposition, whereas soils in the south are in a more advanced acidified state (Warfvinge & Bertills, 2000).

In this study of human-induced acidification, the effect of chloride (Cl^-) deposition is not considered. Cl^- deposition is derived from sea salt and can, temporarily, lead to acidification of soils during sea salt episodes particularly along the Swedish western coast (Akselsson *et al.*, 2013). When SO_4^{2-} deposition has declined, Cl^- has become the dominant anion in many Swedish lakes (Futter *et al.*, 2014). The omission of Cl^- deposition in this study may have somewhat weakened the relationships between deposition and soil acidity variables, mainly in the southwestern part of the country.

When trying to analyse the effect of one single variable on soil acidity (as in Figures 4a-h), the ideal case is if all other variables can be kept constant. That is not the case in this study. In the graphs with respect to RA, the ten dots that represent different RA stages are all average values of a number (150-200) of individual plots. For each of these grouped averages of RA, the distribution of other variables influencing soil acidity such as S deposition, temperature, bedrock and tree species, is different. This may affect each grouped average in different ways and lead to errors in the graphs with respect to RA. In a large dataset like the Swedish Forest Soil Inventory, however, this error should be smaller than in a dataset with fewer sampling points. In the multiple regression analyses (Tables 1 and 2), several variables are considered simultaneously which should decrease the risk for bias. There is still the risk, however, that variables that are not included in the analyses (e.g. tree species, Cl deposition) may have an impact on the results.

6   Conclusions

This study has used a large, statistically representative, national dataset covering all productive forest land in Sweden, to evaluate the acidification of Swedish forest soils hitherto studied at a limited number of experiments or monitoring sites.

Acidification by tree harvesting, counted per unit area, decreases in the order Norway spruce > birch > Scots pine. Comparing harvesting of different tree parts, acidification potential decreases in the order stem wood > logging residues > stumps. The acidifying effect of stump harvesting is fairly small compared to logging residue harvesting, due to the higher contents of base cations in branches and needles than in stem wood. Comparing acidification by tree harvesting with acidification by deposition during 1996-2009, harvesting of Scots pine and birch causes acidification of equal magnitude to that of deposition while Norway spruce harvesting acidifies the soil more than deposition. In the future, forest growth and harvest are expected to increase, particularly for Norway spruce, which will cause increased biological acidification.

Biological acidification and deposition of acidifying substances affect acidity related variables in Swedish forest soils. There is a consistent pattern where the effects of tree growth during one forest generation are limited to the upper soil layers while atmospheric deposition of acidity affects the whole soil profile, but is more evident in deeper soil layers where other processes that contribute to soil acidity have less influence. Deposition of S and N causes lower pH, exchangeable Ca and base saturation and higher exchangeable Al levels throughout the soil profile. Biological acidification is manifested by decreased pH, exchangeable Ca and base saturation and increased exchangeable Al primarily in the O and to some extent in the B horizon. This is particularly evident in the beginning of the trees' life cycle.

With expected continued declines in deposition and increase in biomass growth and harvest, biological acidification may become relatively more important, compared to acidification by deposition, in the future. It is likely, however, that the effects of historic deposition and biomass harvest will remain in Swedish forest soils for a long time to come. Recovery may be delayed due to continued high N deposition and intensive tree harvesting. Continued monitoring of forest soils is important to determine the effects of increased tree growth and harvest and examine to what extent previous high deposition levels will continue to affect the soil.

Försurning av svenska skogsjordar
Utvärdering av data från Markinventeringen

Sammanfattning

Försurning av skogsjordar har påverkat markens näringsstatus negativt och haft toxiska effekter på levande organismer. Syftet med denna avhandling var att studera den kvantitativa effekten av försurning av skogsjordar i Sverige på grund av (1) ökat uttag av trädbränsle, särskilt stubbrytning och (2) försurande deposition av svavel- och kväveföreningar samt biologisk försurning genom trädtillväxt under en skogsgeneration. Storleken av försurning på grund av biomassauttag beräknades med hjälp av skördeuppskattningar för stam-ved, grenar och toppar samt stubbar från en skoglig konsekvensanalys och data över katjoner och anjoner i olika träddelar. Försurning på grund av tillväxt definierades som nettokatjonupptaget (katjoner-anjoner).

Storleken av försurning på grund av deposition uppskattades med hjälp av depositionsdata från fyra avrinningsområden i olika delar av Sverige. Försurning på grund av deposition definierades som det negativa värdet av depositionens syraneutraliserande förmåga.

Den försurande effekten av tillväxt minskar i ordningen gran>björk>tall. Skörd av grenar och toppar orsakar mer markförsurning än skörd av stubbar, trots att massan för stubbar är högre, eftersom barr och grenar har högre halter baskatjoner än stam-ved. Skörd av tall och björk orsakar försurning på ungefär samma nivå som nuvarande försurning på grund av deposition medan skörd av gran orsakar högre försurning än vad som orsakas av deposition.

Data från en storskalig, nationell inventering av skogsmark användes för att studera den försurande effekten i markhorisonterna O (n=1990), B (n=1902) och C (n=1477) av tillväxt under en skogsgeneration respektive deposition. Fyra indikatorer för markens surhet valdes ut: pH-H_2O (pH), basmättnadsgrad (BS), utbytbart calcium (Ca) och utbytbart aluminium (Al). Dessa indikatorer analyserades mot trädens ålder och mot data över svaveldeposition med hjälp av linjär respektive multipel regression. Beträffande trädålder användes ett relativt åldersbegrepp för att kompensera för det faktum att träden växer olika snabbt i olika delar av Sverige.

Tillväxt och deposition av svavel- och kväveföreningar påverkar surhetsrelaterade variabler i svenska skogsjordar. Tillväxt under en skogsgeneration ger lägre pH, BS och Ca och högre Al i O- och i viss mån B-horisonten under första delen av trädens livscykel. Deposition ger lägre pH, BS och Ca och högre Al i O-, B- och C-horisonterna över hela depositionsgradienten i Sverige. Effekten av deposition är tydligast i de lägre marklagren där biologisk försurning har mindre inflytande.

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