

# Measured and Modelled Long-Term Effects of Whole-Tree Harvest

Impact on Soil and Surface Water Acid-Base Status in  
Boreal Forests

Therese Zetterberg

*Faculty of Natural Resources and Agricultural Sciences*

*Department of Aquatic Sciences and Assessment*

*Uppsala*

Doctoral Thesis  
Swedish University of Agricultural Sciences  
Uppsala 2015

Acta Universitatis Agriculturae Sueciae

2015:73

Cover: Logging residues from final felling, Unnaryd  
(photo: T. Zetterberg)

ISSN 1652-6880

ISBN (print version) 978-91-576-8344-1

ISBN (electronic version) 978-91-576-8345-8

© 2015 Therese Zetterberg, Uppsala

Print: SLU Service/Repro, Uppsala 2015

# Measured and Modelled Long-Term Effects of Whole-Tree Harvest. Impact on Soil and Surface Water Acid-Base Status in Boreal Forests.

## Abstract

This thesis examines the impact of whole-tree harvest (WTH) on soils and surface waters acid-base status in coniferous forests compared with conventional harvesting (CH). A combination of field observations (up to four decades) and dynamic modelling was used to describe the impact on soil calcium ( $\text{Ca}^{2+}$ ) pools and surface water acid neutralizing capacity (ANC). The studies were undertaken in northern and southern Sweden at sites belonging to the long-term wood fuel experiment (HELTRAD) and the ICP Integrated Monitoring (IM) programme. One of the most important findings was that WTH and CH caused large depletions (up to 60%) in soil exchangeable  $\text{Ca}^{2+}$  pools, circa 40 years after harvest. Despite these losses, tree growth and vitality has not yet been impaired. The results also implied that the fastest depletion rates occurred in CH-plots why soil  $\text{Ca}^{2+}$  pools between CH and WTH have become more similar with time. However, soil  $\text{Ca}^{2+}$  pools still remained significantly lower after WTH. Measured differences in soil solution showed that the impact of WTH on ANC was small ( $16 \mu\text{Eq l}^{-1}$ ) and temporary but site-specific. This difference was not large enough to counteract the natural recovery from acidification or lead to adverse ecological effects. The results indicate that WTH can have a large impact on soil exchangeable pools without causing surface water acidification in the absence of strong acid mobile anions. The rapidly declining soil  $\text{Ca}^{2+}$  pools may be of concern for sustainable forest management. However, trees will likely respond to lower  $\text{Ca}^{2+}$  availability by e.g. adjusting their uptake or develop biological feed-back mechanisms. The model predictions unanimously suggested that tree growth and net accumulation of nutrients in biomass caused large depletions in soil exchangeable  $\text{Ca}^{2+}$  pools and a decrease in stream ANC during the 19th and 20th centuries, exacerbated by acid deposition. Future predictions also suggested that these  $\text{Ca}^{2+}$  losses would continue accelerated by more intense harvesting practices. The impact on modelled stream ANC was smaller and depended on the concentrations of mobile anions. The empirical data pointed in the same direction as the MAGIC predictions, but the model exaggerated the impact. Until the discrepancy between measured and modelled impact have been resolved, interpretations changes related WTH using dynamic modelling or mass balance budget calculations should be done with caution.

*Keywords:* Acidification, ANC, calcium,  $\text{Ca}^{2+}$ , conventional harvesting, depletion, MAGIC, mass-balance, pH, soil, soil solution, streams, Sweden, whole-tree harvest.

*Author's address:* Therese Zetterberg, IVL Swedish Environmental Research Institute, P.O. Box 53021, 400 14 Gothenburg, Sweden. E-mail: therese.zetterberg@ivl.se

*It was the best of times, it was the worst of times...*

Charles Dickens, A tale of two cities

# Contents

List of Publications	7
Abbreviations	9
<b>1 Introduction</b>	<b>11</b>
<b>2 Objectives</b>	<b>15</b>
<b>3 Material and methods</b>	<b>17</b>
3.1 Measuring the impact on soils and surface waters acid-base status	17
3.2 Overview of study sites	17
3.2.1 The Integrated Monitoring (IM) sites (Paper I and III)	19
3.2.2 The long-term wood fuel experiment (Paper II and IV)	19
3.3 The MAGIC model	23
3.3.1 Modelling the IM-sites with MAGIC (Paper I and III)	24
3.3.2 Modelling the HELTRAD sites with MAGIC (Paper IV)	25
<b>4 Results and Discussion</b>	<b>27</b>
4.1 The impact of WTH on soil acid-base status	27
4.1.1 Modelling results (Paper I, III and IV)	27
4.1.2 Empirical results (Paper II and IV)	32
4.2 The impact of WTH on soil solution and stream water acid-base status (Paper I, II and III)	34
4.2.1 Modelling results (Paper I and III)	34
4.2.2 Empirical results (Paper II)	38
<b>5 Conclusions</b>	<b>43</b>
<b>6 Possible implications for the society and future research needs</b>	<b>45</b>
<b>References</b>	<b>49</b>
<b>Acknowledgements</b>	<b>55</b>



## List of Publications

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

- I Köhler, S. J., Zetterberg, T., Futter, M. N., Fölster, J., Löfgren, S. (2011). Assessment of Uncertainty in Long-Term Mass Balances for Acidification Assessment: A MAGIC model exercise. *Ambio* 40, 891-905.
- II Zetterberg, T., Olsson, B. A., Löfgren, S., von Brömssen, C., Brandtberg, P-O. (2013). The effect of harvest intensity on long-term calcium dynamics in soil and soil solution at three coniferous sites in Sweden. *Forest Ecology and Management* 302, 280-294.
- III Zetterberg, T., Köhler, S. J., Löfgren, S. (2014). Sensitivity analyses of MAGIC modelled predictions of future impact of whole-tree harvest on soil calcium supply and stream acid neutralizing capacity. *Science of the Total Environment* 494-495, 187-201.
- IV Zetterberg, T., Olsson, B. A., Löfgren, S., Hyvönen, R., Brandtberg, P-O. Long-term depletion in soil calcium pools based on measured and modelled data, up to four decades after conventional and whole-tree harvest. (Manuscript).

Papers I-III are reproduced with the kind permission of the publishers.

The contribution of Therese Zetterberg (TZ) to the papers included in this thesis was as follows:

- I TZ was partly involved in defining objectives and the experimental design. Stephan Köhler (SJK) was main responsible for the modelling. TZ was main responsible for input data and partook in the calibrations. TZ contributed with interpreting the results, writing and publishing the paper.
- II TZ, Bent Olsson (BO) and Stefan Löfgren (SL) contributed equally to the central ideas and hypotheses. TZ had the main responsibility for planning and carrying out the work, interpreting the results and writing the paper. TZ was main responsible for the soil solution study (experimental design, planning, organizing, validating and analysing the data). BO was main responsible for the soil study. Per-Olov Brandtberg (POB) conducted the soil sampling and analysis. Claudia von Brömssen (CvB) and BO did the statistical analysis for the soil solution and soil chemistry, respectively. SL contributed with interpreting the results and writing the paper.
- III TZ, SJK and SL contributed equally to the central ideas and. TZ had the main responsibility for planning and carrying out the work, interpreting the results, writing and publishing the paper. All of the modelling was done by TZ. SL contributed with interpreting the results and writing the paper. SJK contributed with text.
- IV TZ, BO and SL contributed equally to the central ideas and hypothesis. TZ had the main responsibility for planning and carrying out the work, interpreting the results and writing the paper. TZ was responsible for the modelling. CvB and BO did the statistical analysis. Riitta Hyvönen (RH) computed the decomposition curves. POB conducted the soil sampling and analysis. SL contributed with interpreting the results and writing the paper.



## Abbreviations

ANC	Acid Neutralizing Capacity
ANOVA	Analysis of Variance
BC	Base cations
BS	Base saturation
CEC	Cation exchange capacity
CH	Conventional Harvesting
EA	Exchangeable Acidity
IM	Integrated Monitoring
MAGIC	Model of Acidification of Groundwater In Catchments
SI	Site Index
SWETHRO	Swedish Throughfall Monitoring Network
WTH	Whole-Tree Harvesting



# 1 Introduction

To mitigate climate change, the member states of the EU have committed to a set of national binding targets to increase the use of renewable energies by 2020 (European Commission, 2007). Forest biomass plays a significant role in this strategy, particularly in countries with vast forest resources such as Sweden where utilization of logging residues has steadily increased (Swedish Forest Agency, 2013). However, increased recovery of biomass in final felling or thinnings may compromise tree productivity and health and lead to soil and surface water acidification (Achat *et al.*, 2015; Kreuzweiser *et al.*, 2008). These effects are caused by a greater removal of nutrients and buffer capacity when branches, tops and needles, in addition to stems, are harvested (WTH) compared with conventional harvest of stems-only (CH) (Iwald *et al.*, 2013).

The short- and medium-term impact of WTH on soils and surface water acid base status is well-described in the literature (Achat *et al.*, 2015; Thiffault *et al.*, 2011; Kreuzweiser *et al.*, 2008; Feller, 2005). Although the results are not unambiguous, increased base cation (BC) depletion after WTH is usually demonstrated. Of all macronutrients, calcium ( $\text{Ca}^{2+}$ ) appears to be frequently affected by increased harvest intensity (McLaughlin, 2014; Thiffault *et al.*, 2011; Federer *et al.*, 1989). The observed differences between CH and WTH are usually explained by a relocation of base cations from decomposing logging residues to the soil exchange complex in CH-plots, thereby creating a measurable difference compared with WTH-plots (Belleau *et al.*, 2006). As a result, the solute leaching from soils subjected to WTH is expected to have lower concentrations of BC, which could prevent or delay a recovery from acidification in surface waters (Futter *et al.*, 2014).

Our understanding of the long-term response in soils and water is limited to a few empirical studies which were established in the seventies or early eighties (van der Heijden *et al.*, 2013; Brandtberg & Olsson, 2012; Saarsalmi *et al.*, 2010; Vanguelova *et al.*, 2010; Walmsley *et al.*, 2009). In Sweden, one of the oldest field experiments for studying the impact of CH and WTH was laid

out in 1974-76 by Björkroth and Rosén (1977). Soil and tree productivity studies in this experiment have shown that WTH can reduce tree growth (Egnell, 2011; Egnell & Valinger, 2003; Egnell & Leijon, 1999), lower the nutrient concentrations in needles (Olsson *et al.*, 2000) and increase soil acidity (Olsson *et al.*, 1996). However, later soil studies from stand age 15 to 25 years showed that the differences between WTH and CH had decreased but not completely diminished (Brandtberg & Olsson, 2012), indicating that the differences are not permanent. These studies have provided valuable insight and empirical evidence on the effects of WTH in Swedish boreal forests. However, the impact of practicing WTH over a rotation period is still unknown making it difficult to assess the long-term sustainability.

As a complement to field experiment, a number of steady state mass balances studies have been conducted to evaluate impact of WTH on soil  $\text{Ca}^{2+}$  pools on different temporal and spatial scales, defined as the difference between inputs (deposition and weathering) and outputs (leaching and tree uptake). Generally, these budgets show negative balances for  $\text{Ca}^{2+}$  for a variety of tree species and soil types across Europe (Johnson *et al.*, 2015; van der Heijden *et al.*, 2011; Akselsson *et al.*, 2007; Joki-Heiskala *et al.*, 2003; Rademacher *et al.*, 2001), Canada (Watmough & Dillon, 2003) and the United States (Vadeboncoeur *et al.*, 2014; Huntington *et al.*, 2000). In Sweden, they have been widely used for assessing the acidifying effect of forestry on soils and for revising environmental goals (Swedish Environmental Protection Agency, 2007a).

Dynamic models that account for time-dependent changes (MAGIC, ForSAFE etc.) have also been used to simulate the biogeochemical response to different harvesting scenarios. In the last three decades, MAGIC has been used to predict the potential effect of land use changes and different forest management scenarios across Europe (Aherne *et al.*, 2012; Oulehle *et al.*, 2007; Ferrier *et al.*, 1995; Durand *et al.*, 1992; Jenkins *et al.*, 1990) and the US (McDonnell *et al.*, 2013). A number of these studies provide examples how land use changes such as afforestation may deplete the soil of base cations (BC) and increase surface water acidity, despite declines in sulphate ( $\text{SO}_4^{2-}$ ) deposition (Ferrier *et al.*, 1995; Jenkins *et al.*, 1990). However, the effect of CH or WTH on surface water acidity in already forested areas has only been considered in a few studies (McDonnell *et al.*, 2013; Aherne *et al.*, 2012; Aherne *et al.*, 2008).

Based on the long-term predictions by mass balances and dynamic models, concern about the long-term sustainability and risk for acidification after WTH has been expressed by Swedish authorities (Swedish Environmental Protection Agency, 2007a). However, the model results have been difficult to verify with

long-term experimental data. By combining the results from empirical studies with mass-balance equations and dynamic modelling, the validity of model results can be assessed.



## 2 Objectives

The overall objective of this thesis was to assess the long-term impact on soil and surface water acid-base status after WTH based on measured and modelled data. The aim was to increase our understanding of the potentially acidifying effect of harvest wood fuel as a mean to reduce the use of fossil fuels and mitigate climate change. A second aim was to increase the credibility of future predictions by validating model outcomes to measured data and identify uncertainties in input parameters.

More specifically, the main objectives of the four papers were to:

- Simulate long-term (1860-2010) trends in soil and stream water chemistry using the dynamic MAGIC model and test the robustness of the modelled outcome by varying a set of key soil parameters (Paper I).
- Determine if WTH leads to lower soil solution  $\text{Ca}^{2+}$  concentrations compared with CH, 27-35 years after clear-cutting (Paper II).
- Use MAGIC to model the long-term changes in soil exchangeable  $\text{Ca}^{2+}$  pools and stream water ANC following a virtual clear-cutting (WTH) and test the sensitivity in model outcome by varying key parameters associated with forestry (Paper III)
- Quantify long-term changes in soil exchangeable  $\text{Ca}^{2+}$  supply after WTH and compare the results with CH-measurements and MAGIC model simulations (Paper IV).





## 3 Material and methods

### 3.1 Measuring the impact on soils and surface waters acid-base status

The impact of WTH on the acid-base status was determined by measuring changes in soil exchangeable (1 M  $\text{NH}_4\text{Cl}$ )  $\text{Ca}^{2+}$  pools and surface water ANC. Calcium is an essential macronutrient for plant development (McLaughlin & Wimmer, 1999; Hepler & Wayne, 1985) and is generally the main cation of the base saturation (BS) of soils. Calcium is also used to determine the acid neutralizing capacity (ANC) of surface waters defined as the differences between strong base cations and strong anions (Reuss & Johnson, 1986). Thus,  $\text{Ca}^{2+}$  plays a key role for the nutrient status of forests and for providing buffer capacity to soils and surface waters.

Calcium is also the base cation typically affected by WTH as indicated by field studies (Thiffault *et al.*, 2011; Kreutzweiser *et al.*, 2008), mass balances (Akselsson *et al.*, 2007; Joki-Heiskala *et al.*, 2003; Rademacher *et al.*, 2001) and dynamic modelling (Aherne *et al.*, 2012).

### 3.2 Overview of study sites

The studies were undertaken at sites which belong to the ICP Integrated Monitoring programme (Paper I and III) and the long-term wood fuel experiment “HELTRAD” (Paper II and IV). The sites were chosen to represent a geographical gradient in terms of climate, past and present deposition of acid pollutants and sea-salt influence (*Figure 1*).

The Integrated Monitoring (IM) sites are part of an international program for monitoring the effects of air pollution on ecosystems (UNECE, 2015). The large number of data collected over the past circa 20 years makes the IM-sites well suited for analyses of soil and surface water acidification trends using

dynamic modelling. On the other hand, forest management has not been carried out. Therefore, simulating the impact of forest practices such as WTH can only be done in theory.

The long-term wood fuel experiment “HELTRAD” was established in the mid-seventies by Björkroth and Rosén (1977). The original purpose was to determine the effect on forest productivity after different levels of biomass removal. With time the program expanded to also include environmental studies. The tree stands have been carefully managed using modern forest management techniques. It is the best studied Swedish experiment for understanding the long-term effect of CH and WTH after final felling in boreal forests. In addition, it belongs to one of the world’s oldest replicated biofuel experiments. However, the sites are not as well described as the IM-sites and pre-treatment data is limited to soil chemistry and tree stand characteristics. Also, since the experiment was laid out as a block experiment (4 replicates), stream data are not available.



Figure 1. Location of the study sites and their affiliation to the research programmes Integrated Monitoring (Δ) and HELTRAD (●).

### 3.2.1 The Integrated Monitoring (IM) sites (Paper I and III)

The four IM-sites are located in semi-natural unmanaged forests in northern (Gammtratten), central (Kindla) and southern (Gårdsjön and Aneboda) Sweden (*Figure 1*). The sites differ in terms of climate, marine influence, past and present deposition of  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ , and  $\text{NH}_4^+$ . The temperature gradient shows a similar pattern with the shortest vegetation period at Gammtratten. Gårdsjön receives the highest annual precipitation (1000 mm) while Kindla has the second highest annual precipitation (900 mm) (Table 1).

Norway spruce (*Picea abies* (L.) Karst.) covers the majority of the IM catchments, mixed with smaller amounts of Scots pine (*Pinus sylvestris* L.) and deciduous trees, notably white birch (*Betula pubescens* Ehrh.) (Table 1). Podzolic soils are the dominant soil type, except for recharge areas where histosols, gleysols, and regosols occur. Sampling and analyses of precipitation, soils, surface waters and vegetation are carried out according to the ICP manual of methodology (ICP Integrated Monitoring, 2013), described in detail by Löfgren et al. (2011). Data necessary for the MAGIC modelling in Paper I and III were extracted from the IM database.

Table 1. General site characteristics of the IM-catchments from Löfgren et al. (2011).

	Gårdsjön	Aneboda	Kindla	Gammtratten
Latitude	58°03'N	57°05'N	59°45'N	63°51'N
Longitude	12°01'E	14°32'E	14°54'E	18°06'E
Altitude (m.a.s.l.)	114-140	210-240	312-415	410-545
Annual precipitation (mm)	1000	750	900	750
Dominant soil type	Podzols	Podzols	Podzols	Podzols
Area (ha)	3.7	18.9	20.4	45
Dominant tree species				
Norway spruce	65%	73%	83%	70%
Scots pine	14%	20%	14%	16%
White birch	17%	3%	2%	13%
Others	-	3%	-	-
Stem volume ( $\text{m}^3 \text{ha}^{-1}$ )	219	319	244	138

### 3.2.2 The long-term wood fuel experiment (Paper II and IV)

The HELTRAD experiment was laid out at four different locations (Tönnersjöheden, Kosta, Lövliden and Lund) following clear-cutting of mature coniferous forests (*Figure 1* and Table 2). After clear-cutting, the sites were replanted with Norway spruce or Scots pine. Three treatments were replicated in four blocks using a randomized block design. Each plot was 25 x 25 meter surrounded by at 5 meter buffer zone. The treatments included a) conventional

harvest of stems only (CH), b) harvest of all above-ground biomass except for needles (BSH) and c) harvest of all above-ground biomass (WTH).

In this thesis, only three out of the four sites (Tönnersjöheden, Kosta and Lövliden) and two out of the three treatments (CH and WTH) were used to assess the long-term effect on soil and soil solution acid-base status. The fourth site, Lund, was of low site quality and considered less representative for forest management. Regeneration has also been less successful at this site due to frost and snow blight fungus (*Phacidium infestans* Karst.). Furthermore, the BSH treatment plots were omitted from the soil solution sampling in 2003-10 due to restricted funding.

All sites are located on well-drained podzols on glacial till but climate, deposition pattern and soil productivity differs. In short, Lövliden is the most well buffered site in terms of soil and soil solution chemistry relative to Kosta and Tönnersjöheden. Lövliden has also received less deposition in terms of sea salts and past and present deposition of sulphate ( $\text{SO}_4^{2-}$ ), ammonium ( $\text{NH}_4^+$ ) and nitrous oxides ( $\text{NO}_x$ ). Precipitation is twice as high in Tönnersjöheden (1140 mm) in comparison to the other two sites. Soil productivity ranges from 3.8 m<sup>3</sup> stemwood per hectare and year (Lövliden) to 10.1 m<sup>3</sup> stemwood per hectare and year (Tönnersjöheden).

Table 2. *Site and stand characteristics of the HELTRAD experiment.*

	<b>Tönnersjöheden</b>	<b>Kosta</b>	<b>Lövliden</b>
<b>General site data</b>			
Latitude	56°42'N	56°52'N	64°18'N
Longitude	13°40'E	15°23'E	19°36'E
Altitude (m)	100	240	260
Annual precipitation (mm)	1040	595	565
Forest type	Mesic dwarf-shrub	Mesic dwarf-shrub	Mesic-wet dwarf-shrub
Soil type	Podzol	Podzol	Podzol
<b>Harvested stand</b>			
Tree species			
Dominant:	Norway spruce (100%)	Scots pine (70%)	Norway spruce (50%)
Co-dominant:		Norway spruce (30%)	Scots pine (50%)
Stand age (year)	70	100	155-175
Site productivity (m <sup>3</sup> ha <sup>-1</sup> yr <sup>-1</sup> )	10.1	5.9	3.8
Site index (H100)	G30	T24	G20
Harvested	Spring 1975	Fall 1975	Fall 1976
<b>New stand</b>			
Tree species	Picea abies (100%)	Pinus sylvestris (100%)	Picea abies (100%)
Planted	Spring 1976	Spring 1976	Spring 1977
Pre-commercial thinning of broadleaves	-	1983, 1989	1998
First thinning	Spring 2004	Autumn 2000	-
Second thinning	-	Autumn 2010	-
Tree diameter measurements	Autumn 1991	Autumn 1991	Spring 1991
	Spring 1997	Spring 1997	Spring 1997
	Autumn 2003	Autumn 2000	Autumn 2002
	Autumn 2007	Autumn 2008	-

### *Soil solution sampling and analyses*

In each plot, five ceramic suction cup lysimeters with a pore diameter of 0.8 µm were installed in the mineral soil at 50 cm depth beneath the ground surface. Two of the lysimeters were installed in 2002-03 as part of a pilot study. Installation of the remaining lysimeters took place in 2008. The samples were collected during spring, summer and autumn. Sampling during the winter period was not possible due to low temperatures and soil frost. The maximum number of possible samples per treatment and location were 24 (2003–2005,

pooled samples) and 100 (2008-2010, discrete samples). However, due to weather conditions and disturbances by e.g. voles and wild boars, the actual numbers of samples were somewhat lower. Furthermore, the soil solution volumes were not always sufficient to conduct a full range of analyses. Nevertheless, the sample size was sufficient to conduct statistical analysis.

A hand operated pump was used to generate an under pressure of 480 mbar. After two days, the samples were transported to the laboratory. All samples were analysed for pH and major anions and cations by accredited laboratories (IVL and SLU). Full details about the analyses can be found in Paper II. The soil solution data was used to determine the long-term (27-35 years) concentration differences between WTH and CH (Paper II). In Paper IV, soil solution data from the first sampling period (2003-05) was used to calibrate the MAGIC model.

### *Soil sampling and analyses*

Details about sampling, methods of analyses and calculations for samples collected between 1990 and 2013 can be found elsewhere (Brandtberg & Olsson, 2012; Olsson *et al.*, 1996). Briefly, the forest floor and mineral soil were collected at 25 locations within each plot. Mineral soil layers were sampled at fixed depths intervals of 0–5, 5–10, 10–15 and 15–20 cm. Forest floor and mineral soil samples were pooled into composite samples forming one sample per soil layer and plot.

Distilled water was used to determine forest floor and mineral soil pH. Concentrations of exchangeable BC,  $H^+$  and  $Al^{n+}$  were measured in a 1M  $NH_4Cl$  solution and exchangeable acidity (EA) was titrated to pH 8.2 in 0.2M KCl. Effective cation exchange capacity ( $CEC_{eff}$ ) at current soil pH was estimated by summing exchangeable base cations,  $H^+$  and  $Al^{n+}$ . Base saturation (%BS) was calculated as the equivalent sum of exchangeable bases ( $Na^+$ ,  $K^+$ ,  $Ca^{2+}$  and  $Mg^{2+}$ ) divided by  $CEC_{eff}$ .

Soil data collected in 2001-02 were included in Paper II to better interpret the soil solution results. In Paper IV, the same data was also used to calibrate the MAGIC model. To compile time-series, data from all three soil sampling campaigns in 1990-91, 2001-02 and 2012-13 were used to determine the long-term (up to four decades) impact after clear-felling.

### *Statistical analyses*

Soil solution data from 2003-05 and 2008-10 were analysed separately using a mixed model analysis (SAS, version 9.2) with fixed and random factors (Paper II). The fixed factors were site and treatment and the random effect factors were block and lysimeter.

Soil chemical differences in 2001-02 were analysed using a nested ANOVA (Statistica 4.1 for Macintosh, StatSoft Inc., 1994) to evaluate treatment effects across sites with site, treatment and block, including the interactions between sites and treatments, as sources of variation (Paper II). To examine soil chemical differences over time (1990-2013) a mixed model analysis was performed, including fixed (site, treatment and time) and interaction effects (treatment\*site, treatment\*time and site\*time) (Paper IV). To determine which treatment means that were significantly different from each other, a Tukey-Kramer post hoc test was performed. A significance level of 0.05 was used for all three tests.

### 3.3 The MAGIC model

The Model of Acidification of Groundwaters in Catchments (MAGIC, version 7) was used to simulate past and future trends in soil and surface water acid-base status. MAGIC is a lumped parameter model developed in the 1980s (Cosby *et al.*, 1985a; Cosby *et al.*, 1985b; Cosby *et al.*, 1985c) and later refined (Cosby *et al.*, 2001) to model the long-term impact of acid deposition on water chemistry. The model structure also makes it suitable to explore the impact of other forcing variables. MAGIC has become widely used for the past 30 years for studying historical and future trends in surface water acidification owing to atmospheric pollution (Moldan *et al.*, 2013; Whitfield *et al.*, 2010; Wright *et al.*, 2005) as well as the impact of other drivers such as land use changes and forestry (McDonnell *et al.*, 2013; Aherne *et al.*, 2012; Jenkins *et al.*, 1990). The model has also become a national tool for surface water acidification assessments in Sweden (Swedish Environmental Protection Agency, 2007b).

One part of the model is used for simultaneously calculating the concentrations of major ions from a set of equilibrium equations. Another part of the model calculates the flux of major ions in and out of compartments from a set of mass balance equations. The input and output of major ions must equal the rate of change in each compartment. The physical and chemical characteristics of the catchment are specified using fixed or adjusted (optimized) parameters. A description of the model and details of the equations can be found in Cosby *et al.* (2001).

The element fluxes vary over time according to user-defined scenarios. These scenarios typically involve total deposition and net vegetation uptake, but additional drivers can be added depending on the purpose of the study. In this thesis, a third driver, decomposition of above- and belowground logging residues, was added in Paper III and IV.

Calibration usually follows a standard protocol where the model is calibrated to one or more years of soil and surface water data. The calibration is done in a step-wise order beginning with strong acid anions ( $\text{SO}_4^{2-}$ ,  $\text{Cl}$  and  $\text{NO}_3^-$ ). Next, BC is calibrated by adjusting weathering rates and initial soil exchangeable fractions. Finally the weak acid-base chemistry is calibrated. Sometimes it may also be necessary to adjust other parameters to achieve a good fit between measured and modelled data.

MAGIC was applied to both the IM-sites (Paper I and III) and the HELTRAD sites (Paper IV) to model potential long-term effects of WTH and examine the robustness of modelled outcome. A brief description of the conceptualization, parametrization and calibration is given below.

### 3.3.1 Modelling the IM-sites with MAGIC (Paper I and III)

In the first paper, MAGIC was calibrated to all four IM-sites (Gammtratten, Gårdsjön, Aneboda and Kindla) to predict acidification trends in soil and stream water in semi-natural forests from 1860 (pre-industrial time) to present time. A two-box model was used to describe the average (aggregated) catchment characteristics including an upper organic and lower mineral soil layer. Elements moved into and out of these boxes on an annual time-step according to the deposition and net tree uptake patterns. Derivations of the deposition sequences were undertaken in several steps involving the use of EMEP-modelled data, measured data and assumptions about marine background concentrations in 1860. Tree uptake sequences were created for each site based on known land-use history, current forest age and measured standing biomass. Parametrizations were based on site specific data from 1996-2008, collected and analysed according to Löfgren et al. (2011). Calibrations followed the standard protocol but less effort was put into achieving a good fit between measured and modelled BC pools owing to considerable variation in soil data. However, the calibrated BC pools were representative for the regions as a whole, and therefore considered satisfactory. The final calibrated sets (SETII) were used as base scenarios for acidification assessment according to the Swedish national criteria (Swedish Environmental Protection Agency, 2007b).

The robustness of model outcomes for assessing acidification and estimates of exchangeable BC pools was then tested by varying a selection of soil parameters (Paper I). These included 1) lowering the final calibrated apparent aluminium solubility constant ( $\log_{10} K_{sp}$ ) in streams by -1 and 2) activating a pH-dependant BC weathering rate instead of using a steady-state BC weathering rate. The latter is achieved by a power function in MAGIC which allows the user to define the relationship between mineral dissolution and pH.



In this study we raised the variable  $H^+$  concentration by a fixed exponent (0.4) for all four BC, sites and soil layers. The impact of varying BC pools by changing soil depth and CEC was also compared using an alternate calibrated data set (SET I). In this dataset, soil depth was set at fixed depth of 15 cm (soil layer 1) and 85 cm (soil layer 2) and CEC was set to 100 mEq  $kg^{-1}$  for the upper soil and 25 mEq  $kg^{-1}$  for the lower soil. In addition to these tests, the outcome for acidification assessment was examined by changing the “pre-industrial year” from 1860 to 1920.

In Paper III, MAGIC was re-calibrated to three (Gammtratten, Kindla and Aneboda) out of the four IM sites. A theoretical clear-cutting and biomass removal (80% of the logging residues) was simulated in 2020 and the future effects on soil and stream water acid-base properties were simulated for one rotation period (base scenario). Uncertainties in future model predictions were then tested in nine alternate scenarios by varying a number of parameters associated with forest practices; 1) the amount of logging residues removed in final felling (0%, 60%, 80% and 100%), 2)  $Ca^{2+}$  concentrations in forest biomass (5<sup>th</sup>, 25<sup>th</sup>, 50<sup>th</sup>, 75<sup>th</sup> and 95<sup>th</sup> percentiles) and 3) site productivity (current site index  $\pm 4$  m). Parameterizations were similar to Paper I since it largely built on the same input data. However, an additional three years of data could be added to the calibration period (1996-2011) allowing for more stable aggregated values. Also, instead of a two-box model, a one-box model was tested. Efforts were also made to calibrate the soil BC pools. An extra model driver (decomposition) was also introduced after harvest. Another difference lied in how total deposition was calculated. In Paper I, total deposition of Cl was normalized against  $Na^+$  deposition. In Paper III, total deposition of Cl was simply calculated by summing bulk deposition (after correcting for dry deposition in bulk collectors; i.e. wet deposition) to dry deposition (difference between wet and throughfall deposition). Finally, small differences also existed in allocating nutrients over time depending on the assumed uptake and deposition scenarios.

### 3.3.2 Modelling the HELTRAD sites with MAGIC (Paper IV)

In Paper IV, the MAGIC model was used to calibrate the HELTRAD sites using a one-box model for each site (Tönnersjöheden, Kosta and Lövliden) and treatment (CH and WTH) to describe changes in the former and current tree stands. Soil data from 2001-02 and soil solution data from 2003-05 were used to calibrate the sites. However, since soil data have only been collected down to 20 cm depth, deeper soil data were taken from the Swedish Forest Soil Inventory database (SLU, 2015). The two soil data sets were combined to form

mass-weighted values for the entire soil column down to lysimeter depth (50 cm).

Three drivers were used; deposition, net tree accumulation and decomposition of above- and belowground logging residues. However deposition was not measured at the HELTRAD sites why data from the IM-sites and SWETHRO (The Swedish Throughfall Monitoring Network) were used. The nutrient content in above- and belowground biomass, including thinning amounts, was calculated by multiplying BC concentrations by biomass dry weight. These amounts were then allocated over time to describe tree uptake during different periods of growth. Decomposing biomass was calculated in a similar way and released over time according to weight loss functions by Hyvönen et al. (2012).

## 4 Results and Discussion

### 4.1 The impact of WTH on soil acid-base status

#### 4.1.1 Modelling results (Paper I, III and IV)

Overall, the results from the MAGIC modelling simulations at the IM- and HELTRAD sites were consistent, indicating that large losses in soil  $\text{Ca}^{2+}$  exchangeable pools had occurred throughout the 19<sup>th</sup> and 20<sup>th</sup> century (Paper I, III and IV). The predictions also suggested that these losses may continue into the future, exacerbated by more intensive harvesting.

At the three IM-sites (Paper III), the hindcast  $\text{Ca}^{2+}$  losses up until the point of the theoretical clear-cutting (2019) ranged from circa 2 to 33  $\text{kmol}_c \text{ha}^{-1}$  (or 10-47%) compared to the original soil pools in 1860 (Table 3). Large depletions in BC pools (8-87  $\text{kmol}_c \text{ha}^{-1}$  or 34-84%) were also noted in Paper I when simulating all four IM-sites for the time period 1860-1996. Much of this loss was associated with  $\text{Ca}^{2+}$  (4-45  $\text{kmol}_c \text{ha}^{-1}$  or 33-87%), which predominated over the other base cations. Similarly, the estimated losses at the HELTRAD sites during the growth of the former tree stands up until the point of clear-cutting (Tönnersjöheden: 1904-1975, Kosta: 1874-1975, Lövliden: 1810-1976) varied from 14 to 22  $\text{kmol}_c \text{ha}^{-1}$  (or 46-66%) (Paper IV).

Table 3. Mass balance equations for  $Ca^{2+}$  inputs (deposition, weathering and decomposition) and outputs (uptake and runoff) for hindcast (1860-2019) and forecast (Gammtratten 2020-2100, Kindla 2020-2085 and Aneboda 2020-2075) simulations. In the SitProd1 and 2 scenarios, the length of the rotation period increased or decreased somewhat depending on site index. Fluxes in  $kmol_c ha^{-1}$ .

	Historic	Future									
	Base	CalConc1	CalConc2	CalConc3	CalConc4	SitProd1	SitProd2	BioRem1	BioRem2	BioRem3	
<b>GAMMTRATTEN</b>											
Deposition	9.41	4.41	4.41	4.41	4.41	4.41	5	3.83	4.41	4.41	4.41
Weathering	45.76	23.17	23.17	23.17	23.17	23.17	26.03	20.31	23.17	23.17	23.17
Uptake	6.58	16.7	9.13	13.95	20.97	25.61	12.79	19.44	16.7	16.7	16.7
Decomposition	0	2.78	2.78	2.78	2.78	2.78	2.78	2.77	5.67	3.5	2.06
Runoff	51.62	18.73	22.64	20.22	16.26	12.44	23.91	14.27	21.15	19.34	18.12
<b>Mass balance:</b>	<b>-3.03</b>	<b>-5.07</b>	<b>-1.41</b>	<b>-3.8</b>	<b>-6.87</b>	<b>-7.69</b>	<b>-2.89</b>	<b>-6.8</b>	<b>-4.6</b>	<b>-4.96</b>	<b>-5.17</b>
<b>KINDLA</b>											
Deposition	16.47	6.22	6.22	6.22	6.22	6.22	6.77	5.68	6.22	6.22	6.22
Weathering	19.2	7.92	7.92	7.92	7.92	7.92	8.52	7.32	7.92	7.92	7.92
Uptake	23.47	19.33	10.52	16.17	24.29	30.92	13.18	19.73	19.33	19.33	19.33
Decomposition	0	5.77	5.77	5.77	5.77	5.77	5.77	5.77	12.46	7.44	4.1
Runoff	13.75	5.05	5.93	5.36	4.55	3.58	6.13	4.78	6.46	5.42	4.68
<b>Mass balance:</b>	<b>-1.54</b>	<b>-4.46</b>	<b>3.47</b>	<b>-1.62</b>	<b>-8.92</b>	<b>-14.58</b>	<b>1.75</b>	<b>-5.74</b>	<b>0.82</b>	<b>-3.16</b>	<b>-5.76</b>
<b>ANEBODA</b>											
Deposition	14.27	4.66	4.66	4.66	4.66	4.66	5.4	3.92	4.66	4.66	4.66
Weathering	14.4	5.04	5.04	5.04	5.04	5.04	5.76	4.32	5.04	5.04	5.04
Uptake	18.76	25.05	13.53	21.02	31.43	44.77	24.47	25.29	25.05	25.05	25.05
Decomposition	0	6.65	6.65	6.65	6.65	6.65	6.7	6.57	14	8.49	4.81
Runoff	42.53	8.22	9.18	8.56	7.66	6.4	9.54	7.07	9.12	8.46	7.97
<b>Mass balance:</b>	<b>-32.62</b>	<b>-16.93</b>	<b>-6.37</b>	<b>-13.24</b>	<b>-22.74</b>	<b>-34.82</b>	<b>-16.17</b>	<b>-17.55</b>	<b>-10.48</b>	<b>-1.533</b>	<b>-1.852</b>

These historical losses were largely explained by net tree accumulation and  $\text{Ca}^{2+}$  leaching owing to mobile  $\text{SO}_4^{2-}$  ions in deposition. At the more well-buffered sites (Gammtratten and Lövliden) naturally occurring anions ( $\text{HCO}_3^-$  and  $\text{RCOO}^-$ ) also explained the  $\text{Ca}^{2+}$  leaching losses, especially during times of low  $\text{SO}_4^{2-}$  deposition. For example, at Lövliden, the sum of  $\text{HCO}_3^-$  and  $\text{RCOO}^-$  constituted 49-95% of the anion sum between 1810 and 1976. Similar values were found at Gammtratten (49-95%) between 1860 and 2019.

The simulated results were consistent with empirical long-term studies confirming that a depletion in soil  $\text{Ca}^{2+}$  exchangeable pools has taken place in Sweden (Hallbäcken, 1992; Falkengren-Grerup *et al.*, 1987), central Europe (Jandl *et al.*, 2004) and the United States (Bedison & Johnson, 2010; Johnson *et al.*, 2008a; Johnson *et al.*, 2008b; Likens *et al.*, 1998; Richter *et al.*, 1994) during the 20<sup>th</sup> century. However, it seemed as if MAGIC over-estimated the  $\text{Ca}^{2+}$  losses at Tönnersjöheden when compared to the estimated declines between 1927 and 1984 in the same area (Hallbäcken, 1992).

In addition to these results, we found that much of the BC depletion (34-76%) occurred prior to 1910-20, a period when sulphur emissions were low (Mylona, 1996) (Paper I). Similar results were found in Papers III and IV for  $\text{Ca}^{2+}$ . This implies that the forest uptake may have been more important at these sites for the modelled declines in soil  $\text{Ca}^{2+}$  pools than acid deposition. Yet, much of the changes in cations pools between 1949 and 1985 have been attributed to acid deposition (Falkengren-Grerup *et al.*, 1987). These results indicate that more research is needed to establish the relationship between soil  $\text{Ca}^{2+}$  depletion, acid deposition and forest harvesting, considering that a pre-industrial reference condition is used to assess surface water acidification owing to acid deposition in Sweden (Swedish Environmental Protection Agency, 2007b).

The large simulated losses at the IM-sites were expected to continue into the future after a theoretical clear-cutting and removal of 80% logging residues in 2020 (base scenario, Paper III) (Table 3). Thus, by the end of the second rotation period, WTH had caused a further decline in  $\text{Ca}^{2+}$  pools by 66% (Gammtratten), 30% (Kindla) and 46% (Aneboda), despite a return of nutrients via decomposing logging residues (which caused a temporary replenishment). However, the sensitivity analysis showed that the impact of WTH varied depending on assumptions made about  $\text{Ca}^{2+}$  concentrations in above-ground tree biomass, site productivity and to a lesser degree, the amount of logging residues removed in final felling (Table 3). Nevertheless, the outcome from the sensitivity analyses generally pointed in the same direction of change as in the base scenario, suggesting that soil  $\text{Ca}^{2+}$  would continue to diminish with time.

The only exception occurred at Kindla where soil exchangeable  $\text{Ca}^{2+}$  pools were predicted to be replenished under a given set of input data (Paper III). These included 1) applying the lowest  $\text{Ca}^{2+}$  concentrations in biomass (5<sup>th</sup> percentile), 2) simulating the lowest site productivity (SI=20 m) or 3) by leaving all the logging residues on site (100%=CH) (Table 3). These results indicated that WTH may be sustainable at some sites.

In Paper IV, MAGIC was used to simulate the impact of an actual clear-cutting in 1975-76 at the HELTRAD sites with (WTH) or without biomass removal (CH). Similar to the results in Paper III, we found a temporary increase in soil  $\text{Ca}^{2+}$  pools during the first circa 15 years in the CH-plots as nutrients were returned to the soil via decomposing logging residues. At the same time tree uptake remained low. A much smaller increase was observed in the WTH-plots owing to decomposition of stumps and roots. As the nutrient uptake of the current stands began to increase, soil  $\text{Ca}^{2+}$  pools began to decline, independent of treatment. When taking into account the entire simulation period, the former and current tree stand in Tönnersjöheden had depleted the soil by 85% (CH) and 88% (WTH) between 1904 and 2013. This can be compared to Lövliden where the soil pools decreased much less (CH=57% and WTH=68%) during almost double the time (1810 to 2013). Similar losses were also found at Kosta, but for a shorter time period (1874-2013): 59% (CH) and 66% (WTH). Thus, the largest losses occurred after WTH when taking into account the entire time period. However, when only considering the time period 1990-2013, the situation was reversed with larger losses from the CH treatment.

The MAGIC modelled  $\text{Ca}^{2+}$  mass balances suggested that tree uptake dominated the fluxes between 1990 and 2013 at the HELTRAD sites (*Figure 2*). Furthermore, the overall fluxes were greater in CH-plots than in WTH-plots at all three locations (*Figure 2*). This indicated that the  $\text{Ca}^{2+}$  cycling in these forests largely depended on  $\text{Ca}^{2+}$  availability. In addition to these results, the mass balances showed that the  $\text{Ca}^{2+}$  weathering was higher in CH-plots than in WTH-plots. However,  $\text{Ca}^{2+}$  inputs via weathering were based on manual calibration and not on actual measurements. In MAGIC, weathering is considered as a source of BC and can therefore make up for underestimates of BC via e.g. deposition or other unidentified pools. Thus, weathering might not necessarily have been higher in CH-plots.

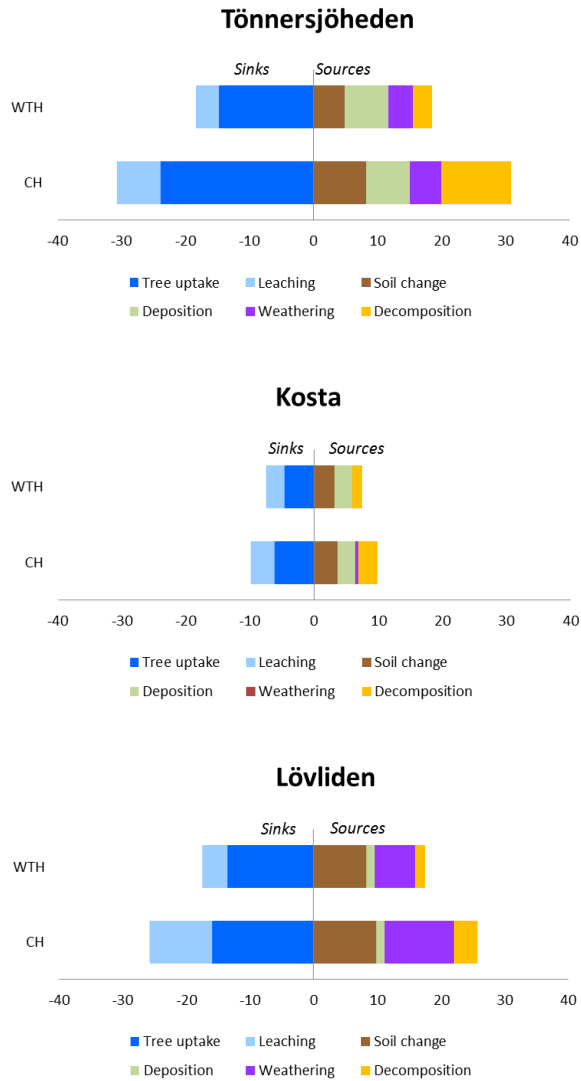


Figure 2. MAGIC modelled  $\text{Ca}^{2+}$  fluxes ( $\text{kmol}_c \text{ha}^{-1}$ ) in CH- and WTH-plots summarized over the period 1990-2013 (Tönnersjöheden), 1991-2012 (Kosta) and 1991-2013 (Lövliden). The fluxes are divided into sinks (uptake, leaching) and sources (deposition, weathering, decomposition, soil change).

The results presented here suggested that nutrient uptake and storage by trees in the past have depleted the exchangeable  $\text{Ca}^{2+}$  pools causing the soils to become more sensitive to acid deposition. Large future depletions were also to be expected following harvest in coniferous boreal forests, in particular after WTH. This is in accordance with other mass-balances (Akselsson *et al.*, 2007; Johnson & Todd, 1998; Likens *et al.*, 1998; Hornbeck *et al.*, 1990; Federer *et al.*, 1989) and dynamic modelling (McDonnell *et al.*, 2013; Aherne *et al.*, 2012; Jenkins *et al.*, 1990) results. For example, Likens *et al.* (1998) estimated a complete depletion in soil exchangeable pools of  $\text{Ca}^{2+}$  within one rotation period following WTH. Large  $\text{Ca}^{2+}$  losses were also simulated by McDonnell *et al.* (2013) who suggested, perhaps ironically, that the only way to restore soil  $\text{Ca}^{2+}$  pools would be a “*complete cessation of tree harvesting*”. We believe, however, that trees will respond to lower soil  $\text{Ca}^{2+}$  pools by adjusting their nutrient uptake (but still be able to meet their physiological need), as indicated by the MAGIC simulated asymptotic  $\text{Ca}^{2+}$  curves for Tönnersjöheden (Paper IV).

#### 4.1.2 Empirical results (Paper II and IV)

Previous studies at the HELTRAD sites by Olsson *et al.* (1996) have confirmed mass-balance (Akselsson *et al.*, 2007) and dynamic model predictions (Aherne *et al.*, 2008); namely that WTH leads to reduced amounts of exchangeable  $\text{Ca}^{2+}$  in the soil, relative to CH, at least in the medium-term. Later studies at the HELTRAD sites showed, however, that the differences between WTH and CH had decreased but not completely diminished from stand age 15 to 25 years (Brandtberg & Olsson, 2012). More specifically, treatment differences in the forest floor and uppermost mineral soil were no longer statistically significant whereas the differences in deeper mineral soil (down to 20 cm) remained. Similar results were found in Paper II when only Tönnersjöheden, Kosta and Lövliden were tested ( $\Delta\text{WTH-CH}$ : -0.29, -0.37 and -0.24  $\text{kmol}_e \text{ ha}^{-1}$  in the 5–10, 10–15 and 15–20 cm soil layer, respectively). The diminishing differences were explained by 1) greater tree uptake of  $\text{Ca}^{2+}$  in CH-plots, 2) increased  $\text{Ca}^{2+}$  weathering rates in WTH-plots and 3) greater  $\text{Ca}^{2+}$  leaching from CH-plots.

The result from the statistical analysis in Paper II also suggested that the effect of WTH varied depending on site. The most pronounced effect was found at the well-buffered site Lövliden while the treatment response at the southern sites Tönnersjöheden and Kosta were much smaller. This is in contrast to what is generally expected from Swedish authorities (Swedish Environmental Protection Agency, 2007a). One explanation might be that these two latter sites soils were richer in exchangeable  $\text{Al}^{\text{n}+}$  and therefore more resilient towards cation exchange reactions, a mechanism proposed by



Bélangier et al. (2003) and Thiffault et al. (2011). Calcium released from decomposing logging residues may therefore not have been sufficient to displace  $Al^{n+}$  ions from the soil complex. On the other hand, it is also possible that the impact of WTH has diminished over time.

Resampling of the soils in 2012-13 (Paper IV) made it possible to determine whether or not the differences between CH and WTH had continued to decrease or if they still persisted in the deeper mineral soil layer, four decades after clear-cutting. The results confirmed the observed trend from 1990-91 to 2001-02, i.e. that the soil exchangeable  $Ca^{2+}$  pools have continued to decrease, independent of treatment from stand age of circa 25 years to 37-38 years (Figure 3). However, the sites responded differently as indicated by a significant time\*site interaction.

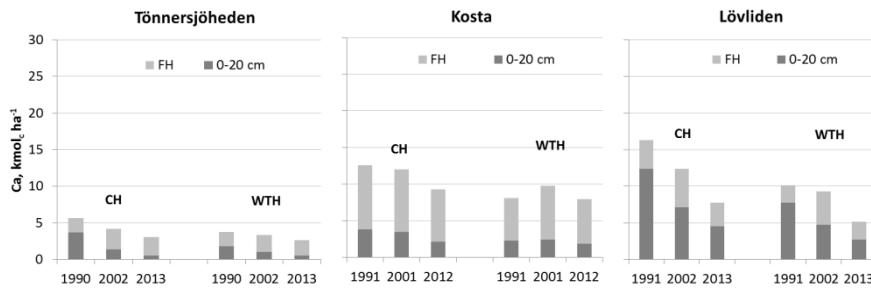


Figure 3. Exchangeable  $Ca^{2+}$  pools in the FH and 0-20 cm soil layer.

The measured  $Ca^{2+}$  losses for the whole period (1990-2013) ranged from 0.2 to 8.6  $kmol_c ha^{-1}$ . In addition, faster soil  $Ca^{2+}$  depletion was observed at the CH-plots (2.6-8.6  $kmol_c ha^{-1}$ ) compared with WTH-plots (0.2-5.0  $kmol_c ha^{-1}$ ). This implies that soil  $Ca^{2+}$  pools have become more similar with time as indicated by a near significant time\*treatment effect ( $p=0.0626$ ). These results were in accordance with the latest tree diameter measurements from 2002 (Tönnersjöheden), 2007 (Lövliden) and 2008 (Kosta) which showed that biomass differences were still present at these points in time (unpublished data). Nevertheless, soil  $Ca^{2+}$  pools still remained significantly lower in the WTH-plots. Whether or not the  $Ca^{2+}$  pools in the CH-plots will continue to decrease at a faster rate and eventually become similar or lower than the  $Ca^{2+}$  pools in the WTH-plots remains to be studied. However, should the current depletion rates in CH-plots continue, soil pools would become comparable within a near future, at least at Lövliden and Tönnersjöheden (Figure 3).

The measured declines in  $Ca^{2+}$  soil pools were greater than what has been found in other studies and were largely explained by the high tree  $Ca^{2+}$  net uptake and high soil  $Ca^{2+}$  availability. But, despite these large  $Ca^{2+}$  losses, tree

growth and vitality has not yet been impaired. In addition, the largest losses were observed at the northern well-buffered site Lövlieden, where CH and WTH is less likely to lead to soil acidification. At the two southern sites (Tönnersjöheden and Kosta), soils were more acid but CH and WTH have not made them more acidic according to the current Swedish soil acidification classification system (Swedish Environmental Protection Agency, 1999).

The measured losses between 1990 and 2013 were however smaller than what MAGIC predicted for the same time period; 3.6-9.9 kmol<sub>c</sub> ha<sup>-1</sup> (CH) and 3.1-8.3 kmol<sub>c</sub> ha<sup>-1</sup> (WTH). Thus, the decline in Ca<sup>2+</sup> pools was verified, but the model exaggerated the depletion. However, both the measured and predicted estimates suggested that the Ca<sup>2+</sup> losses were greater following CH than WTH although the model failed to predict the rate by which the soil Ca<sup>2+</sup> pools have become more similar over time.

The discrepancy between modelled and measured impact of WTH was likely due to an inability to understand and incorporate biological feed-back mechanisms in the scenario making. These may include an underestimation of the available soil Ca<sup>2+</sup> by neutral salt extractions (Vadeboncoeur *et al.*, 2014; Yanai *et al.*, 2005), diffusion from deeper soil layers (Grigal & Ohmann, 2005), development of a deeper root system (Dijkstra & Smits, 2002) or direct uptake of Ca<sup>2+</sup> from the atmosphere by the foliage (van der Heijden *et al.*, 2014; Berger *et al.*, 2006). Recently, van der Heijden (2015) also put forward the hypothesis of an internal Ca<sup>2+</sup> oxalate tree pool. It has also been shown that Norway spruce can mobilize Ca<sup>2+</sup> in the topsoil via acidification (Berger *et al.*, 2006). Additionally, the ability of trees to increase weathering fluxes through the association with “rock-eating” fungi has been discussed (Blum *et al.*, 2002; Jongmans *et al.*, 1997). According to this theory, uptake of nutrients will increase via ectomycorrhizal symbiosis when trees show signs of depletion. Not including ignored soil pools and/or a biotic control in mass balances or dynamic modelling may lead to a risk for exaggerating the impact of WTH. We believe that future research should aim at identifying and quantifying overlooked Ca<sup>2+</sup> sources and potential biotic control mechanisms.

## 4.2 The impact of WTH on soil solution and stream water acid-base status (Paper I, II and III)

### 4.2.1 Modelling results (Paper I and III)

The MAGIC modelling results (hindcast scenarios) for the IM-sites Gammtratten, Kindla and Aneboda, indicated that all three streams started with positive ANC, which gradually declined from 1860 to late 1980s and early 1990s (Paper III) (*Figure 4*). After that, MAGIC predicted a recovery in ANC,

but not a return to pre-industrial levels by the end of the simulation period. The overall change in ANC was closely related to changes in  $\text{SO}_4^{2-}$  deposition and stream water  $\text{SO}_4^{2-}$  concentrations (data not shown). Similar trends in ANC were found in Paper I when modelling all four sites (including Gårdsjön). These results were consistent with the significant increase in stream ANC documented at these sites (Löfgren *et al.*, 2011). The results were also in agreement with the general recovery from acidification in surface water demonstrated for a large number of lakes and streams across Europe (Garmo *et al.*, 2014; Evans *et al.*, 2001).

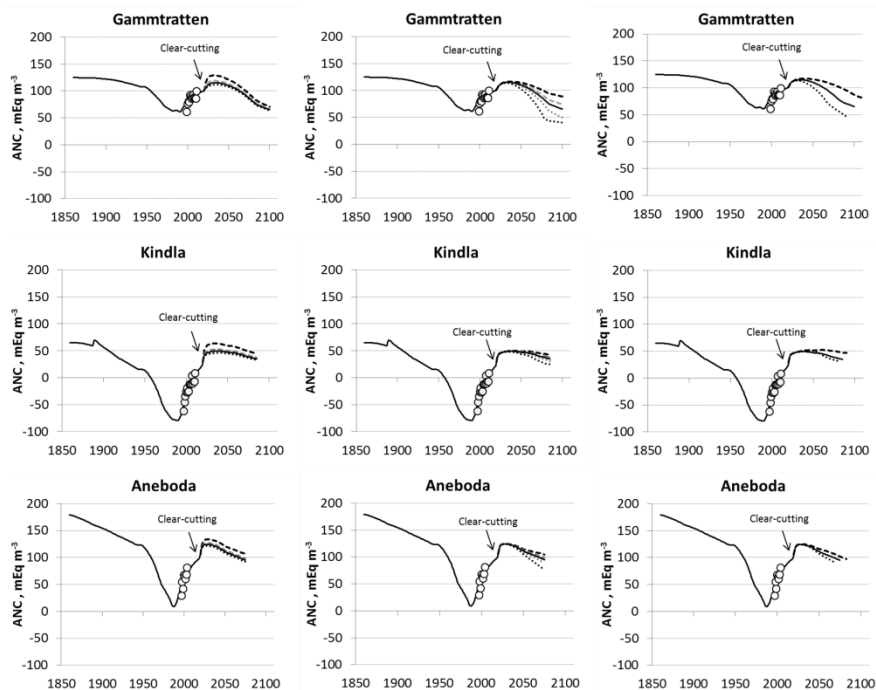


Figure 4. Measured (white circles) and modelled streamwater ANC (solid black line) under the base scenario along with the nine alternate scenarios. Left panel: varying biomass (BioRem1–3); middle panel: varying  $\text{Ca}^{2+}$  concentration in above-ground biomass (CalConc1–4) and right panel: varying site productivity (SitProd1–2). Black dashed line= BioRem1, CalConc1 and SitProd1, 1; grey dashed line = BioRem2 and CalConc2; grey dotted line = BioRem3 and CalConc3, black dotted line= BioRem4, CalConc4 and SitProd2. Note that the minimum age of cutting differs between the base, SitProd1 and SitProd2 scenarios. In 2020, the virtual clear-cutting takes place (arrow).

In Sweden, hindcast predictions by MAGIC are used to classify lakes as acidified if there has been a change in pH by more than 0.4 units from a pre-industrial state (1860) to present time (Fölster *et al.*, 2007; Swedish Environmental Protection Agency, 2007b). The estimated change in ANC ( $55\text{--}211 \text{ mEq m}^{-3}$ ) between 1860 and 2000 corresponded to a pH-change between 0.4 and 1.4 units (Paper I). Thus, prior to the theoretical clear-cutting in 2020 (Paper III) all four streams were classified as acidified, despite the increase in pH and ANC that occurred after 1990 (Paper I). However, the predicted response in  $\Delta\text{ANC}$  and  $\Delta\text{pH}$  changed when a selection of key input parameters were varied. These included e.g. lowering the aluminium solubility in stream

and letting BC weathering rates to change depending on soil acidity (Paper I). Changing the apparent aluminium solubility constant ( $\log_{10} K_{sp}$ ) in streams increased  $\Delta\text{pH}$  to 0.9-1.7 units, suggesting that the streams would have been more severely affected by anthropogenic acidification in the past. On the other hand, releasing more BC when soil acidity increased lowered stream  $\Delta\text{pH}$  to 0.3-1.2 units. The lower value ( $\Delta\text{pH}=0.3$ ) belonged to Gammtratten. Thus, by including a weathering feedback in MAGIC, Gammtratten would no longer be classified as acidified according to the current system (Paper I). A similar result was achieved when the pre-industrial reference year (1860) was postponed to 1920.

One of the consequences of using input data associated with large uncertainties is that streams or lakes could be misclassified as acidified and thereby wrongfully subjected to liming. Alternatively, streams or lakes in need of liming may be overlooked. Thus using site-specific data based on long-term monitoring is important for our ability to accurately predict trends in surface waters. However, the results in Paper I showed at the same time that using another input data set (SET I) resulted in comparable estimates in  $\Delta\text{pH}$  and  $\Delta\text{ANC}$  although the initial BC soil pools were overestimated by a factor of 2. Therefore, putting efforts into soil sampling to reduce a potential large standard deviation in catchment BC pools may not necessarily lead to more accurate predictions in stream pH and ANC. These results also showed that substantial changes may occur in the soil without causing a large impact on surface water acid-base status (Paper I).

In 2020, the IM-catchments were theoretically clear-cut and replanted with Norway spruce, leaving 20% of the above-ground logging residues to decompose (base scenario, Paper III) (*Figure 4*). During the first circa 5-10 years after felling, this caused a further increase in ANC. But, as the contribution from decomposing residues started to decline and the nutrient demand of the forest increased, ANC began to decrease. By the end of the second rotation period (forecast scenario), WTH had led to a reversal of the positive trend in ANC at Gammtratten, Kindla and Aneboda. The magnitude of impact on stream ANC varied, however, depending on site and the concentration of mobile strong acid anions. Contrary to common beliefs, the largest decrease in modelled ANC was observed at the well-buffered site Gammtratten. The effects at Kindla and Aneboda were much more limited and not large enough to offset the general recovery from acidification.

Variations in the future possible impact of WTH were examined in nine alternate scenarios (see Chapter 3.3.1) by changing a set of input parameters related to forestry (Paper III). The results showed that modelled stream ANC varied the most when changing the  $\text{Ca}^{2+}$  concentrations in tree biomass (*Figure*

4). Site productivity was the second most important variable while harvesting different amount of logging residues (0-100%) only marginally affected the results. The results showed that the reliability of modelled outcome would increase by using site-specific  $\text{Ca}^{2+}$  concentrations in tree biomass and field determined identification of site productivity.

Overall, the results from Paper I and III indicated that the impact on stream ANC was much less than the impact on soil exchangeable  $\text{Ca}^{2+}$  pools. Similar results were found by Jenkins et al (1990). These modelled results were also well in agreement with the long-term measured impact of WTH on  $\text{Ca}^{2+}$  and ANC in the soil solution at the HELTRAD sites (Paper II).

#### 4.2.2 Empirical results (Paper II)

Results from the first soil solution sampling at the HELTRAD sites in 2003-05 (27-30 years after harvest) demonstrated few statistically significant differences between CH and WTH (Paper II, Table 4). The greatest treatment difference was noted for  $\text{Ca}^{2+}$ , which was significantly lower ( $17 \mu\text{Eq l}^{-1}$  or 40%) in WTH-plots compared with CH at 50 cm soil depth. These results agreed well with the observed differences in  $\text{Ca}^{2+}$  concentrations and pools in deeper mineral soil layers (down to 20 cm) from 2001-02 (Paper II and IV). In addition to these results, ANC differed in the same order of magnitude ( $16 \mu\text{Eq l}^{-1}$ ) as  $\text{Ca}^{2+}$  and pH was slightly lower after WTH (-0.05 units).

Table 4. Average soil solution chemistry and results from the statistical analysis. Bold numbers indicate statistically significant differences at  $p \leq 0.05$ .

	Treatments		Test of fixed effects					
	CH	WTH	Main effects			Interaction effects		
			Site	Treatment	Season	Site x treatment	Site x season	Treatment x season
<b>2003-2005</b>								
pH	4.92 (0.63)	4.87 (0.66)	<.0001	<b>0.0295</b>	0.5675	0.5539	0.6249	0.7224
ANC ( $\mu\text{eq l}^{-1}$ )	9 (68)	-7 (62)	<b>0.0001</b>	<b>0.0191</b>	<.0001	<b>0.0233</b>	0.0657	0.0775
DOC ( $\text{mg l}^{-1}$ )	5 (2)	4 (2)	<b>0.0142</b>	0.3772	<b>0.0002</b>	0.2561	0.9577	0.1968
Ca <sup>2+</sup> ( $\mu\text{eq l}^{-1}$ )	43 (34)	26 (18)	<b>0.0095</b>	<b>0.0123</b>	0.4139	0.5104	<b>0.0086</b>	0.4980
Mg <sup>2+</sup> ( $\mu\text{eq l}^{-1}$ )	24 (9)	19 (10)	0.5105	0.0877	<b>0.0001</b>	0.5214	0.2659	0.3606
Na <sup>+</sup> ( $\mu\text{eq l}^{-1}$ )	152 (117)	138 (118)	<b>0.0001</b>	0.9257	<b>0.0001</b>	0.6910	0.7123	0.6361
K <sup>+</sup> ( $\mu\text{eq l}^{-1}$ )	8 (4)	9 (8)	0.5014	0.8609	0.6329	<b>0.0484</b>	<b>0.0013</b>	0.7882
SO <sub>4</sub> <sup>2-</sup> ( $\mu\text{eq l}^{-1}$ )	91 (47)	76 (52)	<b>0.0028</b>	0.1880	<b>0.0063</b>	0.0999	0.1042	0.3628
Cl <sup>-</sup> ( $\mu\text{eq l}^{-1}$ )	124 (112)	118 (121)	<.0001	0.2370	<b>0.0253</b>	0.4377	0.8635	0.0689
RCOO <sup>-</sup> ( $\mu\text{eq l}^{-1}$ )	37 (19)	30 (16)	0.1422	0.287	<b>0.0001</b>	0.2392	0.9753	0.1584
HCO <sub>3</sub> <sup>-</sup> ( $\mu\text{eq l}^{-1}$ ) <sup>1</sup>	60 (24)	43 (11)	-	0.2724	0.1256	-	-	0.1402
Alt ( $\mu\text{g l}^{-1}$ )	403 (277)	396 (349)	<.0001	0.2375	<b>0.0013</b>	0.8406	<b>0.0136</b>	0.4070
Alo ( $\mu\text{g l}^{-1}$ )	141 (158)	109 (108)	<.0001	0.482	<b>0.0023</b>	0.1572	<.0001	0.6267
Ali ( $\mu\text{g l}^{-1}$ )	311 (163)	336 (240)	<.0001	0.7709	0.0538	0.5066	<b>0.0158</b>	0.7859

	Treatments		Test of fixed effects					
	CH	WTH	Main effects			Interaction effects		
			Site	Treatment	Season	Site x reatment	Site x season	Treatment x season
<b>2008-2010</b>								
pH	5.0 (0.7)	5.2 (0.6)	<.0001	0.7264	0.2681	0.4009	<.0001	0.2969
ANC ( $\mu\text{eq l}^{-1}$ )	54 (68)	43 (60)	<b>0.0166</b>	0.4552	<.0001	0.0958	<.0001	0.1972
DOC ( $\text{mg l}^{-1}$ )	8 (13)	5 (4)	<b>0.0002</b>	0.1238	<.0001	0.0610	0.5248	0.8930
Ca <sup>2+</sup> ( $\mu\text{eq l}^{-1}$ )	47 (32)	37 (23)	<b>0.0023</b>	0.0573	<.0001	<b>0.0282</b>	<.0001	0.3077
Mg <sup>2+</sup> ( $\mu\text{eq l}^{-1}$ )	23 (15)	24 (18)	0.3387	0.5818	<.0001	0.6326	<.0001	0.9265
Na <sup>+</sup> ( $\mu\text{eq l}^{-1}$ )	143 (143)	124 (101)	<.0001	0.2862	<.0001	0.0847	<b>0.0145</b>	0.2378
K <sup>+</sup> ( $\mu\text{eq l}^{-1}$ )	14 (22)	14 (16)	0.6540	0.4017	<.0001	0.4168	0.5934	0.1912
SO <sub>4</sub> <sup>2-</sup> ( $\mu\text{eq l}^{-1}$ )	68 (50)	68 (53)	<.0001	0.3101	<b>0.0168</b>	<b>0.0089</b>	<b>0.0019</b>	0.8247
Cl <sup>-</sup> ( $\mu\text{eq l}^{-1}$ )	105 (145)	88 (109)	<.0001	0.2508	<.0001	0.0937	<b>0.0060</b>	0.1174
RCOO <sup>-</sup> ( $\mu\text{eq l}^{-1}$ )	60 (73)	43 (28)	<b>0.0013</b>	0.1637	<.0001	0.072	0.4052	0.9681
HCO <sub>3</sub> <sup>-</sup> ( $\mu\text{eq l}^{-1}$ ) <sup>1</sup>	86 (34)	75 (23)	-	0.1683	<0.001	-	-	0.7029
Alt ( $\mu\text{g l}^{-1}$ )	407 (481)	358 (426)	<.0001	0.7034	<b>0.0028</b>	0.4762	<.0001	0.1498
Alo ( $\mu\text{g l}^{-1}$ )	205 (310)	131 (204)	<.0001	0.4443	0.2264	<b>0.0166</b>	0.1462	0.2106
Ali ( $\mu\text{g l}^{-1}$ )	179 (201)	219 (345)	<.0001	0.8526	0.1566	0.7096	<.0001	0.2898



Despite these measured differences, ANC remained positive at the well-buffered northern site Lövliden, independent of treatment (CH=111  $\mu\text{Eq l}^{-1}$ ; WTH= 67  $\mu\text{Eq l}^{-1}$ ). Therefore, at this site, the loss of  $\text{Ca}^{2+}$  and the subsequent decline in ANC following WTH was not likely to have caused adverse effects on aquatic biota in receiving streams or lakes. The two southern sites were more acidic in terms of ANC (-28 to -10  $\mu\text{Eq l}^{-1}$  at Tönnersjöheden and -58 to -24  $\mu\text{Eq l}^{-1}$  at Kosta depending on treatment). At these sites, WTH was expected to have a large impact on the acid-base status. However when comparing the treatment differences to the general increase in ANC following the natural recovery in this area, the small difference between CH and WTH almost became negligible. For example, the difference in ANC between CH and WTH at Tönnersjöheden was less than 8  $\mu\text{Eq l}^{-1}$ . The difference at Kosta was slightly higher (12  $\mu\text{Eq l}^{-1}$ ). These figures can be compared to the general increase in ANC (9.6-176  $\mu\text{Eq l}^{-1}$ ) recorded in 10 small forest streams in southern Sweden (Löfgren *et al.*, 2009). A similar positive trend (2-20  $\mu\text{Eq l}^{-1}$ ) in the soil solution have also been shown for 32 out of 68 monitoring sites in the same region (Löfgren & Zetterberg, 2011). This implies that the long-term effect of WTH was not likely to have fully counterbalanced the natural recovery from acidification during the study period at these sites. Similar results were found in Paper III when modelling the IM-sites. However, acidification effects prior to the study period (<27 years) cannot be ruled out.

The results from the first soil solution sampling thus showed that WTH caused the soil solution to be more acidic. However, the impact on  $\text{Ca}^{2+}$  concentrations was only temporary as the results from the second soil solution sampling in 2008-10 (32-35 years after harvest) showed no main treatment response (although the difference was nearly significant;  $p=0.0573$ ). There was, however, an interaction effect between site and treatment which indicated still present effects on the  $\text{Ca}^{2+}$  concentrations at the northern well-buffered site Lövliden. Hence, the effect of WTH on the soil solution composition was not only a function of time but also of site and can therefore be expected to last longer at some sites.

To find the underlying cause for the differences in soil solution  $\text{Ca}^{2+}$  concentrations, we examined the anion composition as it has been shown that the mobility of BC depend on the concentrations of mobile anions (Reuss & Johnson, 1986). Knowing how these mobile anions form, we could gain insight into the processes which caused lower  $\text{Ca}^{2+}$  leaching in the WTH-plots.

At the well-buffered site Lövliden, biological processes appeared to be important for regulating the concentrations of anion and cations. At this site, mineralisation of organic matter was probably higher in the CH-plots as indicated by higher concentrations of organic acids ( $\text{RCOO}^-$ ), bicarbonate

( $\text{HCO}_3^-$ ) and  $\text{SO}_4^{2-}$  in these plots (Figure 5). It is also possible that desorption of previously adsorbed sulfate was higher. Together, these anions seemed to be responsible for the greater  $\text{Ca}^{2+}$  leaching from the CH-plots. The same line of reasoning was used at the two southern sites Tönnersjöheden and Kosta, although the interpretation was more difficult due to the small differences in  $\text{Ca}^{2+}$  concentrations between CH and WTH. However, it was argued that since  $\text{HCO}_3^-$  was lacking at both sites, differences in  $\text{Ca}^{2+}$  concentrations were mainly due to higher concentrations of  $\text{SO}_4^{2-}$  (Tönnersjöheden) and  $\text{RCOO}^-$  (Kosta) from the CH-plots, resulting from higher mineralisation and/or desorption of sulfate.

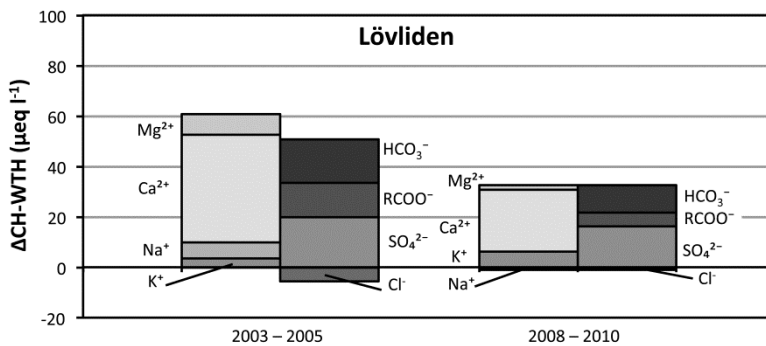


Figure 5. The difference in soil solution concentrations of major cations and anions between conventional harvest (CH) and whole-tree harvest (WTH) at Lövliden. Positive values indicate higher ion concentrations in CH-plots and negative values higher ion concentrations in WTH-plots.

Overall, the field measurement and modelling results showed that the effect of WTH on surface-water acidity depended on the concentration of mobile anions. The results suggest that practicing WTH in times of low acid deposition does not cause long-term acidification (low ANC and pH) in surface waters except for areas with naturally occurring strong acid mobile anions ( $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$  and  $\text{Cl}^-$ ). Other areas are generally well-buffered by  $\text{HCO}_3^-$  (circumneutral pH) or weak organic acids (low pH).

## 5 Conclusions

- MAGIC simulations at the IM and HELTRAD sites suggested that tree growth and net storage of  $\text{Ca}^{2+}$  in tree biomass caused large depletions in soil exchangeable  $\text{Ca}^{2+}$  pools during the 19<sup>th</sup> and 20<sup>th</sup> centuries, exacerbated by acid deposition. The impact of tree growth and acid deposition also caused streams to become acidified in terms of ANC and pH.
- The modelled depletions in soil exchangeable  $\text{Ca}^{2+}$  pools and the decrease in ANC were expected to continue to decline in the future at the IM-sites following a theoretical clear-cutting and biomass removal (WTH and CH), despite a temporary recovery during the first circa 5-10 years after clear-cutting.
- In all MAGIC future simulations, the impact of WTH was larger than after CH. However, varying a selection of key input parameters had a large impact on the WTH modelled outcome although the results generally pointed in the same direction of change as in the WTH base scenario.
- Field measurements at the HELTRAD sites confirmed that WTH and CH caused large depletions in soil exchangeable  $\text{Ca}^{2+}$  pools, two to four decades after clear-cutting. Despite rapidly diminishing differences between WTH and CH, soil  $\text{Ca}^{2+}$  pools remained lower in plots where logging residues had been harvested. Soil solution measurements also showed that WTH caused a temporary but site-dependant decrease in ANC.
- Despite rapidly declining soil  $\text{Ca}^{2+}$  pools, tree growth and vitality has not been impaired at the HELTRAD sites. Nor did the measured differences in ANC between CH and WTH offset the natural recovery at the southern sites Kosta and Tönnersjöheden. At the well-buffered northern site, Lövliden, the ecological impact of WTH on ANC was considered insignificant.
- A comparison between modelled and measured soil  $\text{Ca}^{2+}$  pools indicated that MAGIC exaggerated the losses at the HELTRAD sites. Furthermore, MAGIC could not reproduce the rapidly diminishing differences between

CH and WTH. Uncertainties in model parameters, underestimation of soil  $\text{Ca}^{2+}$  pools or a lack of understanding biological feed-back mechanisms could explain this discrepancy. Until these have been resolved, interpretations of  $\text{Ca}^{2+}$  changes related to CH or WTH using dynamic modelling should be done with caution.

## 6 Possible implications for the society and future research needs

Sustainable forest management aims to achieve a balance between society's need for forest products and the long-term health and productivity of the forest ecosystem. For the past decades, WTH has become a common practice in Sweden and will no doubt continue to play an important role in the future. However, the use of WTH poses several threats to the ecosystem which may not be compatible with sustainable forest management, relative to CH. This thesis has provided valuable insights into the long-term effect of WTH on soils and surface water acid-base status in coniferous boreal forests, up to four decades after stand development. The practical implication of these results and need for future research is listed below.

- The results can provide significant input in efforts to identify regions in Sweden where WTH can be carried out without compromising the long-term (several harvest cycles) soils  $\text{Ca}^{2+}$  supply or adversely affect the acid-base status of soils and surface waters.
- According to our results, it would appear as if WTH can be practiced in large parts of Sweden. There are several reasons for this conclusion. First, despite large variations in site characteristics, marine influence and past and present deposition of  $\text{SO}_4^{2-}$ , the long-term impact of WTH on surface water ANC was relatively small, especially at the southern sites. The effect at the northern sites were larger but considered ecologically insignificant due to bicarbonate buffering. As long as the concentration of strong mobile anions remains low, the supply of  $\text{Ca}^{2+}$  (and other BC) is large enough to maintain reasonably high ANC at the investigated sites. Still, temporary short-term decreases in ANC cannot be ruled out due to e.g. large inputs of sea salts during heavy storms or an increase in  $\text{NO}_3^-$  concentrations following clear-cutting, storm felling or insect outbreaks. Second, WTH did not make the

soils at the HELTRAD sites more acidic according to the Swedish soil acidification classification system, despite the impact on soil  $\text{Ca}^{2+}$  exchangeable pools. Third, earlier studies at the HELTRAD sites have shown that tree growth and health were not impaired by the soil exchangeable  $\text{Ca}^{2+}$  losses according needle analysis and unpublished tree diameter measurements. On the contrary, the trees were still able to take up  $\text{Ca}^{2+}$  at concentrations that far exceeded their optimal needs. These results suggest that WTH can be carried out over large parts of Sweden without compromising tree health and productivity or lead to acidification effects on soils and waters. We do, however, recognize that WTH may lead to other unwanted effects such as a reduction of decaying dead wood in the forests, which may reduce the biodiversity and put future limitations to the use of logging residues.

- The results can also be used to quantify to what extent WTH should be compensated for by ash-recycling to maintain tree health and productivity as well as counteracting soil and surface water acidification. At present, the Swedish Forest Agency (2008) recommends a low dose (3 tonnes  $\text{ha}^{-1}$ , and on occasion 6 tonnes  $\text{ha}^{-1}$  per rotation period) of ash to mitigate acidification effects caused by the additional removal of base cations during WTH. However, long-term forest liming studies have shown that the downward movement of added  $\text{Ca}^{2+}$  is very slow which is why improvements in soils acid-base status so far has been restricted to the uppermost soil layers (Löfgren *et al.*, 2009). Furthermore, ash recycling on mineral soils may not necessarily counteract surface water acidification during periods when the concentrations of strong mobile anions are low. At the current rate of natural recovery from acidification in streams, ash recycling may seem redundant in the short-term. From a socioeconomic view, ash should rather be used as a fertilizer on peat soils (Ekvall *et al.*, 2014). In terms of maintaining tree health and productivity, previous studies at the HELTRAD sites by Egnell (2011) have shown that the trees are limited by nitrogen rather than BC. From these considerations, ash recycling at the HELTRAD sites is not required. Over several harvesting cycles though, ash recycling could however become necessary. Furthermore, practicing WTH on thin, base poor soils may require compensatory measures.
- Future research should focus on identifying and quantifying overlooked  $\text{Ca}^{2+}$  sources and potential biotic control mechanisms so that uncertainties in mass balances and dynamic modelling can be reduced. A number of ignored sources have been proposed including an underestimation of soil exchangeable  $\text{Ca}^{2+}$  pools from common extraction methods. However, as

tree net uptake was identified as the major cause for soil  $\text{Ca}^{2+}$  depletion, attention should be given to examining how trees exert control of their nutrient uptake. This can be achieved by means of e.g. isotopic tracing experiment using enriched  $^{44}\text{Ca}$  to better understand how  $\text{Ca}^{2+}$  is cycled through the soil and the vegetation.

- There is a lack of long-term empirical studies where the effect of CH and WTH can be compared (Thiffault *et al.*, 2011). The HELTRAD experiment is one of few European experiments, and the only on-going experiment in Sweden. This experiment has provided valuable and unique insight into the long-term impact of WTH relative to CH in Swedish coniferous boreal forests. The results have also helped shaping policies for sustainable forest management. It is important that funding can be provided to maintain this type of long-term experiment so that the impact of WTH over an entire rotation period can be studied.
- Although the HELTRAD sites offer many future possibilities to explore the impact of WTH there are some limitations which should be considered. For example, all of the logging residues were removed in the WTH-plots. This is higher than what is generally recommended (80%) by the Swedish Forest Agency (2008). In reality, there is always some slash left on site why the impact of WTH on soil  $\text{Ca}^{2+}$  exchangeable pools may be lower than expected from this thesis. Also, the trees were planted at a stand density of circa 3 300 trees per hectare which is higher than what is currently practiced. This could have led to greater depletion of  $\text{Ca}^{2+}$  via net nutrient uptake and an exaggeration of the impact of WTH. In addition, the block experiment did not include uncut mature trees why changes over time owing to other stressors cannot be separated from that of CH and WTH. Finally, since the experiment was primarily designed to measure differences in tree productivity, pre-treatment data for other measures than soil and stands characteristics are lacking. To ensure that the results from the HELTRAD sites are reproducible, new similar experiments should therefore be established.





## References

- Achat, D.L., Deleuze, C., Landmann, G., Pousse, N., Ranger, J. & Augusto, L. (2015). Quantifying consequences of removing harvesting residues on forest soils and tree growth – A meta-analysis. *Forest Ecology and Management*, 348, pp. 124-141.
- Aherne, J., Posch, M., Forsius, M., Lehtonen, A. & Härkönen, K. (2012). Impacts of forest biomass removal on soil nutrient status under climate change: a catchment-based modelling study for Finland. *Biogeochemistry*, 107(1), pp. 471-488.
- Aherne, J., Posch, M., Forsius, M., Vuorenmaa, J., Tamminen, P., Holmberg, M. & Johansson, M. (2008). Modelling the hydro-geochemistry of acid-sensitive catchments in Finland under atmospheric deposition and biomass harvesting scenarios. *Biogeochemistry*, 88(3), pp. 233-256.
- Akselsson, C., Westling, O., Sverdrup, H., Holmqvist, J., Thelin, G., Uggla, E. & Malm, G. (2007). Impact of Harvest Intensity on Long-Term Base Cation Budgets in Swedish Forest Soils. *Water, Air, & Soil Pollution: Focus*, 7(1), pp. 201-210.
- Bedison, J.E. & Johnson, A.H. (2010). Seventy-Four Years of Calcium Loss from Forest Soils of the Adirondack Mountains, New York. *Soil Science Society of America Journal*, 74(6), pp. 2187-2195.
- Bélanger, N., Pare, D. & Yamasaki, S.H. (2003). The soil acid-base status of boreal black spruce stands after whole-tree and stem-only harvesting. *Canadian Journal of Forest Research*, 33(10), pp. 1874-1879.
- Belleau, A., Brais, S. & Paré, D. (2006). Soil Nutrient Dynamics after Harvesting and Slash Treatments in Boreal Aspen Stands. *Soil Science Society of America Journal*, 70(4), pp. 1189-1199.
- Berger, T.W., Swoboda, S., Prohaska, T. & Glatzel, G. (2006). The role of calcium uptake from deep soils for spruce (*Picea abies*) and beech (*Fagus sylvatica*). *Forest Ecology and Management*, 229(1-3), pp. 234-246.
- Björkroth, G. & Rosén, K. (1977). *Biomassa och näringsmängder på fyra ståndorter*. (PHU rapport, 49). Umeå.
- Blum, J.D., Klaue, A., Nezat, C.A., Driscoll, C.T., Johnson, C.E. & Siccama, T.G. (2002). Mycorrhizal weathering of apatite as an important calcium source in base-poor forest ecosystems. *Nature*, 417, pp. 729-731.

- Brandtberg, P.-O. & Olsson, B.A. (2012). Changes in the effects of whole-tree harvesting on soil chemistry during 10 years of stand development. *Forest Ecology and Management*, 277, pp. 150-162.
- Cosby, B.J., Ferrier, R.C., Jenkins, A. & Wright, R.F. (2001). Modelling the effects of acid deposition: refinements, adjustments and inclusion of nitrogen dynamics in the MAGIC model. *Hydrology and Earth System Sciences*, 5(3), pp. 499-517.
- Cosby, B.J., Hornberger, G.M., Galloway, J.N. & Wright, R.F. (1985a). Modeling the Effects of Acid Deposition - Assessment of a Lumped Parameter Model of Soil-Water and Streamwater Chemistry. *Water Resources Research*, 21(1), pp. 51-63.
- Cosby, B.J., Hornberger, G.M., Galloway, J.N. & Wright, R.F. (1985b). Time Scales of Catchment Acidification. *Environmental Science & Technology*, 19(12), pp. 1144-1149.
- Cosby, B.J., Wright, R.F., Hornberger, G.M. & Galloway, J.N. (1985c). Modeling the Effects of Acid Deposition - Estimation of Long-Term Water-Quality Responses in a Small Forested Catchment. *Water Resources Research*, 21(11), pp. 1591-1601.
- Dijkstra, F.A. & Smits, M.M. (2002). Tree species effects on calcium cycling: The role of calcium uptake in deep soils. *Ecosystems*, 5(4), pp. 385-398.
- Durand, P., Neal, C., Lelong, F. & Didonlescot, J.F. (1992). Effects of Land-Use and Atmospheric Input on Stream and Soil Chemistry - Field Results and Long-Term Simulation at Mont-Lozere (Cevennes-National-Park, Southern France). *Science of the Total Environment*, 119, pp. 191-209.
- Egnell, G. (2011). Is the productivity decline in Norway spruce following whole-tree harvesting in the final felling in boreal Sweden permanent or temporary? *Forest Ecology and Management*, 261(1), pp. 148-153.
- Egnell, G. & Leijon, B. (1999). Survival and growth of planted seedlings of *Pinus sylvestris* and *Picea abies* after different levels of biomass removal in clear-felling. *Scandinavian Journal of Forest Research*, 14(4), pp. 303-311.
- Egnell, G. & Valinger, E. (2003). Survival, growth, and growth allocation of planted Scots pine trees after different levels of biomass removal in clear-felling. *Forest Ecology and Management*, 177(1-3), pp. 65-74.
- Ekvall, H., Löfgren, S. & Bostedt, G. (2014). Ash recycling - A method to improve forest production or to restore acidified surface waters? *Forest Policy and Economics*, 45, pp. 42-50.
- European Commission *Communication from the Commission to the European Council and the European Parliament - an energy policy for Europe {SEC(2007) 12} /\* COM/2007/0001 final \*/[2015-08-05].*
- Evans, C.D., Cullen, J.M., Alewell, C., Kopáček, J., Marchetto, A., Moldan, F., Prechtel, A., Rogora, M., Veselý, J. & Wright, R. (2001). Recovery from acidification in European surface waters. *Hydrology and Earth System Sciences*, 5(3), pp. 283-297.
- Falkengren-Grerup, U., Linnermark, N. & Tyler, G. (1987). Changes in Acidity and Cation Pools of South Swedish Soils between 1949 and 1985. *Chemosphere*, 16(10-12), pp. 2239-2248.
- Federer, C.A., Hornbeck, J.W., Tritton, L.M., Martin, C.W., Pierce, R.S. & Smith, C.T. (1989). Long-term depletion of calcium and other nutrients in eastern US forests. *Environmental Management*, 13(5), pp. 593-601.

- Feller, M.C. (2005). Forest harvesting and streamwater inorganic chemistry in western North America: A review. *Journal of the American Water Resources Association*, 41(4), pp. 785-811.
- Ferrier, R.C., Wright, R.F., Cosby, B.J. & Jenkins, A. (1995). Application of the Magic Model to the Norway Spruce Stand at Solling, Germany. *Ecological Modelling*, 83(1-2), pp. 77-84.
- Futter, M., Valinia, S., Löfgren, S., Köhler, S. & Fölster, J. (2014). Long-term trends in water chemistry of acid-sensitive Swedish lakes show slow recovery from historic acidification. *Ambio*, 43(1), pp. 77-90.
- Fölster, J., Andrén, C., Bishop, K., Buffam, I., Cory, N., Goedkoop, W., Holmgren, K., Johnson, R., Laudon, H. & Wilander, A. (2007). A Novel Environmental Quality Criterion for Acidification in Swedish Lakes – An Application of Studies on the Relationship Between Biota and Water Chemistry. *Water, Air, & Soil Pollution: Focus*, 7(1-3), pp. 331-338.
- Garmo, Ø., Skjelkvåle, B., Wit, H., Colombo, L., Curtis, C., Fölster, J., Hoffmann, A., Hruška, J., Høgåsen, T., Jeffries, D., Keller, W.B., Krám, P., Majer, V., Monteith, D., Paterson, A., Rogora, M., Rzychon, D., Steingruber, S., Stoddard, J., Vuorenmaa, J. & Worsztynowicz, A. (2014). Trends in Surface Water Chemistry in Acidified Areas in Europe and North America from 1990 to 2008. *Water, Air, & Soil Pollution*, 225(3), pp. 1-14.
- Grigal, D.F. & Ohmann, P.R. (2005). Calcium and forest systems: Diffusion from deep sources. *Soil Science*, 170(2), pp. 129-136.
- Hallbäck, L. (1992). Long-Term Changes of Base Cation Pools in Soil and Biomass in a Beech and a Spruce Forest of Southern Sweden. *Zeitschrift Fur Pflanzenernahrung Und Bodenkunde*, 155(1), pp. 51-60.
- Hepler, P.K. & Wayne, R.O. (1985). Calcium and Plant Development. *Annual Review of Plant Physiology and Plant Molecular Biology*, 36, pp. 397-439.
- Hornbeck, J.W., Smith, C.T., Martin, Q.W., Tritton, L.M. & Pierce, R.S. (1990). Effects of Intensive Harvesting on Nutrient Capitals of 3 Forest Types in New-England. *Forest Ecology and Management*, 30(1-4), pp. 55-64.
- Huntington, T.G., Hooper, R.P., Johnson, C.E., Aulenbach, B.T., Cappellato, R. & Blum, A.E. (2000). Calcium depletion in a southeastern United States forest ecosystem. *Soil Science Society of America Journal*, 64(5), pp. 1845-1858.
- Hyvönen, R., Olsson, B.A. & Ågren, G.I. (2012). Dynamics of soil C, N and Ca in four Swedish forests after removal of tops, branches and stumps as predicted by the Q model. *Scandinavian Journal of Forest Research*, 27(8), pp. 774-786.
- ICP Integrated Monitoring (2015-08-03). *Methodology and reporting of subprogrammes*. [http://www.syke.fi/en-US/Research\\_Development/Maintaining\\_ecosystem\\_services\\_and\\_biodiversity/Monitoring/Integrated\\_Monitoring/Manual\\_for\\_Integrated\\_Monitoring/7\\_Methodology\\_and\\_Reporting\\_of\\_Subprogrammes](http://www.syke.fi/en-US/Research_Development/Maintaining_ecosystem_services_and_biodiversity/Monitoring/Integrated_Monitoring/Manual_for_Integrated_Monitoring/7_Methodology_and_Reporting_of_Subprogrammes).
- Iwald, J., Löfgren, S., Stendahl, J. & Karlton, 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.
- Jandl, R., Alewell, C. & Prietzel, J. (2004). Calcium loss in Central European forest soils. *Soil Science Society of America Journal*, 68(2), pp. 588-595.

- Jenkins, A., Cosby, B.J., Ferrier, R.C., Walker, T.A.B. & Miller, J.D. (1990). Modeling Stream Acidification in Afforested Catchments - an Assessment of the Relative Effects of Acid Deposition and Afforestation. *Journal of Hydrology*, 120(1-4), pp. 163-181.
- Johnson, A.H., Moyer, A., Bedison, J.E., Richter, S.L. & Willig, S.A. (2008a). Seven Decades of Calcium Depletion in Organic Horizons of Adirondack Forest Soils. *Soil Science Society of America Journal*, 72(6), pp. 1824-1830.
- Johnson, D.W. & Todd, D.E. (1998). Harvesting effects on long-term changes in nutrient pools of mixed oak forest. *Soil Science Society of America Journal*, 62(6), pp. 1725-1735.
- Johnson, D.W., Todd, D.E., Trettin, C.F. & Mulholland, P.J. (2008b). Decadal Changes in Potassium, Calcium, and Magnesium in a Deciduous Forest Soil. *Soil Science Society of America Journal*, 72(6), pp. 1795-1805.
- Johnson, J., Aherne, J. & Cummins, T. (2015). Base cation budgets under residue removal in temperate maritime plantation forests. *Forest Ecology and Management*, 343, pp. 144-156.
- Joki-Heiskala, P., Johansson, M., Holmberg, M., Mattsson, T., Forsius, M., Kortelainen, P. & Hallin, L. (2003). Long-term base cation balances of forest mineral soils in Finland. *Water Air and Soil Pollution*, 150(1-4), pp. 255-273.
- Jongmans, A.G., van Breemen, N., Lundström, U., van Hees, P.A.W., Finlay, R.D., Srinivasan, M., Unestam, T., Giesler, R., Melkerud, P.A. & Olsson, M. (1997). Rock-eating fungi. *Nature*, 389(6652), pp. 682-683.
- Kreutzweiser, D.P., Hazlett, P.W. & Gunn, J.M. (2008). Logging impacts on the biogeochemistry of boreal forest soils and nutrient export to aquatic systems: A review. *Environmental Reviews*, 16, pp. 157-179.
- Likens, G.E., Driscoll, C.T., Buso, D.C., Siccama, T.G., Johnson, C.E., Lovett, G.M., Fahey, T.J., Reiners, W.A., Ryan, D.F., Martin, C.W. & Bailey, S.W. (1998). The biogeochemistry of calcium at Hubbard Brook. *Biogeochemistry*, 41(2), pp. 89-173.
- Löfgren, S., Aastrup, M., Bringmark, L., Hultberg, H., Lewin-Pihlblad, L., Lundin, L., Karlsson, G. & Thunholm, B. (2011). Recovery of Soil Water, Groundwater, and Streamwater From Acidification at the Swedish Integrated Monitoring Catchments. *Ambio*, 40(8), pp. 836-856.
- Löfgren, S., Cory, N., Zetterberg, T., Larsson, P.E. & Kronnäs, V. (2009). The long-term effects of catchment liming and reduced sulphur deposition on forest soils and runoff chemistry in southwest Sweden. *Forest Ecology and Management*, 258(5), pp. 567-578.
- Löfgren, S. & Zetterberg, T. (2011). Decreased DOC concentrations in soil water in forested areas in southern Sweden during 1987–2008. *Science of the Total Environment*, 409(10), pp. 1916-1926.
- McDonnell, T.C., Sullivan, T.J., Cosby, B.J., Jackson, W.A. & Elliott, K.J. (2013). Effects of Climate, Land Management, and Sulfur Deposition on Soil Base Cation Supply in National Forests of the Southern Appalachian Mountains. *Water, Air, and Soil Pollution*, 224(10), pp. 1-18.
- McLaughlin, J.W. (2014). Forest Soil Calcium Dynamics and Water Quality: Implications for Forest Management Planning. *Soil Science Society of America Journal*, 78(3), pp. 1003-1020.
- McLaughlin, S.B. & Wimmer, R. (1999). Tansley Review No. 104 - Calcium physiology and terrestrial ecosystem processes. *New Phytologist*, 142(3), pp. 373-417.

- Moldan, F., Cosby, B.J. & Wright, R.F. (2013). Modeling Past and Future Acidification of Swedish Lakes. *Ambio*, 42(5), pp. 577-586.
- Mylona, S. (1996). Sulphur dioxide emissions in Europe 1880-1991 and their effect on sulphur concentrations and depositions. *Tellus*, 48B, pp. 662-689.
- Olsson, B.A., Bengtsson, J. & Lundkvist, H. (1996). Effects of different forest harvest intensities on the pools of exchangeable cations in coniferous forest soils. *Forest Ecology and Management*, 84(1-3), pp. 135-147.
- Olsson, B.A., Lundkvist, H. & Staaf, H. (2000). Nutrient status in needles of Norway spruce and Scots pine following harvesting of logging residues. *Plant and Soil*, 223(1-2), pp. 161-173.
- Oulehle, F., Hofmeister, J. & Hruska, J. (2007). Modeling of the long-term effect of tree species (Norway spruce and European beech) on soil acidification in the Ore Mountains. *Ecological Modelling*, 204(3-4), pp. 359-371.
- Rademacher, P., Mueller-Using, B., Meesenburg, H. & Meiwes, K.J. (2001). Inventory and budget of nutrients in managed German forest stands - a method for sustainable nutrient management. In: Horst, W.J., Schenk, M.K., Bürkert, A., Claassen, N., Flessa, H., Frommer, W.B., Goldbach, H., Olf, H.W., Römheld, V., Sattelmacher, B., Schmidhalter, U., Schubert, S., Wirén, N. & Wittenmayer, L. (eds) *Plant nutrition: food security and sustainability of agro-systems through basic and applied research* Boston: Kluwer Academic Publisher, pp. 906-907.
- Reuss, J.O. & Johnson, D.W. (1986). *Acid deposition and the acidification of soils and waters*. New York, USA: Springer-Verlag.
- Richter, D.D., Markewitz, D., Wells, C.G., Allen, H.L., April, R., Heine, P.R. & Urrego, B. (1994). Soil Chemical Change during Three Decades in an Old-Field Loblolly Pine (*Pinus Taeda* L.) Ecosystem. *Ecology*, 75(5), pp. 1463-1473.
- Saarsalmi, A., Tamminen, P., Kukkola, M. & Hautajärvi, R. (2010). Whole-tree harvesting at clear-felling: Impact on soil chemistry, needle nutrient concentrations and growth of Scots pine. *Scandinavian Journal of Forest Research*, 25(2), pp. 148-156.
- SLU (2015). *Swedish Forest Soil Inventory*. <http://www.slu.se/en/collaborative-centres-and-projects/swedish-forest-soil-inventory/> [2015-08-06].
- Swedish Environmental Protection Agency (1999). *Bedömningsgrunder för miljökalitet. Skogslandskapet*. (Rapport 4917). Stockholm: Swedish Environmental Protection Agency.
- Swedish Environmental Protection Agency (2007a). *Bara naturlig försurning. Bilagor till underlagsrapport till fördjupad utvärdering av miljömålen*. (Report 5780). Stockholm: Swedish Environmental Protection Agency.
- Swedish Environmental Protection Agency (2007b). *Bara naturlig försurning. Underlagsrapport till fördjupad utvärdering av miljötillståndet*. (Report, 5766). Stockholm: Swedish Environmental Protection Agency.
- Swedish Forest Agency (2008). *Rekommendationer vid uttag av avverkningsrester och askåterföring*. (Meddelande, 2-2008). Jönköping: Swedish Forest Agency.
- Swedish Forest Agency (2013). *Swedish Statistical Yearbook of Forestry 2013*. Jönköping: Swedish Forest Agency.

- Thiffault, E., Hannam, K.D., Paré, D., Titus, B.D., Hazlett, P.W., Maynard, D.G. & Brais, S. (2011). Effects of forest biomass harvesting on soil productivity in boreal and temperate forests — A review. *Environmental Reviews*, 19, pp. 278-309.
- UNECE (2015). *ICP Integrated Monitoring. International Cooperative Programme on Integrated Monitoring of Air Pollution Effects on Ecosystems*.  
<http://www.unece.org/env/lrtap/workinggroups/wge/im.html> [2015-08-06].
- Vadeboncoeur, M.A., Hamburg, S.P., Yanai, R.D. & Blum, J.D. (2014). Rates of sustainable forest harvest depend on rotation length and weathering of soil minerals. *Forest Ecology and Management*, 318, pp. 194-205.
- Walmsley, J.D., Jones, D.L., Reynolds, B., Price, M.H. & Healey, J.R. (2009). Whole tree harvesting can reduce second rotation forest productivity. *Forest Ecology and Management*, 257(3), pp. 1104-1111.
- van der Heijden, G., Dambrine, E., Pollier, B., Zeller, B., Ranger, J. & Legout, A. (2015). Mg and Ca uptake by roots in relation to depth and allocation to aboveground tissues: results from an isotopic labeling study in a beech forest on base-poor soil. *Biogeochemistry*, 122(2-3), pp. 375-393.
- van der Heijden, G., Legout, A., Nicolas, M., Ulrich, E., Johnson, D.W. & Dambrine, E. (2011). Long-term sustainability of forest ecosystems on sandstone in the Vosges Mountains (France) facing atmospheric deposition and silvicultural change. *Forest Ecology and Management*, 261(3), pp. 730-740.
- van der Heijden, G., Legout, A., Pollier, B., Mareschal, L., Turpault, M.P., Ranger, J. & Dambrine, E. (2013). Assessing Mg and Ca depletion from broadleaf forest soils and potential causes - A case study in the Morvan Mountains. *Forest Ecology and Management*, 293, pp. 65-78.
- van der Heijden, G., Legout, A., Pollier, B., Ranger, J. & Dambrine, E. (2014). The dynamics of calcium and magnesium inputs by throughfall in a forest ecosystem on base poor soil are very slow and conservative: evidence from an isotopic tracing experiment ( $^{26}\text{Mg}$  and  $^{44}\text{Ca}$ ). *Biogeochemistry*, 118(1-3), pp. 413-442.
- Vanguelova, E., Pitman, R., Luro, J. & Helmisaari, H.-S. (2010). Long term effects of whole tree harvesting on soil carbon and nutrient sustainability in the UK. *Biogeochemistry*, 101(1-3), pp. 43-59.
- Watmough, S.A. & Dillon, P.J. (2003). Base cation and nitrogen budgets for seven forested catchments in central Ontario, 1983-1999. *Forest Ecology and Management*, 177(1-3), pp. 155-177.
- Whitfield, C.J., Aherne, J., Cosby, B.J. & Watmough, S.A. (2010). Modelling boreal lake catchment response to anthropogenic acid deposition. *Journal of limnology*, 69, pp. 135-146.
- Wright, R.F., Larssen, T., Camarero, L., Cosby, B.J., Ferrier, R.C., Helliwell, R., Forsius, M., Jenkins, A., Kopáček, J., Majer, V., Moldan, F., Posch, M., Rogora, M. & Schöpp, W. (2005). Recovery of acidified European surface waters. *Environmental Science & Technology*, 39(3), pp. 64a-72a.
- Yanai, R.D., Blum, J.D., Hamburg, S.P., Arthur, M.A., Nezat, C.A. & Siccama, T.G. (2005). New Insights into Calcium Depletion in Northeastern Forests. *Journal of Forestry*, 103(1), pp. 14-20.

## Acknowledgements

The first time I stepped into the Physical Geography building at Umeå University, I knew that one day I would like to become a PhD-student. This was way back in 1993, but it would take me many years before my dream could be realized. The opportunity arose when I started working with Stefan Löfgren. One day when we were out in the field, collecting water samples, I boldly asked him if he was open to the idea of becoming my supervisor. He looked surprised but quickly said yes. At least, that is how I remember it. It took us some time and energy to formalize an agreement since I was (and still am) working at IVL. Also, we needed a great idea for a PhD-project and someone willing to pay for it. After a year of intense work, I was finally able to call myself a proud PhD-student at the department of Aquatic Sciences and Assessment at SLU in Uppsala. This was in March, 2009.

Several years later, at the age of 41, I have succeeded in completing my dissertation. Like many others it was not an easy task. It has been the best of times but also the worst of times. It was the best of times because I was able to realize my dream. It was the worst of times due to several personal tragedies. Needless to say that it has been challenging at times. However, I am surrounded by a number of great people who has helped me overcome bumps in the road, both professionally and personally.

To begin with I would like to thank my supervisor, Stefan Löfgren. When I began this quest I told you that I wanted you to make me a better scientist. I believe you have. Through endless conversations, mostly over the phone, you have guided me through this journey. You have taken on the role as supervisor, mentor, teacher, collaborator and not least, as a friend. I hope that we will continue to work together in the future since I still have a lot to learn from you.

I also want to thank Bengt Olsson, my assistant supervisor. Who knew, back in 1998, when I was citing your exciting work in a literature study that I would end up working with you one day? I am overcome with joy for being

able to work with you and the HELTRAD sites. Let's continue to do so. And as a supervisor, you and Stefan have complemented each other perfectly.

I am also grateful for my other assistant supervisors, official as well as unofficial, for being involved in my studies and supported me whenever needed. Especially in answering endless questions about MAGIC...Therefore, a big thank you to Filip Moldan and Stephan Köhler is in order but also to Anneli Ågren, John Munthe and Lars Lundin. I have also had great help from my co-authors not already mentioned; Riitta Hyvönen, Per-Olov Brandtberg and Claudia von Brömssen.

There are also many friends and colleagues at SLU and IVL who has made this journey possible, I would like to thank all of you for supporting me. The people at SLU who always made me feel welcome when I came to visit but also my colleagues at IVL who showed respect for my studies but always included me in everything.

I would also like to acknowledge the executive board at IVL who let me conduct my studies full time while still being employed by the company. This allowed me to stay in Gothenburg where my friends and family are. Also, this project would never have been realized without the financial support from the Swedish Energy Agency to which I extend my sincerest thanks and appreciation.

Finally, I want to thank my family, extended family and close friends. First, my grandmother, Solveig, who encouraged me as a child to pursue a career in science, but unfortunately never got to see me finish my PhD-studies. My mother, Synnöve, for always making me believe in me and for being my biggest cheerleader. My father, Göran who has always supported me in everything although he can no longer express how proud he is of me. My stepdad, Lloyd, who is always there for me and makes me laugh from endless (bad) jokes. My extended family; Ulla, Ellert, Karin, Mattias, Linda, Jim and the children for bringing joy into my life and forcing me to take a break from work sometimes. And all my friends, old ones as well as new ones, with whom I've got to share good and bad experiences with. Hopefully we will have more time to see each other now?

I have saved the best for last; Mattias, my beloved husband who is always by my side. Who would I be without you? As a couple, we have endured a lot during the years we've been together, especially during the latter years. However, we have overcome everything in our way and today we are stronger than ever. I feel optimistic about the future, maybe great things are about to happen?