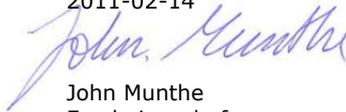


Long-term effects of clear-cutting and biomass removal on soil water chemistry at three coniferous sites in Sweden

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February 2011

The report approved:  
2011-02-14



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Forskningschef



<p><b>Organization</b></p> <p>IVL Swedish Environmental Research Institute Ltd.</p>	<p><b>Report Summary</b></p>
<p><b>Address</b></p> <p>P.O. Box 5302 SE-400 14 Göteborg</p>	<p><b>Project title</b></p> <p>Long-term effects on soil water chemistry following whole-tree harvesting, P30885-1</p>
<p><b>Telephone</b></p> <p>+46 (0)31-725 62 00</p>	<p><b>Project sponsor</b></p> <p>Swedish Energy Agency</p>
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<p><b>Title and subtitle of the report</b></p> <p>Long-term effects of clear-cutting and biomass removal on soil water chemistry at three coniferous sites in Sweden.</p>	
<p><b>Summary</b></p> <p>The long-term effects (27-36 years) of conventional and whole-tree harvest on soil water chemistry (50 cm depth) have been examined at three coniferous sites. One of the most important findings of our study were that WTH caused a long-term (27-30 years), but temporary loss, of exchangeable Ca<sup>2+</sup> measured as lower soil water concentrations at 50 cm depth as a result of large amounts of Ca<sup>2+</sup> being removed during harvest. In addition, WTH resulted in lower soil water pH and ANC. Treatment effects were also noted for SO<sub>4</sub><sup>2-</sup> which could be the combined results of lower dry deposition in WTH-plots and/or increased retention in the mineral soil. The main treatment differences are temporary and had disappeared during the second study period between 2008 and 2010 (i.e. 32 to 35 years after harvest). These results are in agreement with previous soil studies carried out at the same sites. The implications of WTH effect are discussed in the context of validating model forecasts and need for ash-recycling.</p>	
<p><b>Keyword</b></p> <p>Acidification, base cations, bioenergy, buffering capacity, clear-cutting, coniferous, forest fuel, logging residues, harvest, pine, soil solution, soil water, spruce, Sweden, whole-tree harvest.</p>	
<p><b>Bibliographic data</b></p> <p>IVL Report B1959</p>	
<p><b>The report can be ordered via</b></p> <p>Homepage: <a href="http://www.ivl.se">www.ivl.se</a>, e-mail: <a href="mailto:publicationservice@ivl.se">publicationservice@ivl.se</a>, fax+46 (0)8-598 563 90, or via IVL, P.O. Box 21060, SE-100 31 Stockholm Sweden</p>	



## **Preface**

The Swedish Energy Agency works towards achieving an economically and ecologically sustainable energy system by e.g. supporting biofuel research. In 2007, the IVL Swedish Environmental Research Institute was granted funding from the energy research program “Sustainable supply and upgrading of biomass feedstock for energy purposes” to resume a pilot study that ran from 2003 to 2005. The work is carried out in collaboration with the Swedish University of Agricultural Sciences, Department of Ecology.

In this report we present unique results on long-term effects of clear-cutting and biomass removal on soil water chemistry at three coniferous sites in Sweden during 2008-2010. We have also included the results from a pilot study performed at the same sites in order to examine temporal trends. The sites were established 35-36 years ago and are among the oldest experimental sites in the world in this field of research. We are very grateful for the early work carried out by Göran Björkroth and Kaj Rosén which provided us with the opportunity to work on these sites.

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Gothenburg 2011-02-14

## Summary

The effect of harvest intensity (whole-tree vs. stem-only), site (Lövliden, Kosta and Tönnersjöheden) and season (spring, summer and autumn) on soil water chemistry (50 cm depth, ~B/C-horizon) has been determined at three coniferous sites. The sites were harvested 35-36 years ago (1975-76) and are located in southern (Kosta and Tönnersjöheden) and northern (Lövliden) Sweden. This report primarily describes results from 2008 to 2010 but data from a pilot investigation performed between 2003 and 2005 has been included in order to study temporal trends. The sites differ in terms of geographic location, climate, deposition, tree species, soil characteristics etc. which is reflected in the soil water chemical composition. The sites have also been subjected to different silvicultural practices since the clear-cutting in 1975-76. In addition, the southern part of Sweden was severely hit by a storm in 2005 (Gudrun) and 2007 (Per) leading to large inputs of sea-salts. When studying temporal trends and treatment differences, these factors must be taken into consideration.

One of the most important findings of our study were that Whole Tree Harvest (WTH) caused a long-term (27-30 years), but temporary loss, of exchangeable  $\text{Ca}^{2+}$  measured as lower soil water concentrations at 50 cm depth, as a result of large  $\text{Ca}^{2+}$  removal during harvest. In addition, WTH resulted in lower soil water pH and ANC. Treatment effects were also noted for  $\text{SO}_4^{2-}$  which could be the combined result of lower dry deposition in WTH-plots and/or increased retention in the mineral soil.

The main treatment differences in soil water chemistry are temporary and had disappeared during the second study period between 2008 and 2010 (i.e. 32 to 35 years after harvest). We have no soil water data from earlier time periods of the experiment, but the effect of WTH have probably been more pronounced in the past. The results are in agreement with previous soil studies showing a long-term, but temporary effect, on base saturation and exchangeable acidity in the humus layer and upper mineral soil. With time, the treatment differences has decreased or almost disappeared. Remaining effects on soil Ca pools is most pronounced at Lövliden.

Although examining temporal trends was not a primary focus of this study some interesting observations were made. Firstly, all three sites show increasing trends in soil water pH, ANC and decreasing trends in  $\text{SO}_4^{2-}$ . The decreasing trend in  $\text{SO}_4^{2-}$  concentration at Tönnersjöheden and Kosta is likely to be the combined result of decreased deposition and thinnings carried out in conjunction with the soil water sampling. In contrast, the decreasing trend in soil water  $\text{SO}_4^{2-}$  concentration at Lövliden is explained by decreased deposition only. No other effects of the thinnings were seen on soil water chemistry. Secondly, the sea salt events in 2005 and 2007 temporarily offset the recovery process at Tönnersjöheden and Kosta. Thirdly, the concentration of Cl<sup>-</sup> in Lövliden has decreased over time while deposition has remained constant. Reasons for this need to be explored further.

The results from this study can be used for validating model forecast as well as discussing the need for ash-recycling for the long-term sustainability of the forest.

## Sammanfattning

I mitten av 1970-talet anlades ett antal randomiserade blockförsök runt om i Sverige i brukad skogsmark. Syftet med försöken var att studera de ekologiska effekterna av helträdsuttag (dvs. skörd av stammar, grenar, toppar och barr) jämfört med stamvedsuttag vid slutavverkning. Försöken, som drivs i regi av Sveriges Lantbruksuniversitet, Enheten för skoglig fältforskning, har varit föremål för en rad olika studier under årens lopp, framför allt med avseende på skogsproduktion och markkemi. Under perioden 2003-2005 genomförde IVL Svenska Miljöinstitutet (IVL) en pilotstudie på huruvida markvattenkemin (sammelpöror) på 50 cm djup (~B/C-horisonten) skilde sig åt mellan stamved- och helträdsytter. Resultaten pekade på att helträdsuttag ledde till surare markvatten mätt som minskade koncentrationer av baskatjoner.

För att bekräfta dessa observationer beviljades IVL medel av Energimyndigheten för att på ett mer noggrant sätt undersöka om helträdsuttag verkligen leder till ett surare och mindre välbuffrat markvatten. Studien genomfördes under perioden 2008-2010 i samarbete med Sveriges Lantbruksuniversitet (SLU). Försöksområdena är etablerade i södra (Kosta och Tönnersjöheden) och norra (Lövliden) Sverige. Under försöksperioden samlades ett antal markvattenprover in för att belysa skillnaden mellan avverkningsintensitet (stamved- och helträdsuttag), lokal (Lövliden, Kosta och Tönnersjöheden) och säsong (vår, sommar och höst). Analyserna av mikro- och makrokonstituenterna gjordes på diskreta prover. Efter slutavverkningen har bestånden varit föremål för olika skogsskötselåtgärder. Dessutom drabbades södra Sverige av två kraftiga stormar 2005 (Gudrun) och 2007 (Per). Tillsammans medför detta att behandlingsskillnader och trender över tiden kan vara svårare att upptäcka.

Resultaten från denna studie bekräftade emellertid observationerna från den tidigare pilotstudien, nämligen att helträdsuttag leder till en långsiktig (27-30 år), men tillfällig förlust av utbytbar  $\text{Ca}^{2+}$  kopplat till bortförelsen av  $\text{Ca}^{2+}$  i biomassa. Dessutom ledde helträdsuttag till lägre pH,  $\text{SO}_4^{2-}$  och syraneutraliserande förmåga (ANC=Acid Neutralizing Capacity). Lägre halter av  $\text{SO}_4^{2-}$  skulle kunna bero på lägre torrdeposition och/eller högre retention i mineraljorden. Skillnaderna hade emellertid försvunnit mellan 2008-2010 (32-35 år efter avverkning). Tidigare försurningseffekter (<27 år) kan dock inte bortses från. Resultaten är i överensstämmelse med de markkemiska studierna som har visat på en långsiktig men tillfällig effekt på basmättnadsgraden och utbytbara aciditet i humuslagret och den övre mineraljorden. Skillnaden har minskat med tiden och nästan försvunnit 2002-2004. Kvarstående effekter på Ca-förråden är mest uttalade i Lövliden vilket också tillhör det mest välbuffrade området (och således minst behov av askåterföring).

Huvudsyftet med denna studie var i första hand att studera behandlingsskillnader mellan stamveds- och helträdsuttag. Mätningarna från pilotstudien möjliggjorde emellertid att även skillnader över tid kunde observeras (dock inte statistiskt testade). Bland annat uppvisar samtliga tre försöksområden en återhämtning från försurning mätt som en ökad trend i pH och ANC och en minskad trend i  $\text{SO}_4^{2-}$ . Den minskade  $\text{SO}_4^{2-}$  depositionen (som sannolikt har lett till denna återhämtning) är ett resultat av europeiska utsläppsbegränsningar (och genomförda gallringar i södra Sverige). Återhämtningsprocessen i södra Sverige bromsades tillfälligt av ett havssaltsnedfall som stormen Gudrun och Per orsakade. I norra Sverige har Cl- koncentrationen även minskat trots att depositionen har legat på en konstant nivå. Anledningen till detta har inte kunnat utredas inom ramen för detta projekt men indikerar en fastläggning av Cl- i marken.

Resultaten från långliggande studier är värdefulla och kan användas för att validera modellresultat såväl som att diskutera behovet av askåterföring.



# 1 Introduction

Biomass from renewable forest resources is a main provider of energy in Sweden. In 2008, the total supply of biofuels, peat, waste etc. amounted to 123 TWh (or 20 % of the total energy supply) used primarily in district heating plants, electricity production, forest product industry and for heating of residential buildings (Swedish Energy Agency, 2009). According to binding energy policies, the use of renewable energies is expected to increase even further in the future while reducing the use of fossil fuels (European Commission, 2007; directive 2009/28/EC). Increased utilization of logging residues (above and below ground) will contribute to the goal of reducing our fossil fuel dependency. At the same time, valuable nutrients and buffer capacity is removed from the forest which could prevent or delay a recovery from acidification in forest soils and surface waters (Swedish Environmental Protection Agency, 2007).

There are a number of natural and anthropogenic processes which governs soil and surface water acidification measured as a decrease in pH and loss of base cations. For example, trees naturally acidify the soil via cation uptake, predominantly base cations. Hallbäck & Tamm (1986) found an age-related decline in soil pH by almost one unit in the A-horizon of a spruce stand during a period of 55 years. Processes that consume hydrogen ions ( $H^+$ ), and thus neutralize the acidity, include decomposition and weathering. Decomposition and nutrient release from various logging residues (stems, roots, branches, needles and leaves) after cutting are well studied (e.g. Arthur et al., 1993; Fahey et al., 1991; Fahey and Arthur, 1994; Fahey et al., 1988; Hyvönen et al., 2000; Palviainen et al., 2004). The results show that nutrient rich material (leaves and needles) is a primary source of nutrients followed by more woody material (branches and roots) (e.g. Hyvönen et al., 2000). Additionally, weathering introduces new base cations into the system. In general, soil pH increases with depth as a result of weathering processes (Ilvesniemi et al., 2000). If no trees are harvested, nutrient uptake and decomposition will compensate each other in the long-term. However, following clear-cutting and harvesting, the ability to neutralize  $H^+$  via decomposition decreases, principally by the removal of base cations in harvested biomass. The amount of nutrients removed in above-ground biomass via whole-tree harvest (WTH) is 2–4 times higher than in conventional stem-only harvest (CH). Consequently, introducing WTH in forestry inevitably leads to a greater loss of buffer capacity in the soil. A change in soil acidity causes changes in the chemical composition of soil water but the magnitude and nature of the changes is difficult to predict since the solution composition depends not only on ion exchange processes, but also on deposition of sea salt and acidifying pollutants, redox reactions, leaching, nutrient uptake, etc. Nevertheless, if the concentrations of exchangeable soil base cations decrease, the soil solution quickly equilibrates resulting in lower soil water base cation concentrations, acid neutralizing capacity (ANC) and pH, and vice versa.

In the absence of long-term field studies, different model approaches have been used to predict the long-term sustainability following one or two rotation periods. The word sustainability has different meanings but generally describes a system where economic, social and environmental goals are met. In managed forest systems this is achieved when site productivity is maintained without negative environmental effects such as acidification, eutrophication and loss of biodiversity. According to the most recent mass-balance calculations, weathering and deposition alone cannot compensate for the net losses in exchangeable calcium ( $Ca^{2+}$ ) and magnesium ( $Mg^{2+}$ ) pools down to 50 cm depth for large parts of Sweden, regardless of harvesting method (Akselsson et al., 2007). Losses of potassium ( $K^+$ ) following CH in pine stands are compensated for by weathering and deposition but not in spruce stands following WTH. Similar results have been found in Finland and Germany using the same input-output approach for  $Ca^{2+}$ ,  $Mg^{2+}$  and  $K^+$  (Joki-Heiskala et al., 2003; Rademacher et al., 2001). In addition to mass-balance calculations, dynamic models are applied to

include temporally variable parameters. Using the dynamic model Model of Acidification of Ground water In Catchments (MAGIC), Aherne et al. (2008) showed that WTH will lead to a long-term re-acidification of soils and lakes (with ANC<0) despite further reductions in emissions of acidifying compounds. Similarly, Watmough et al. (2003) found that any harvesting “*in south-central Ontario will potentially have a dramatic impact on lake chemistry*”, especially in the WTH scenario according to the Steady-State Water Chemistry (SSWC) model. Consequently, in order to maintain sustainability, ash-recycling and N-fertilization may become necessary according to these forecasts.

Despite major research, there is limited empirical evidence that forest fuel extraction causes long-term surface water acidification. In addition, little is known about changes in weathering rates which may compensate for the loss of buffering base cations. One reason for this is the lack of long-term experiments and monitoring. Pioneering bioenergy research in Sweden began during the 1960's aiming at studying the effects on tree growth in CH- and WTH-plots (Andersson, 1983). However, information about the experiment is not well documented (e.g. amount of biomass removed, nutrient content in harvest biomass, soil conditions, soil vegetation etc) making these sites less suitable for long-term ecological studies. It was not until the oil crisis in 1973 that proper biofuel research programs began, designed in a more scientific manner (Anonymous, 1977). Consequently, the oldest field-experiments in Sweden aiming at studying the ecological effects of WTH originate from this period. At this time, four WTH experiments were established in northern (Lund and Lövliden) and southern (Tönnersjöheden and Kosta) part of Sweden which provide an excellent opportunity to study the long-term (27-36 years) effects of harvesting logging residues in final felling. The original purpose of these sites was to study the effects on tree productivity (Anonymous, 1976), but over the last two decades a large number of studies have also been performed on biomass and nutrient content (Björkroth and Rosén, 1977), plant survival and growth (Björkroth, 1977; Egnell, 2011; Egnell and Leijon, 1999; Egnell and Valinger, 2003), vegetation changes (Olsson, 1995; Olsson and Staaf, 1995), soil acidity (Nykvist and Rosén, 1985), soil carbon and nitrogen (Olsson et al., 1996b), exchangeable base cations (Olsson et al., 1996a; Olsson et al., 2004), decomposition and nutrient release (Hyvönen et al., 2000), needle chemistry (Olsson et al., 2000) and soil water chemistry (Zetterberg et al., 2008).

According to the soil chemical studies, small temporary differences in pH were measured following WTH but had disappeared 26-28 years after harvest (Olsson et al., 2004). However, statistically significant differences in base cation pools, base saturation and exchangeable acidity were still measurable in the upper part of the soil (humus, 0-20 cm mineral soil) indicating lower buffer capacity due to WTH. An increase in soil acidity and a decrease in buffer capacity should be reflected in the soil solution. Preliminary results from a pilot soil water study (at 50 cm depth, i.e. below the rooting zone) carried out at the same sites indicated higher acidity and lower base cation concentrations (Zetterberg et al., 2008) which is in agreement with the soil data. In order to verify these observations, a more detailed study was performed from 2008 to 2010. The null hypothesis ( $H_0$ ) is that WTH does not lead to higher acidity and loss of buffer capacity in soil water in comparison to CH. The alternative hypothesis ( $H_1$ ) states that WTH leads to higher acidity and loss of buffer capacity in the soil in comparison to CH.

## 2 Material and methods

### 2.1 Study area

Only three out of the original four sites have been included in this experiment. The study sites are located in southern and northern Sweden (Figure 1) at elevations varying from 100 to 260 meters above sea level (Table 1). Mean annual precipitation is 1040, 595 and 564 mm per year for Tönnersjöheden, Kosta and Lövliden, respectively. The emissions of acidifying pollutants have been reduced and as a result the deposition of S and N has decreased (Appendix 1). Present day deposition rates of total S vary between 0-2 kg per hectare in the northern parts of Sweden and 3-6 kg per hectare in the southern part (Figure 2). However, some areas on the southwest coast still receive more than 6 kg per hectare and year. The total N deposition has also decreased but not to the same extent. Consequently, Lövliden has not been subjected to acid deposition to the same extent as Tönnersjöheden and Kosta. Areas with the highest deposition of S and N also coincide with areas receiving high deposition of sea salts (NaCl) due to the close proximity to the sea. Thus, the highest precipitation amounts occur over the south-west Sweden which coincides with the area most affected by acid deposition and sea salt events.

Soils are classified as iron podzols developed on glacial till of gneissic and or granitic type (Table 1). The soils are freely drained and do not experience periods of saturation (i.e. reduced conditions). The southern sites are more acid than the northern site in terms of pH, exchangeable base cations, base saturation and exchangeable acidity (Appendix 2). According to two out of three criteria put forward by the Swedish Environmental Protection Agency (1999), the soils in Tönnersjöheden and Kosta are moderately acid with respect to pH (4.4-5.5) but highly acidic in terms of base saturation (<6%). Lövliden, on the other hand, has a very low soil acidity (pH >5.5 and very high base saturation >50%) at 15-20 cm soil depth (B-horizon). Consequently, the soils at Lövliden have a greater ability to resist changes in pH due to high buffer capacity. More information about soil chemical characteristics is given in Staaf and Olsson (1991) and Olsson et al. (1996ab, 2004).

The existing stand prior to harvest consisted of a mesic dwarf-shrub forest with Norway spruce (*Picea abies*) or Scots pine (*Pinus sylvestris*) as dominant species (Table 1). The sites were manually harvested in 1975 (Tönnersjöheden and Kosta) and 1976 (Lövliden). All merchantable stems (>5cm top diameter) were removed from the CH-plots. Remaining biomass was evenly spread on the surface. In the WTH-plots, all above-ground biomass (100 %) was harvested. Similar amounts of slash were removed at Tönnersjöheden and Kosta (Table 1 and Table 2). However, stem density was much higher at Kosta in comparison to Tönnersjöheden. This would have resulted in lower mean stem diameter and, consequently, greater amounts of biomass per tree. The lowest biomass volumes were removed from Lövliden. Estimates of nutrient removal show that larger amounts of nutrients are lost via WTH in comparison to CH (Table 2). On average, N and P losses following WTH exceeded those in CH by a factor of 2, while for Ca, Mg and K, losses are between 1.1 to 1.6 times higher. Although larger amounts of biomass and nutrients were removed during harvest at the southern sites, the greatest losses of Ca in absolute numbers occurred at Lövliden (217 kg per ha<sup>-1</sup>).

After patch scarification, two-year old spruce or pine seedlings were planted (3600 seedlings ha<sup>-1</sup>). To avoid shadowing by fast-growing broadleaves, pre-commercial thinnings took place at Kosta in 1983 and 1989, respectively (Table 1). First stage thinning occurred in 2000 and 2004 in Kosta and Tönnersjöheden, respectively. After thinning, biomass was either harvested (Kosta) or left on the forest floor (Tönnersjöheden), regardless of the initial treatment (CH or WTH). In contrast, the

forest stands in Lövliden has never been subjected to thinning operations except for a pre-commercial thinning of broadleaves in 1998.

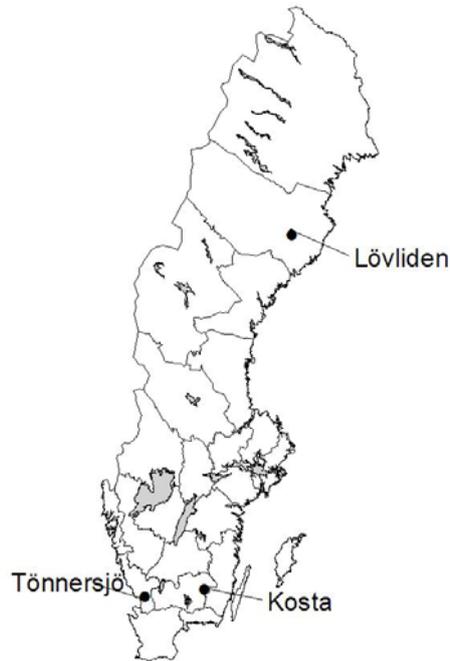


Figure 1. Location of the research sites. Lövliden is situated in the Vindelälven river basin while Tönnersjöheden and Kosta are located in the south-western and southern part of Sweden.

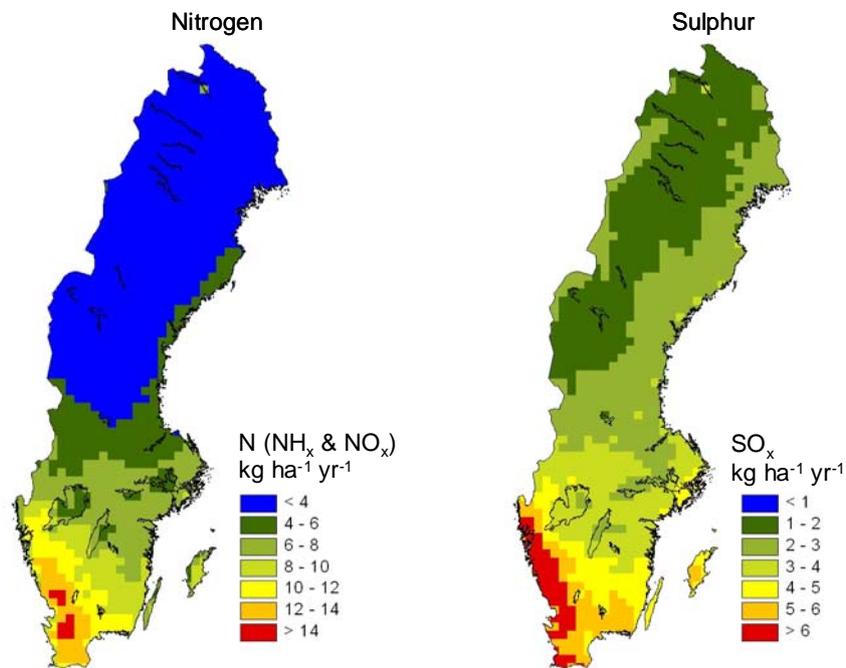


Figure 2. Total deposition of N and S in forests expressed as a mean during the period 2003-2005. The maps are based on MATCH ("Swedish model") with a resolution of 20\*20 km. From IVL Swedish Environmental Institute.

Table 1. Site characteristics and silvicultural history (from Björkroth and Rosén, 1977 and documentation by G. Egnell, SLU).

	Tönnersjöheden	Kosta	Lövliden
<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
Mean annual precipitation (mm yr <sup>-1</sup> )	1040	595	565
Length of growing season (days >5°)	204	195	260
Forest type	Mesic dwarf-shrub	Mesic dwarf-shrub	Mesic dwarf-shrub
Soil moisture class <sup>a</sup>	Fresh	Fresh	Fresh/moist
Soil type	Iron podsol	Iron podsol	Iron podsol
Site productivity (m <sup>3</sup> ha <sup>-1</sup> yr <sup>-1</sup> )	10.1	5.9	3.8
Site index (H <sub>100</sub> )	G30	T24	G20
<b>Harvested stand</b>			
Tree species Dominant Co-dominant	<i>Picea abies</i> (100%) -	<i>Pinus sylvestris</i> (70%) <i>Picea abies</i> (30%)	<i>Picea abies</i> (50%) <i>Pinus sylvestris</i> (50%)
Stand age (yr)	70	100	155 (spruce) 175 (pine)
Stem volume (m <sup>3</sup> ha <sup>-1</sup> ) CH-plots (average) WTH-plots (average)	327 342	300 301	255 237
Number of trees per hectare CH-plots (average) WTH-plots (average)	372 392	796 848	740 1208
Harvest (yr)	Spring 1975	Fall 1975	Fall 1976
Amounts of slash produced at felling (Mg ha <sup>-1</sup> dry wt) CH-plots (average) WTH-plots (average)	39.7 41.8	39.4 42.9	32.8 33.3
<b>New stand</b>			
Tree species	<i>Picea abies</i>	<i>Pinus sylvestris</i>	<i>Picea abies</i>
Planted (yr)	Spring 1976	Spring 1976	Spring 1977
Soil scarification	Patch scarification	Patch scarification	Patch scarification
Number of seedlings (ha <sup>-1</sup> )	3600	3600	3600
Number of seedlings per plot (25*25)	196	196	196
Age of seedlings (yr)	2	2	2
Stand age (yr) <sup>c</sup>	36	36	35
Canopy cover (%) 1983 1991	36 69	20 57	5 24
Pre-commercial thinning of broadleaves	no data	1983, 1989	1998
Thinning (date) Stem density, before ( stems ha <sup>-1</sup> ) <sup>d</sup> Stem density, after (( stems ha <sup>-1</sup> ) <sup>d</sup>	March-April 2004 1565-1950 385-839	Autumn 2000 2018-2653 1293-1519	- - -

<sup>a</sup>According to the Swedish Forest Soil Inventory 2010

<sup>d</sup>Min and max for the individual plots (n=8).

Table 2. Biomass and nutrient removal by conventional and whole-tree harvest (from Björkroth and Rosén, 1977). Average values(n=4 blocks).

Site	Treatment	Biomass (tonnes ha <sup>-1</sup> )	Nutrients removed (kg ha <sup>-1</sup> )				
			N	P	K	Ca	Mg
Tönnersjöheden	CH	150	126	12	56	103	23
	WTH	199	414	36	126	241	53
	<b>ΔWTH-CH</b>	<b>49</b>	<b>288</b>	<b>24</b>	<b>70</b>	<b>138</b>	<b>30</b>
Kosta	CH	155	106	10	50	141	25
	WTH	197	352	35	158	278	49
	<b>ΔWTH-CH</b>	<b>42</b>	<b>246</b>	<b>25</b>	<b>108</b>	<b>137</b>	<b>24</b>
Lövliden	CH	135	114	13	56	207	20
	WTH	161	262	31	133	424	38
	<b>ΔWTH-CH</b>	<b>26</b>	<b>148</b>	<b>18</b>	<b>77</b>	<b>217</b>	<b>18</b>

## 2.2 Experimental design

The experiments are arranged in the form of randomized block with three treatments repeated in four blocks, i.e. 12 plots per site (Appendix 3). The treatments are (i) conventional harvest of stems-only (CH), (ii) whole-tree harvest of all above-ground biomass (WTH) and (iii) whole-tree harvest of all above-ground biomass except for needles (BSH). The BSH treatment resembles the recommendations issued by the Swedish Forest Agency (2008) but in this study, only the CH and WTH treatments are included.

Each plot is 25m \* 25m surrounded by a 2.5 m border strip in order to minimize disturbances from the neighbouring plots and treatments (Figure 3). Soil and soil water sampling is collected within a 20m\*20m net plot inside the larger plot. The distance between each net plot is thus 5 m. Within each net plot, five lysimeters (A-E) are installed at a soil depth of circa 50 cm (from the surface) in the mineral soil corresponding to the B/C-horizon. At this depth, most of the nutrient and water uptake has already taken place. Consequently, the chemical composition of the water leaving the rooting zone (0-50 cm) can be used to indicate e.g. nutrient losses to groundwater and possibly surface waters. The depth was determined based upon rooting zone and soil type. The spatial distribution of the lysimeters was selected to represent soil variability by placing them inside the net parcel in the shape of a cross. Occasionally, small adjustments were made from the original shape due to soil compacting and rockiness.

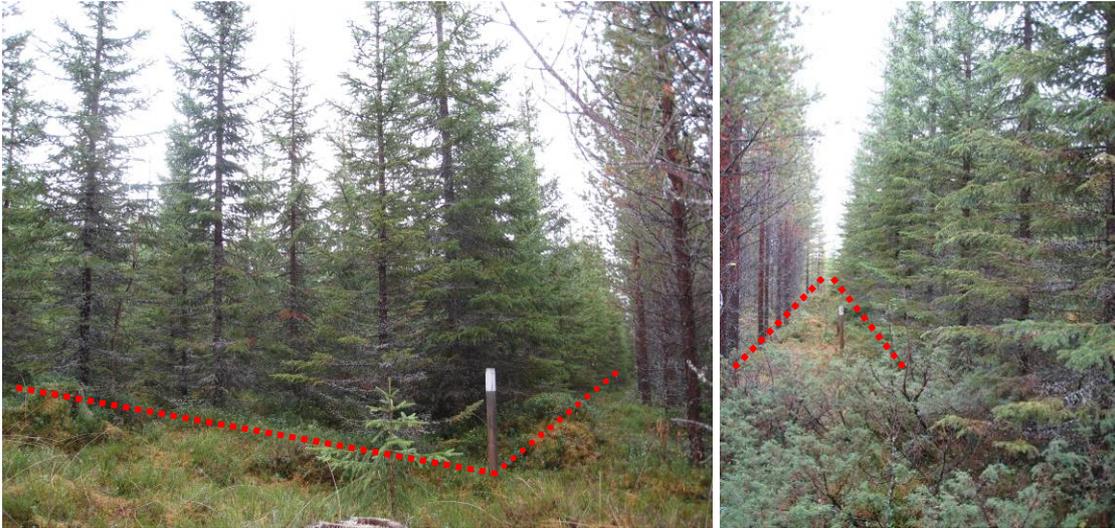


Figure 3. Example of a 25\*25 m plot in Lövliden, Northern Sweden (left). At this particular site, the sampling plots are situated in rows (4 plots per row, see Appendix 3). Each row is separated by a corridor (not to be confused with the border strip) indicated by red dots.

## 2.3 Sampling and analysis

The soil solution was collected using ceramic suction cups (P80) manufactured at the Department of Aquatic Sciences and Assessment (SLU). The permanently installed lysimeters consist of a two part system connected to each other via a PVC tube, assembled by IVL. The suction cups are placed in distilled water for periods between 1 and 4 weeks and completely saturated at the time of installation. Vacuum is applied by means of a hand-operated vacuum pump (NALG 6131-0020, 480 mbar) which forces pore water to penetrate into the ceramic cup and move towards the surface inside the silicon tube before emptying into a polyethylene bottle (250 ml). The plastic bottle is located at the top of the soil surface, stored at ambient soil temperature and protected from light by a ca. 25 cm tall drainage tube (Ø11 cm) (Figure 4). The soil water in the collection bottle was removed after circa two days for transport to the laboratory for chemical analysis. Upon arrival at the laboratory, the soil water volume was recorded for each individual sample but not used in this report.



Figure 4. Soil pore water is collected in a plastic bottle and protected from light and rodents by drainage tubes.

Lysimeter A and B were installed in 2002 and 2003, prior to lysimeters C, D and E. Installation of the remaining lysimeters took place in 2008. After installation the lysimeters were left in the ground on average 179 (2003-2005) and 142 (2008-2010) days for calibration before collecting the first samples. During this time, the lysimeters were rinsed at least once (maximum three times) in the field. The procedure for rinsing is identical to the actual sampling, but instead of saving the water for analysis it is emptied onto the ground. The soil solution was sampled in two periods; between 2003-2005 (from lysimeter A and B) and 2008-2010 (from lysimeter A-E). In order to reduce the cost of analyses, soil water from lysimeter A and B were pooled into a composite sample from 2003 to 2005. But from 2008 to 2010, analysis was made on discrete samples from lysimeter A-E. Consequently, comparisons between the time periods should be done with caution.

Sampling frequency has varied through time but normally took place during spring, summer and autumn (Appendix 5). Sampling during the winter period is normally not possible due to low temperatures and soil frost. The total number of samples on each sampling occasion is  $n=24$  (2003-2005, pooled samples) and  $n=120$  (2008-2010, discrete samples). However, due to weather conditions and disturbances by e.g. vole and wild boars, the actual number of samples was lower. Furthermore, the collected soil water volume was not always adequate enough to permit a complete analysis of all the parameters. Therefore, it was necessary to prioritize the order of analysis with respect to parameter and volume needed to perform the analysis, see below.

All samples were analysed for pH and subsamples were filtered through 0.8  $\mu\text{m}$  acetate cellulose filter (Advantec) at IVL as an extra measure to remove any excess particles. Soil water pH was measured using a pH-electrode (Radiometer). The major anions; sulfate ( $\text{SO}_4^{2-}$ ) nitrate ( $\text{NO}_3^-$ ) and chloride ( $\text{Cl}^-$ ) were analysed by ion chromatography (Dionex ICS-1100) and major cations; calcium ( $\text{Ca}^{2+}$ ), magnesium ( $\text{Mg}^{2+}$ ), potassium ( $\text{K}^+$ ), sodium ( $\text{Na}^+$ ) and manganese ( $\text{Mn}^{2+}$ ) by ion chromatography (Dionex ICS-2000). Ammonium ( $\text{NH}_4^+$ ) was determined via flow injector analysis (FIA) and alkalinity by titration to  $\text{pH}=5.4$ . Subsamples of total aluminium (Alt), organic monomeric aluminium (Alo), iron ( $\text{Fe}^{n+}$ ) and total organic carbon (TOC) were analysed by the Department of Aquatic Sciences and Assessment (SLU). Alt and Alo (after cation-exchange fractioning by Driscoll, 1984) were analysed by ICP-AES. Filtered TOC-samples (from now on

termed DOC) was determined using a Shimadzu TOC 5050 analyzer. The order of chemical analysis is given in Table 3 and information about the analysis is given in Appendix 4.

Table 3. The order of chemical analysis when the total volume needed for analysis was less than 175 ml.

Parameter	Volume needed for analysis (ml)	Priority
pH	10	1
SO <sub>4</sub> <sup>2-</sup> , NO <sub>3</sub> <sup>-</sup> , Cl <sup>-</sup> , Ca <sup>2+</sup> , Mg <sup>2+</sup> , K <sup>+</sup> , Na <sup>+</sup> and Mn <sup>2+</sup>	10	2
NH <sub>4</sub> <sup>+</sup>	10	3
Alt and Fe <sup>n+</sup>	25	4
Aluminium fractioning and Alo	65	5
TOC (DOC)	25	6
Alkalinity	30	7
<b>Total volume</b>	<b>175 ml</b>	<b>-</b>

Acid neutralizing capacity (ANC) was calculated according to Reuss and Johnson (1986). Inorganic cationic aluminium (Ali) was calculated by subtracting organic aluminium (Alo) from total aluminium (Alt). Negative Ali values (Alt<Alo) or values equal to zero (Alt=Alo) were set to 0.0001. Measured values that fall below the detection limit were assigned a value of the detection limit divided by two and used in the statistical analysis. Unfortunately the detection limit has not remained constant between and during the two study periods (2003-2005 and 2008-2010), which presents a problem. In addition, from 2009, detection limit is no longer used according to the Swedish Board for Accreditation and Conformity Assessment (SWEDAC). Instead a quantification level is used which corresponds to the lower limit of the measuring range. For statistical analysis and calculations, only one detection (or quantification) limit were chosen upon for each individual parameter corresponding to the highest level.

## 2.4 Statistical analysis

Differences in soil water concentrations at 50 cm depth were tested separately for the two time periods (2003-2005 and 2008-2010) using SAS version 9.2 for mixed model analysis with both fixed and random effects factors. The fixed factors were site (Tönnersjöheden, Kosta and Lövliden), treatment (WTH and CH) and season (spring, summer and autumn) and the random effect factors; block (1-4) and lysimeter (A-D). Unfortunately, irregularity in soil water sampling during the initial phase led to uneven grouping of data why these had to be omitted from the analysis (Table 4). However, the data has been included in time-series diagrams in order to interpret temporal trends at the individual sites.

The fixed factors are used to describe statistically significant difference ( $p \leq 0.05$ ) in soil water concentrations from an overall mean, either as a result of site, treatment or season. Interactions between these fixed factors were modelled as well. In contrast, random effect factors describe a variance. Thus, statistically significant differences ( $p \leq 0.05$ ) in soil water concentrations as a result of block or treatment makes up some of the overall variability. Between the measurements of each (lysimeter) at different time points, the autocorrelation structure was estimated using the power model  $\sigma^2 \rho^{d_{ij}}$ , where  $\sigma^2$  is the autocorrelation between two subsequent days and  $d_{ij}$  is the distance of two measurements in time (days). The model assumes that the data come from a normally distributed population. For variables not reasonably distributed normally, log-transformations were made. Values consistently falling below the detection limit (e.g. Fe<sup>n+</sup> and Mn<sup>2+</sup>) produce a straight line over time which makes it impossible to perform a statistical analysis. Consequently, only pH,

ANC, DOC, SO<sub>4</sub><sup>2-</sup>, Cl<sup>-</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup>, Na<sup>+</sup>, K<sup>+</sup>, Alt, Alo and Ali are included in the analysis. Kenward-Rogers (1997) were used to adjust the degrees of freedom in unbalanced data sets (e.g. missing values).

Table 4. The statistical analysis was performed on two individual data sets. Sampling dates in brackets were not included in the statistical analysis.

	Tönnersjöheden	Season	Kosta	Season	Lövliden	Season
Time period 1	(2003-06-13)	-	(2003-06-09 <sup>a</sup> )	-	(2003-06-06)	-
	-	-	-	-	2003-06-23	-
	2003-11-18	Autumn	2003-11-14	Autumn	2003-10-20	Autumn
	(2003-12-16)	-	-	-	-	-
	2004-04-15	Spring	2004-04-22	Spring	2004-05-10	Spring
	2004-10-04	Autumn	2004-09-27	Autumn	2004-10-04	Autumn
	2005-05-09	Spring	2005-05-11	Spring	2005-05-23	Spring
	2005-08-17	Summer	2005-08-15	Summer	2005-08-16	Summer
	2005-10-24	Autumn	2005-10-27	Autumn	2005-10-17	Autumn
Time period 2	2008-10-23	Autumn	2008-10-08	Autumn	2008-10-13	Autumn
	2009-05-25	Spring	2009-05-11	Spring	2009-05-25	Spring
	2009-07-20	Summer	2009-07-22	Summer	2009-07-20	Summer
	2009-10-19	Autumn	2009-10-21	Autumn	2009-10-12	Autumn
	2010-04-12	Spring	2010-04-12	Spring	2010-06-07	Spring

<sup>a</sup>Data missing. It has not been possible to find out whether or not soil water samples were analysed and the results lost, or if the samples were lost in the mail.

## 3 Results and discussion

### 3.1 Outcome of the statistical analysis

Main and interaction effects of site, treatment and season on soil water chemistry are summarized in Table 5 (2003-2005) and Table 6 (2008-2010). The main effect of *site* on soil water chemistry was statistically significant for all parameters except Mg<sup>2+</sup> and K<sup>+</sup> during 2003-2005 (Table 5). These site differences were still present during the second study period, with the exception of ANC (Table 6). The results also showed a main *treatment* effect on pH, ANC, SO<sub>4</sub><sup>2-</sup> and Ca<sup>2+</sup> (Table 5). However, during 2008-2010 the treatment effect had disappeared (Table 6). Main *seasonal* effects were present for most variables during the two time periods.

In addition to the main effects, some sites occasionally displayed a different response as indicated by the interactions between site\*treatment and site\*season. In addition, one occasion with a significant treatment\*season interaction was also found (Table 5 and Table 6). For example, there was an overall site effect on pH (p<0.0001) independent of location during 2008-2010, but the difference was not always statistically significant depending on site and season (site\*season, p<0.0001, Table 6). A closer examination of the results show that differences in soil water pH are statistically significant for all sites and seasons except for Kosta and Tönnersjöheden during autumn (p=0.3234, Appendix 6). As a result, the main effect (site) does not collectively explain the influence on pH but also season. And, in order to fully interpret the results, interaction effects must be considered.

There are also examples of interaction effects but no main effects. For example, no treatment effect was found for K<sup>+</sup> during 2003-2005 but the interaction between site\*treatment was statistically significant (p<0.0368, Table 5). Consequently, WTH did not result in an overall difference in soil water K<sup>+</sup> concentrations in comparison to CH, regardless of site. However, lower soil water K<sup>+</sup> concentrations were found in Lövåden following WTH in comparison to CH (p=0.0128, Appendix 6). Further details about statistically significant interaction effects between site and treatment are found in Appendix 6.

Table 5. Results (p-values) from the statistical analysis (mixed model) for the period 2003 to 2005. Variables with values often falling below the detection and/or quantification limit have not been tested statistically. Significant effects are reported at the 0.05 level. NS=Non-Significant.

Variable	Test of fixed effects					
	Main effects			Interaction effects		
	site	treatment	season	site * treatment	site * season	treatment * season
pH	<0.0001	0.0142	NS	NS	NS	NS
Alkalinity	-	-	-	-	-	-
ANC	<0.0001	0.0114	NS	0.0113	NS	NS
SO <sub>4</sub> <sup>2-</sup>	0.0068	0.0583	0.0053	NS	NS	NS
Cl <sup>-</sup>	<0.0001	NS	0.0439	NS	NS	0.0496
NO <sub>3</sub> <sup>-</sup>	-	-	-	-	-	-
NH <sub>4</sub> <sup>+</sup>	-	-	-	-	-	-
Ca <sup>2+</sup>	0.0088	0.0111	NS	NS	0.0061	NS
Mg <sup>2+</sup>	NS	NS	<0.0001	NS	NS	NS
Na <sup>+</sup>	0.0001	NS	0.0018	NS	NS	NS
K <sup>+</sup>	NS	NS	NS	0.0368	0.0015	NS
Mn <sup>2+</sup>	-	-	-	-	-	-
Fe <sup>n+</sup>	-	-	-	-	-	-
DOC	0.0142	NS	0.0002	NS	NS	NS
Alt	<0.0001	NS	0.0012	NS	0.0237	NS
Alo	<0.0001	NS	0.0009	NS	0.0004	NS
Ali	0.0012	NS	0.0147	NS	0.0047	NS

Table 6. Results (p-values) from the statistical analysis (mixed model) for the period 2008 to 2010. Variables with values often falling below the detection and/or quantification limit have not been tested statistically. Significant effects are reported at the 0.05 level. NS=Non-Significant.

Variable	Test of fixed effects					
	Main effects			Interaction effects		
	site	treatment	season	site * treatment	site * season	treatment * season
pH	<0.0001	NS	NS	NS	<0.0001	NS
Alkalinity	-	-	-	-	-	-
ANC	NS	NS	NS	NS	0.0003	NS
SO <sub>4</sub> <sup>2-</sup>	0.0003	NS	0.0408	0.0008	0.0186	NS
Cl <sup>-</sup>	<0.0001	NS	<0.0001	NS	0.0226	NS
NO <sub>3</sub> <sup>-</sup>	-	-	-	-	-	-
NH <sub>4</sub> <sup>+</sup>	-	-	-	-	-	-
Ca <sup>2+</sup>	0.0031	NS	<0.0001	<0.0001	0.0459	NS
Mg <sup>2+</sup>	NS	NS	<0.0001	NS	<0.0001	NS
Na <sup>+</sup>	<0.0001	NS	0.0001	NS	0.0186	NS
K <sup>+</sup>	NS	NS	0.0036	NS	NS	NS
Mn <sup>2+</sup>	-	-	-	-	-	-
Fe <sup>n+</sup>	-	-	-	-	-	-
DOC	<0.0001	NS	<0.0001	NS	NS	NS
Alt	<0.0001	NS	0.0280	NS	<0.0001	NS
Alo	<0.0001	NS	NS	0.0333	0.0174	NS
Ali	<0.0001	NS	NS	NS	<0.001	NS

### 3.2 Temporal trends

Over the years, the sites have been subjected to different silvicultural practices (no thinning or thinning with and without slash removal) since the original clear-cutting in 1975-1976 (Table 1). In addition, the southern part of Sweden was severely hit by a storm in 2005 (Gudrun) and in 2007 (Per) leading to large inputs of  $\text{Na}^+$  and  $\text{Cl}^-$ . Since the thinning operations and sea salt events occurred just prior, during or after the initial soil water sampling (2003-2005) it becomes more complicated to describe overall temporal trends.

In general, the soils are more acidic at the southern sites (Kosta and Tönnersjöheden) than at the northern site (Lövliden), see Appendix 2. As a consequence, measured soil water pH in Lövliden is circa one unit higher (pH~6) with an ANC around 80  $\mu\text{eq}$  per litre (Figure 5 and Figure 6). There appears to have been a positive development with time in terms of pH and ANC, especially at the southern sites. During the initial sampling period (2003-2005), pH varied between 4.6 and 4.9 at Kosta and Tönnersjöheden but in the second period (2008-2010) pH increased to 4.7-5.2 (Figure 5). No such trend is observed at Lövliden. Similarly, ANC tended to increase at all three sites (Figure 6). During 2005, the positive trend in ANC at the southern sites was temporarily offset by a decline in ANC caused by large inputs of  $\text{Cl}^-$  (Figure 8) corresponding to the sea salt event in January 8<sup>th</sup>, 2005.

The general increase in pH and ANC may result from a decrease in soil water  $\text{SO}_4^{2-}$  concentrations (Figure 7) as a result of emission reductions and decreased S deposition. Deposition of  $\text{SO}_4\text{-S}$  in throughfall precipitation collected from nearby sites indicates a decreasing trend during the study period (2003-2010) (Appendix 1). Similar trends are observed in open field  $\text{SO}_4\text{-S}$  deposition at Tönnersjöheden and Kosta but not at Lövliden. At Timrilt, located circa 10 kilometres northeast of Tönnersjöheden, throughfall deposition of non-marine  $\text{SO}_4\text{-S}$  decreased from 12.2 to 5.1 kg per hectare and year between 1996 and 2004. Timrilt was severely damaged by the storm Gudrun why throughfall deposition data from 2005 and onwards should be treated with care. A similar reduction is seen in a pine stand at Attsjö situated circa 17 kilometres northeast of Kosta where throughfall deposition decreased from 5.2 to 2.0 kg  $\text{SO}_4\text{-S}$  per hectare and year between 1997 and 2008. In a spruce stand at Bäcksjö, 60 kilometres southeast of Lövliden,  $\text{SO}_4\text{-S}$  deposition decreased from 4.7 to 2.0 kg per hectare and year between 1992 and 2008 (Appendix 1). A decrease in  $\text{SO}_4^{2-}$  concentration and an increase in pH and ANC at the southern sites may also be attributed to the thinning operations at Tönnersjöheden and Kosta in 2004 and 2000, respectively (Table 1). Thinning of the stands would lower the filtering capacity which in turn would lead to decreased dry deposition and soil water concentrations. Thus, the decreasing trend in  $\text{SO}_4^{2-}$  concentration at Tönnersjöheden and Kosta is likely to be the combined result of decreased deposition and thinnings. In contrast, the decreasing trend in soil water  $\text{SO}_4^{2-}$  concentration at Lövliden is explained by decreased deposition only.

In addition, leaving slash to decompose and mineralize on the forest floor after thinning at Tönnersjöheden could increase soil water base cation concentrations (especially  $\text{K}^+$ ) via leaching from above-lying horizons. An increase in base cations would lead to an increase in both pH and ANC. However, no such increase was observed at Tönnersjöheden (or Kosta). There may be several reasons for this. For example, the thinning at Tönnersjöheden was carried out in the autumn of 2004. It is possible that decomposition and mineralization of fresh litter was delayed and/or that the base cations were effectively retained in the humus layer and upper mineral soil. In addition, the released nutrients may have been quickly leached and effectively utilized by the remaining trees as indicated by  $\text{NO}_3^-$  concentrations near or below the detection limit (Appendix 7). Furthermore, all of the slash was removed from both the CH and WTH plots after the thinning at

Kosta in 2000. Even so, some material is inevitable left on the ground but the amounts were probably too small to contribute to increased concentrations. Thus, the thinnings at Kosta and Tönnersjöheden did not alter soil water chemistry, with the possible exception of  $\text{SO}_4^{2-}$ . In contrast, the concentration of  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$  and  $\text{K}^+$  appear to have decreased during 2003-2005 at Kosta and Tönnersjöheden (Figure 9 to Figure 11). Following the law of electron neutrality principle, a decrease in soil water  $\text{SO}_4^{2-}$  concentration will be met by a by an equal decrease in base cations and vice versa. Thus, a decrease in soil water anion concentration via decreased deposition of  $\text{SO}_4^{2-}$  and  $\text{Cl}^-$  will lead to lower base cation concentrations.

The temporal trend at Lövliden differs to some extent from those at Tönnersjöheden and Kosta. During the first sampling period, pH, ANC and base cations ( $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{K}^+$ ,  $\text{Na}^+$ ) increased but decreased during the second sampling period (Figure 5, Figure 6, and Figure 9 to Figure 12). In contrast,  $\text{SO}_4^{2-}$  and  $\text{Cl}^-$  showed a clear decrease during the entire sampling period (2003-2010) (Figure 7 and Figure 8). Lövliden has not been subjected to any silvicultural practices since 1976, except for a pre-commercial thinning of broadleaves in 1998 (Table 1). As a result, the cause for these trends needs to be sought for elsewhere. For instance, changes in atmospheric deposition and/or differences in tree canopy with time could be responsible for the trends. Looking at deposition data at Lövliden between 2003 and 2010, deposition of  $\text{Cl}^-$  has remained more or less constant while  $\text{SO}_4^{2-}$  has decreased (Appendix 1). As discussed above, the decrease in soil water concentration of  $\text{SO}_4^{2-}$  is likely to be the result of decreased  $\text{SO}_4^{2-}$  deposition. However, the decrease in soil water  $\text{Cl}^-$  concentration cannot be explained by changes in  $\text{Cl}^-$  deposition during the same period.

It has been shown in many studies that the concentration of  $\text{Cl}^-$  and  $\text{SO}_4^{2-}$  in soil water, groundwater and surface water decrease following harvest (e.g. Adamson et al., 1987; Neal et al., 2004; Parfitt et al., 2002). The decrease is generally attributed to a decrease in dry deposition following the removal of trees (Robertson et al., 2000). Treatment effects are generally observed during the clear-cut phase but as the forest canopy closes, soil water concentrations generally returns to pre-harvest conditions. However, McLaughlin & Phillips (2006) measured lower soil water  $\text{Cl}^-$  concentrations (-14.4  $\mu\text{eq}$  per litre) in the mineral soil (25-50 cm) 16-18 years after WTH in comparison to an uncut control forest. No explanation was given but reasons which could help explain these results are differences in crown structure leading to lower dry deposition in the regenerating stand and/or increased  $\text{Cl}^-$  retention in the upper part of the soil. Although  $\text{Cl}^-$  is considered to be a relative inert ion that moves through the soil without participating in exchange processes (Johnson and Cole, 1980) it has been shown that inorganic  $\text{Cl}^-$  can be converted into organic chlorine in the upper part of the soil (Öberg, 1998). Whether or not these processes are responsible for the decreasing  $\text{Cl}^-$  trends in Lövliden can only be speculated upon. However, by visual inspection, the tree crowns at the CH- and WTH-plots do not appear to differ.

The increase and decrease in base cation concentrations during the study period is also intriguing. As the concentration of anions decrease, so would the base cations according to the electron neutrality principle. Instead the trends show the opposite pattern between 2003 and 2005. A satisfying explanation for these trends has not been found and lies beyond the scope of this report.

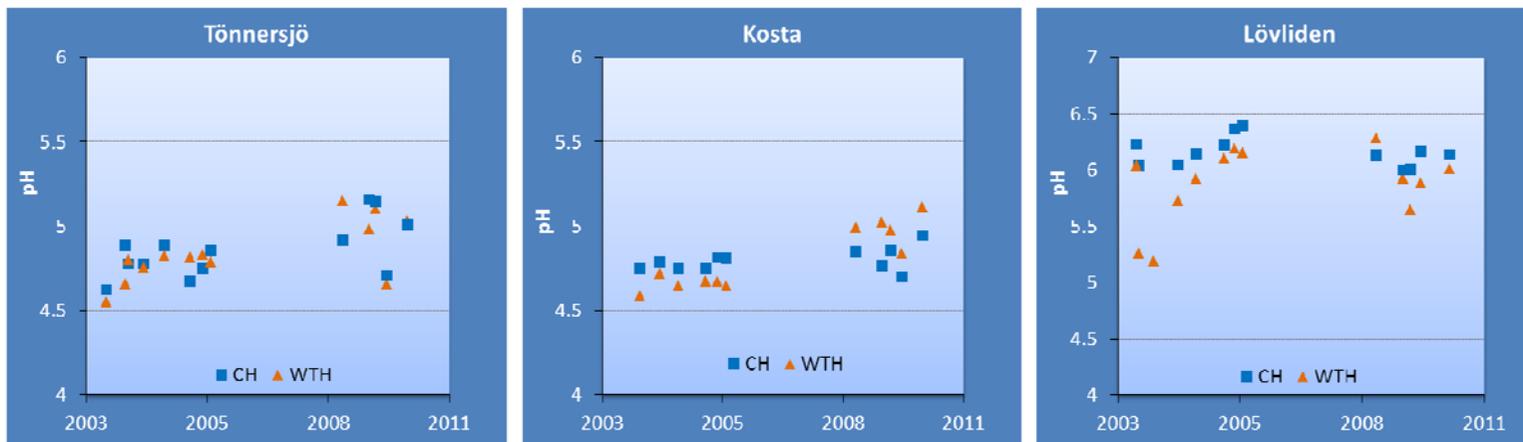


Figure 5. Volume weighed soil water pH at 50 cm depth (~B/C-horizon). Data from 2003 to 2005 are based on composite samples from lysimeter A and B but from 2008 to 2010, analysis was made on discrete samples from lysimeter A-E. Soil water samples were occasionally missing during dry periods. Alternatively, the collected volume was not enough to allow for a complete analysis. As a result, the number of samples from each lysimeter varies why the volume-weighted means for each parameter may be based on  $n < 2$  (2003-2005) and  $n < 5$  (2008-2010). Note differences in scales.

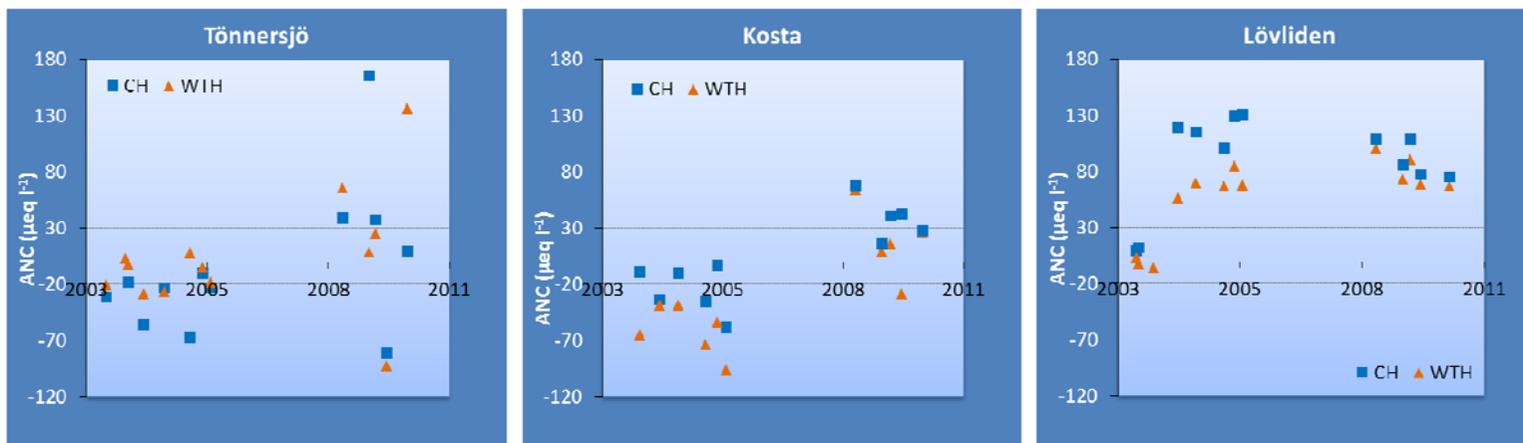


Figure 6. Acid neutralizing capacity (ANC, volume weighed) at 50 cm depth (~B/C-horizon). Legends as in Figure 5.

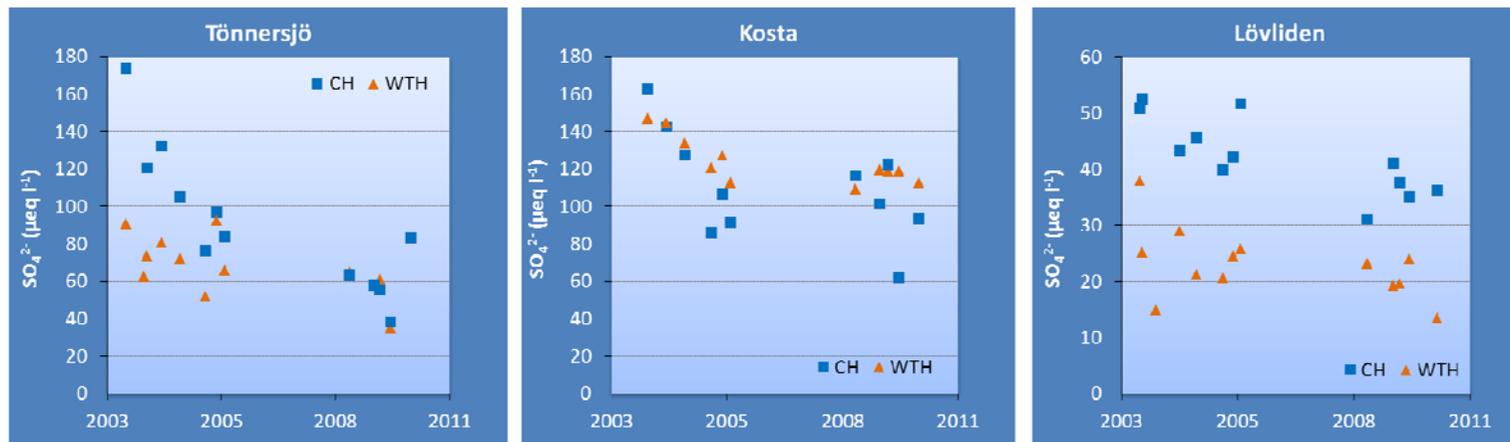


Figure 7. Sulfate ( $\text{SO}_4^{2-}$ ) concentration (volume weighed) at 50 cm depth (~B/C-horizon). Legends as in Figure 5. Note differences in scale.

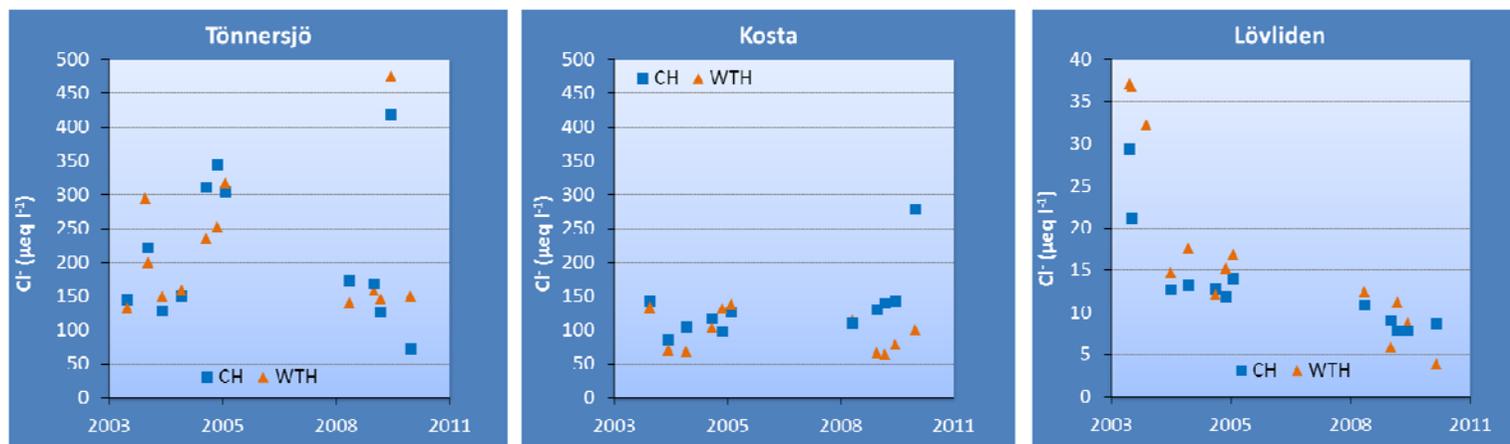


Figure 8. Chloride ( $\text{Cl}^-$ ) concentration (volume weighed) at 50 cm depth (~B/C-horizon). Legends as in Figure 5. Note differences in scale.

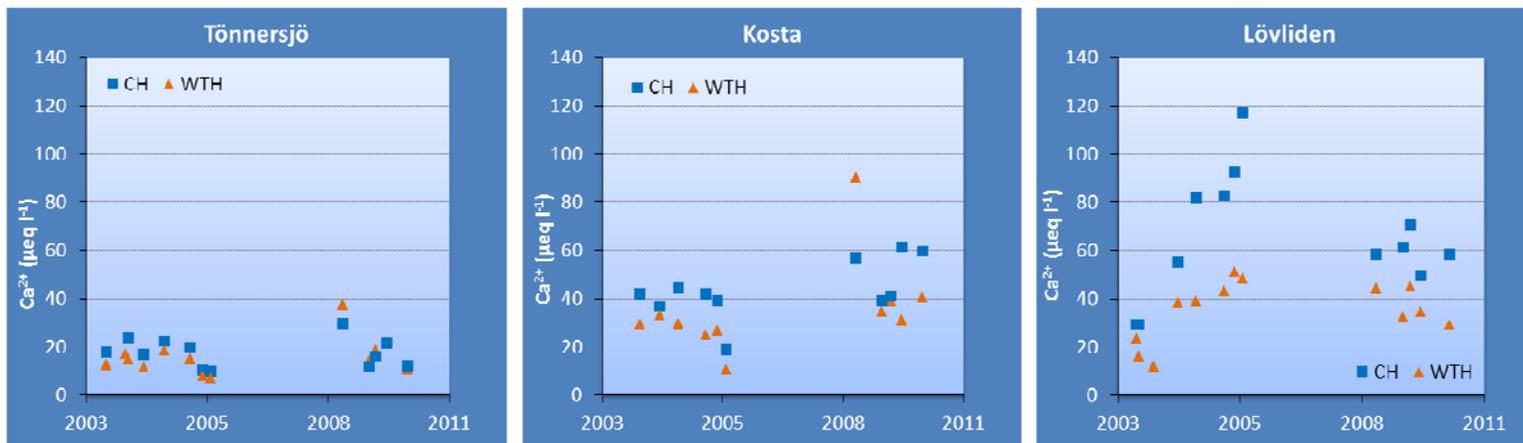


Figure 9. Calcium ( $\text{Ca}^{2+}$ ) concentration (volume weighed) at 50 cm depth (~B/C-horizon). Legends as in Figure 5.

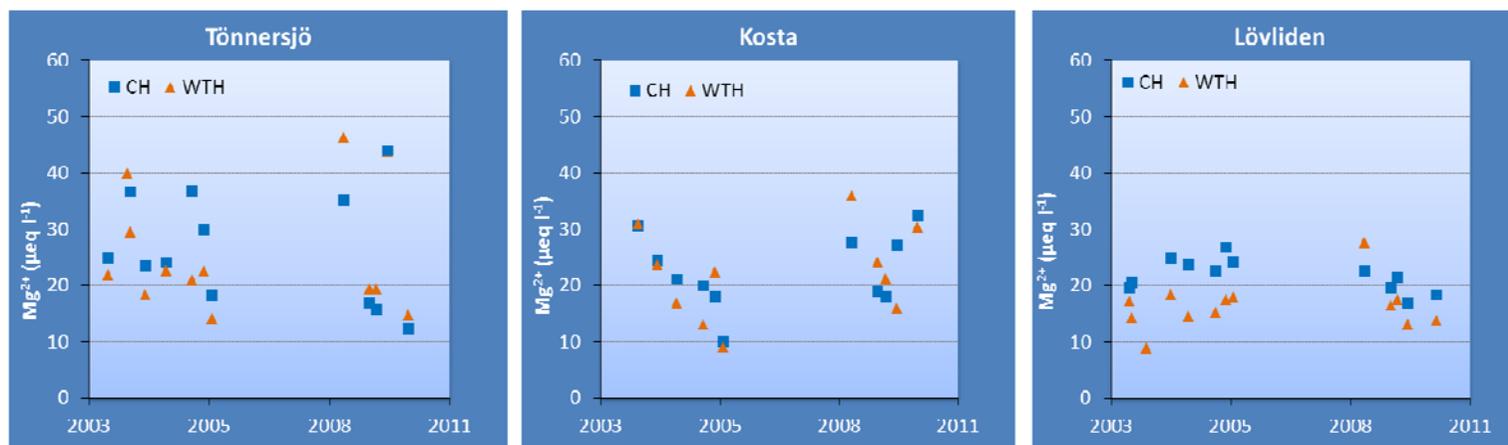


Figure 10. Magnesium ( $\text{Mg}^{2+}$ ) concentration (volume weighed) at 50 cm depth (~B/C-horizon). Legends as in Figure 5.

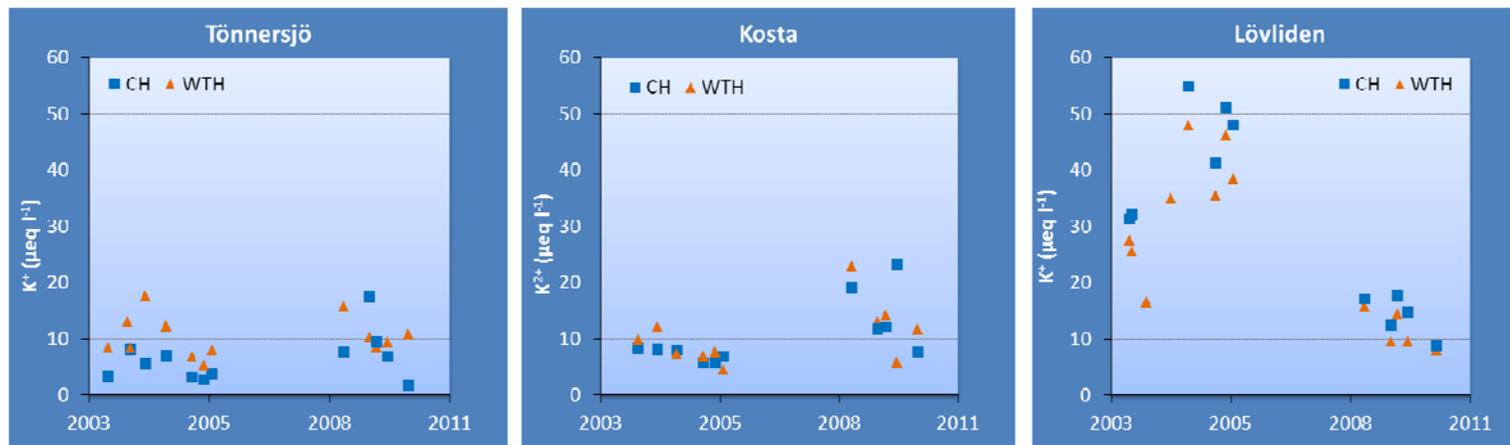


Figure 11. Potassium ( $K^+$ ) concentration (volume weighed) at 50 cm depth (~B/C-horizon). Legends as in Figure 5.

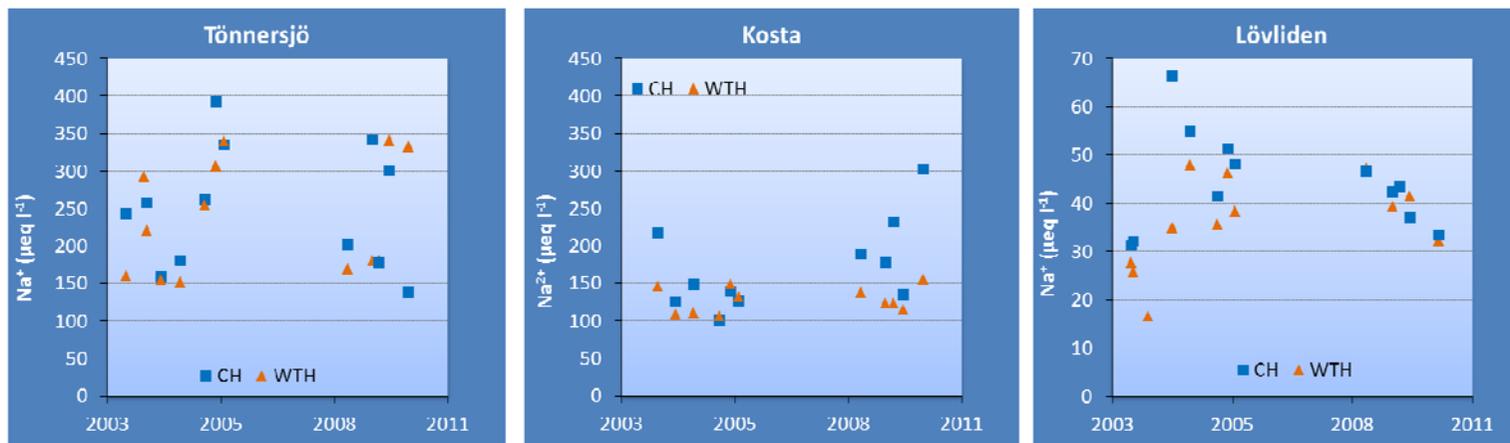


Figure 12. Sodium ( $Na^+$ ) concentration (volume weighed) at 50 cm depth (~B/C-horizon). Legends as in Figure 5.

### **3.3 Treatment effects**

In this study, WTH resulted in lower pH, ANC and lower concentrations of  $\text{Ca}^{2+}$  and  $\text{SO}_4^{2-}$  in comparison to CH between 2003 and 2005, i.e. 27-30 years after harvest (Table 5). The results from the statistical analysis also showed an interaction between site\*treatment for  $\text{SO}_4^{2-}$  and  $\text{Ca}^{2+}$  between 2008 and 2010. In terms of pH, the greatest difference was found at Lövliden and Kosta whereas the pH at Tönnersjöheden did not appear to differ between treatments (Figure 5). A similar pattern was found for ANC and  $\text{Ca}^{2+}$  (Figure 6 and Figure 9). The  $\text{SO}_4^{2-}$  concentration on the other hand, differed between the WTH-and CH-plots at Tönnersjöheden and Lövliden, but not at Kosta (Figure 7). Thus, the main treatment effect of harvest intensity is long-term but of limited duration (Table 6).

The results from the present study are in agreement with previous soil studies at the same sites, showing that WTH caused lower pH of the organic horizon and the upper mineral soil layers, lower base saturation and higher exchangeable acidity compared to CH (Nykqvist and Rosén, 1985; Olsson et al., 1996a; Staaf and Olsson, 1991). A soil study made prior to the present soil water study indicated that those effects were still present, except for soil pH (Olsson et al. 2004). However, the magnitude of previously found treatment effects on soil had decreased or almost diminished, and the still present effects on soil Ca pools were most pronounced at Lövliden.

Initial treatment differences in base cation concentrations between WTH and CH may result from decomposition and mineralization of logging debris. For example, several studies have shown that the leaching of cations is lower during the early post-harvest period following WTH in comparison to conventionally harvested areas where logging residues are left on site (e.g. Rosén and Lundmark-Thelin, 1987; Staaf and Olsson, 1994; Westling et al., 2004) although there are reports of no treatment effects at all (Wall, 2008) or even higher  $\text{Ca}^{2+}$  leaching following WTH (Hendrickson et al., 1989). With time, changes in e.g. weathering rates, nutrient uptake by the new stand, differences in leaching and dry deposition may reinforce or weaken the effects in the long run.

In the present study, the main treatment effect had disappeared by the end of the second period (2008-2010). However, a statistically significant interaction effect between site\*treatment for  $\text{Ca}^{2+}$  still existed at Lövliden ( $p=0.0022$ ) suggesting a longer-lasting effect of WTH in comparison to Tönnersjöheden and Kosta. The observed treatment differences in soil water  $\text{Ca}^{2+}$  concentrations between WTH and CH are likely to be the result of different levels of biomass removal. Thus the removal of logging residues has resulted in a long-term, but temporary decrease in exchangeable  $\text{Ca}^{2+}$ . It is possible that a change in weathering rate or deposition patterns have modified or partly compensated the treatment effect on soil water chemistry, but these processes have apparently not been able to offset the treatment effect. Treatment effects on ANC and pH probably arose as a result of differences in  $\text{Ca}^{2+}$  concentrations, but are not further discussed.

The treatment effect appeared to differ between sites. The long-term effect of WTH on soil water  $\text{Ca}^{2+}$  concentrations at Lövliden is likely to have been caused by greater amounts of Ca being removed from the site (426 kg per hectare) in comparison to Tönnersjöheden (241 kg per hectare) and Kosta (278 kg per hectare) (Table 2). The higher biomass  $\text{Ca}^{2+}$  content at Lövliden is likely to be the result of greater Ca-pools in the humus layer and upper mineral soil (Olsson et al., 1996a) and stand age (Table 2). Via transpiration,  $\text{Ca}^{2+}$  ions are taken up trees and used in cells for metabolism and other functional roles, but significant amounts of excess  $\text{Ca}^{2+}$  are deposited in the bark or as extracellular calcium oxalate crystals in the foliage (e.g. Fink, 1991; Hudgins et al., 2003; Tillman-Sutela and Kauppi, 1999). At Lövliden, this has resulted in higher  $\text{Ca}^{2+}$  concentrations in the biomass as the stand grew old (Table 2). This excess uptake of  $\text{Ca}^{2+}$  may have a greater impact

on ecosystem nutrient cycling which may be overlooked if only the functional need of forest trees is considered.

The difference in soil water  $\text{SO}_4^{2-}$  concentration between the CH- and WTH-plots is more difficult to explain. Compare with Cl, sulfate is more tightly held in the soil and depends on biological and chemical processes (Johnson and Cole, 1980). Thus, some soils are able to retain  $\text{SO}_4^{2-}$ . One important factor which has been shown to be important is pH, with  $\text{SO}_4^{2-}$  retention inversely related to pH. In addition, sulfate retention is reversible (Sanger et al., 1994). For example, Nodvin et al. (1986) studied the effects of pH and ionic strength on  $\text{SO}_4^{2-}$  adsorption by using soil samples from a podzolic B-horizon. Depending on  $\text{SO}_4^{2-}$  concentration and pH in the solution, the net removal or release of  $\text{SO}_4^{2-}$  varied according to Figure 13. A small change in pH can thus significantly increase the  $\text{SO}_4^{2-}$  retention at low pH-values. However, in soils with high pH-values (>6) the retention of  $\text{SO}_4^{2-}$  is low.

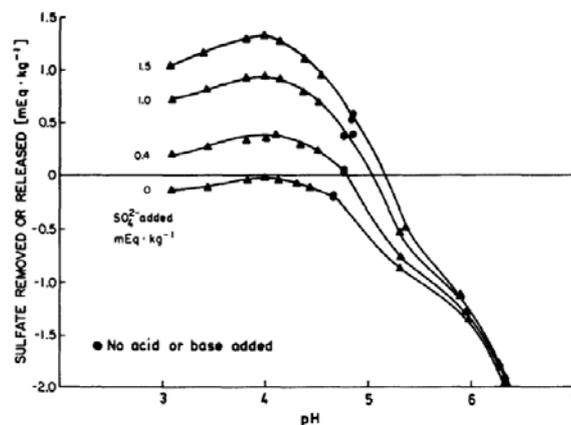


Figure 13. The net retention (positive values) or release (negative values) of  $\text{SO}_4^{2-}$  as a function of soil solution concentration and pH. From Nodvin et al. (1986).

In this study, soil water pH was significantly lower in WTH-plots 2003 and 2005 (Table 5). However, by 2008-2010 the treatment effect was gone (Table 6). A lower pH in WTH-plots could have led to increased  $\text{SO}_4^{2-}$  retention in the soil and lower soil water concentrations at the southern sites. However, at the northern site, soil water pH varies around 6 (Figure 5). At this level  $\text{SO}_4^{2-}$  retention is negligible (Figure 13).

An alternative explanation could lie in canopy differences between the stands. For example, Egnell (2011) found a statistically significant difference in stand growth between CH- and WTH-plots in Lövliden, 31 years following cutting. The basal area in the WTH plots averaged 10.5 m<sup>2</sup> per hectare in comparison to 14.0 m<sup>2</sup> per hectare at the CH plots. It is possible that these differences would be responsible for a less dense tree canopy in the WTH-plots which in turn could lead to lower dry deposition (and soil water concentrations). However, in areas with low S deposition, the relative importance of dry deposition for measured soil water concentrations must be questioned. Other factors known to affect soil water concentrations of  $\text{SO}_4^{2-}$  are dilution following reduced evapotranspiration rates (Hornbeck and Kropelin, 1982) and anaerobic conditions leading to S reductions (Löfgren et al., 2009). Since the soils are freely drained, the latter two processes are not likely to be of importance at Lövliden. Finally, differences in element cycling of S (i.e. plant uptake, growth rates, litter fall and decomposition) could be responsible.

## **4 Implications for practical forestry and future research**

The results from long-term field experiments can be used to discuss the need for ash recycling. According to the Swedish Forest Agency (2008) the aim of ash recycling is to maintain forest sustainability by e.g. counteracting soil and surface water acidification following WTH. In this study, WTH resulted in measurable changes in the chemical composition of soil water which is in agreement with previous studies on soil chemistry, tree growth and nutrition levels carried out at these sites. When combined, these results indicate that WTH effects are distributed throughout various parts of the forest ecosystem at decadal time scales. The effect of ash recycling is likely to be distributed in the ecosystem at a similar pace as when removing logging residues. Thus, based on our findings, ash products used for compensating base cations should have a dissolution rate of at least 30 years if counteracting soil water acidification, and possibly, run-off water is a primary objective.

Another issue related to ash recycling is to what extent the observed effects of WTH on soil water chemistry can be used as a motive for ash-recycling? In our view, the relationship between magnitude of change and the need for ash-recycling is more likely to be negatively than positively related. The reason to this is that large treatment differences between WTH and CH on soil water exchangeable base cation concentrations may be a consequence of higher base saturation and greater surplus of base cations in relation to the demand from growing trees. For example, the largest treatment differences took place at a well-buffered site in northern Sweden (Lövliden) where treatment effects were seen both in soil and soil water chemistry. At this site, WTH have caused significant growth reduction in comparison to CH as a result of lower N availability (Olsson et al. 1999, Egnell 2011) and not likely due to lower concentrations of base cations. Furthermore, the effects of WTH on the soil and soil water chemistry at Lövliden have taken place within a range of moderate soil acidity. If the primary aim of ash-recycling is to prevent soil and surface water acidification in strongly acidified areas, ash recycling at Lövliden may seem as an unnecessary measure considering the relatively well-buffered soils.

The southern sites have suffered historically from acid deposition, depleting the soils of buffer capacity, superimposed by forestry. If we would use the less pronounced effects at the southern sites as an indicator, we could perhaps argue that there is little need for ash-recycling. However, even a small change at these sites could prevent the soils from fully restoring their buffer capacity. In addition, there is a risk that acidification effects prior to this investigation are overlooked, thereby underestimating the effect of WTH because of lack of soil water data. We may also underestimate the need for ash-recycling if only taking soil water chemistry into account since the effect of WTH is recognized in other parts of the ecosystem. In conclusion, the variation in treatment effects (i.e. the magnitude of change) may not be the best measure when deciding upon ash-recycling and should therefore be used with caution. Instead, the need for ash-recycling at a particular site must be viewed in a greater context.

The results from this study can also be used to validate computer simulations used to predict clear-cutting effects over one or two rotations periods. According to recent model simulations, WTH will lead to a long-term re-acidification of soils and lakes despite further emission reductions (Aherne et al., 2008). Our results are partly in agreement with the model simulation since WTH resulted in a temporary loss of base cations and more acid soil water. Whether or not the treatment difference was large enough to jeopardize the long-term sustainability of the forest ecosystem can be discussed. Furthermore, the treatment differences have disappeared with time (<31 years) which

suggests that the effect of WTH on soil and soil water chemistry is temporary as oppose to the model simulations forecasts which points at treatments effects lasting more than (100 years (Aherne et al., 2008)).

Future studies could help us to explain differences in treatment response between the sites by including information about weathering rates, decomposition, deposition and organic matter content of chlorine. In addition, the input and output of nutrients should also be quantified in order to establish nutrient budgets for each individual site. This would be helpful when discussing the magnitude of change, the risk for unwanted effects and need for ash recycling. Data from these sites could also be used to discuss the choice of parameters needed to decide upon the need for ash recycling. Finally, with enough information, the study sites offer a unique chance to model the long-term effects of biomass removal on surface water acidity using MAGIC.

## **5 Conclusions**

The experimental sites used in this study belong to the oldest in the world and provided us with a unique opportunity to study the long-term effects of biomass removal on soil water chemistry, 27 to 35 years after harvest. Previous studies on soil chemistry offered additional insight which helped us to explain treatment differences. The most important findings are listed below:

- The hypothesis ( $H_0$ ) of no differences in soil water concentrations between WTH and CH was rejected. In particular, the soil water soil water  $Ca^{2+}$  concentrations at 50 cm depth were lower following WTH in comparison to CH. In addition, WTH resulted in lower soil water pH and ANC. Treatment effects were also noted for  $SO_4^{2-}$  that could be the combined results of lower dry deposition in WTH-plots and/or increased retention in the mineral soil.
- The main treatment differences are of long-term (27-30 years) but limited duration since they had disappeared during the second study period between 2008 and 2010 (i.e. 32 to 35 years after harvest).
- The results are in agreement with previous soil studies carried out at the same sites.
- Although examining temporal trends was not a primary focus of this study, all three sites showed recovery from acidification measured as an increase in pH and ANC and a decrease in  $SO_4^{2-}$  deposition and soil water concentrations. The recovery process at the southern sites was temporary offset by sea salt events in 2005-2007.
- The results from long-term field experiments can be used to validate computer simulations used to predict clear-cutting effects over one or two rotations periods. The results can also be used to discuss the need for ash-recycling. Future studies could help us to explain differences in treatment response between the sites by including information about weathering rates, decomposition, deposition and organic matter content of e.g. chlorine.

## 6 Acknowledgement

We are very grateful to the Swedish Energy Agency for their financial support. We would also like to thank Karol Koos, Hans-Göran Nilsson and Morgan Erixon for their field work and Claudia von Brömssen for the statistical analysis. We would also like to express our sincere gratitude to SLU for allowing us access to the research sites. We are also thankful to Johan Berg who provided useful and helpful comments on an earlier version which helped to improve the report. Finally, deposition data were kindly being made available by The Swedish Throughfall Monitoring Network which is run by IVL Swedish Environmental Research Institute and financed by regional air quality protection associations, county administrative boards and the Swedish Environmental Protection Agency.

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## Appendix 1. SO<sub>4</sub>-S and Cl deposition between 1987 and 2009



Figure 14. Deposition of non-marine SO<sub>4</sub>-S and Cl in throughfall and open field, collected at nearby stations part of the Throughfall Monitoring Network (<http://www.krondroppsnetet.ivl.se/>). Timrilt is a spruce dominated forest close (<10 kilometres) to Tönnersjöheden. Attsjö is a pine dominated forest located circa 17 kilometres from Kosta while Bäcksjö (a spruce forest) is situated circa 60 kilometres southeast of Lövliden.

## Appendix 2. Soil chemical characteristics between 2002 and 2004

pH (H<sub>2</sub>O) from Olsson et al., 2004.

	Tönnersjöheden		Kosta		Lövliden	
	CH	WTH	CH	WTH	CH	WTH
FH	3.89	3.81	4.05	4.02	3.90	3.92
0-5	3.94	3.88	3.78	3.79	4.04	4.10
5-10	4.20	4.19	4.19	4.28	4.65	4.55
10-15	4.43	4.43	4.47	4.51	5.20	5.04
15-20	4.71	4.60	4.60	4.51	5.55	5.50

Exchangeable base cations (kmol<sub>c</sub> ha<sup>-1</sup>) from Olsson et al., 2004.

	Tönnersjöheden		Kosta		Lövliden	
	CH	WTH	CH	WTH	CH	WTH
FH	5.24	4.75	10.99	9.80	6.76	5.97
0-5	1.93	1.88	2.98	2.38	2.73	2.22
5-10	0.77	0.68	0.98	0.68	2.08	1.52
10-15	0.50	0.41	0.64	0.43	2.36	1.39
15-20	0.53	0.40	0.32	0.29	1.68	1.01

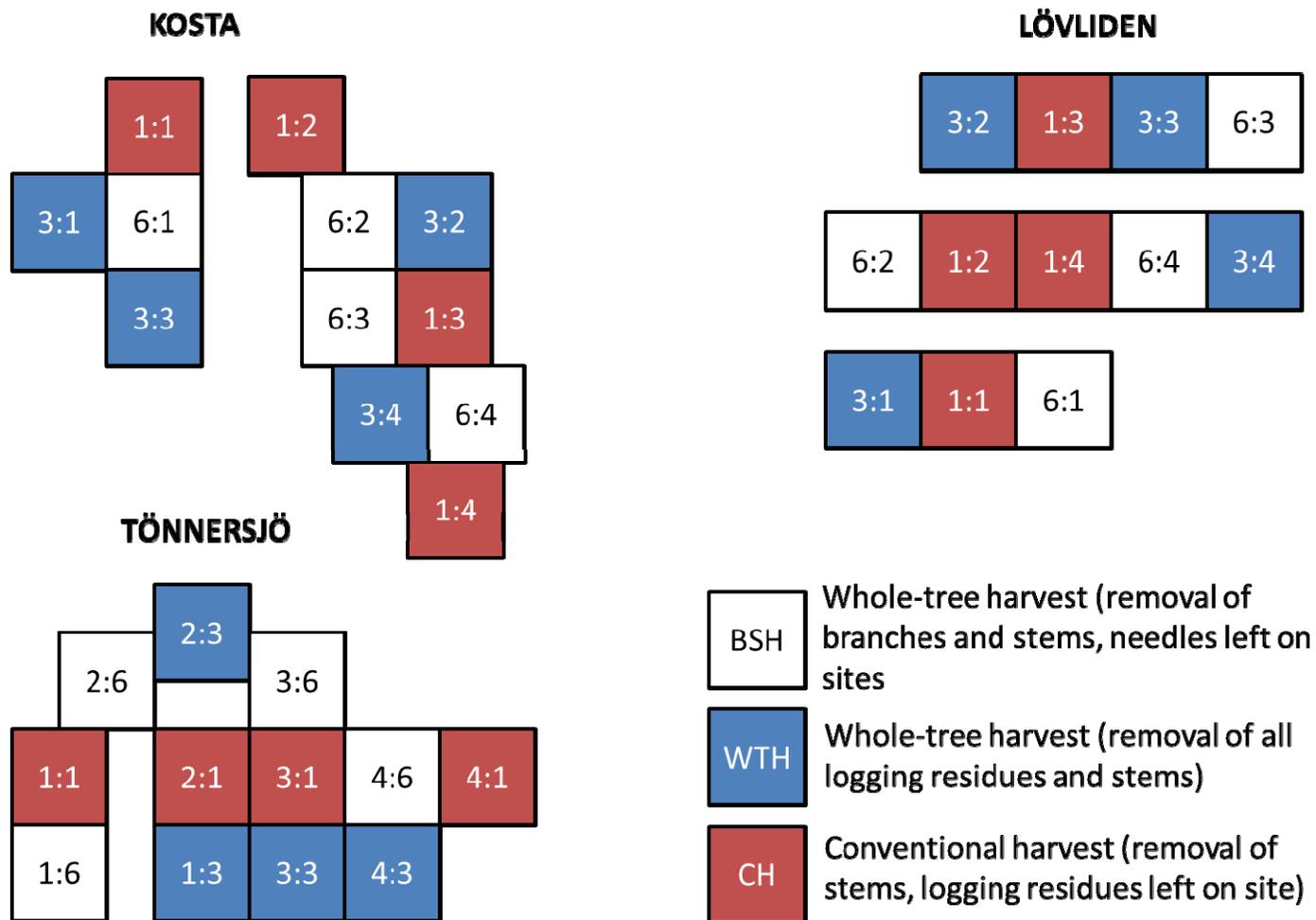
Base saturation (%) from Olsson et al., 2004.

	Tönnersjöheden		Kosta		Lövliden	
	CH	WTH	CH	WTH	CH	WTH
FH	49.6	43.8	53.6	46.4	79.8	76.6
0-5	13.4	12.6	22.6	18.7	41.2	30.3
5-10	5.6	5.1	8.8	6.8	28.2	20.6
10-15	4.6	4.2	8.4	5.9	47.7	40.4
15-20	5.8	5.3	8.6	7.3	82.0	72.7

Exchangeable acidity (µmole H<sup>+</sup> per gram dry substance) from Olsson et al., 2004.

	Tönnersjöheden		Kosta		Lövliden	
	CH	WTH	CH	WTH	CH	WTH
FH	46.6	50.1	51.0	64.4	17.0	18.2
0-5	28.0	27.8	38.2	39.1	10.7	13.3
5-10	30.1	28.2	40.6	37.7	13.8	15.2
10-15	27.7	24.8	-	-	5.6	5.4
15-20	12.3	13.3	-	-	1.1	1.1

### Appendix 3. Experimental design



## Appendix 4. Soil water analysis

Parameter	Analytical method	Name	Measuring range without dilution	Instrument
<b>pH</b>	SS 02 81 22, utg. 2	Determination of pH-value of water	3-8 units	<b>Electrode</b>
<b>Alkalinitet</b>	SS-EN ISO 9963-2, utg. 1, mod.	Determination of alkalinity – Part 2: Determination of carbonate alkalinity (ISO 9663-2:1994).	0,01-4 mmole/l	<b>Titration</b>
<b>Sulfate (SO<sub>4</sub><sup>2-</sup>-S)</b>	SS-EN ISO 10 304-1, utg. 1	Determination of dissolved fluoride, nitrite, orthophosphate, bromide, nitrate and sulfate ions, using liquid chromatography of ions – Part 1_ Method for water with low contamination (ISO 10304-1:1992).	0,01-28 mg S/l	<b>Ion chromatography (IC)</b>
<b>Chloride</b>	SS-EN ISO 10 304-1, utg. 1	See sulfate	0,05-32 mg/l	<b>Ion chromatography (IC)</b>
<b>Nitrate (NO<sub>3</sub>-N)</b>	SS-EN ISO 10 304-1, utg. 1	See sulfate	0,01-5,6 mg/l	<b>Ion chromatography (IC)</b>
<b>Calcium</b>	SS-EN ISO 14911, utg. 1	Determination of dissolved Li <sup>+</sup> , Na <sup>+</sup> , NH <sub>4</sub> <sup>+</sup> , K <sup>+</sup> , Mn <sup>+</sup> , Ca <sup>2+</sup> , Mg <sup>2+</sup> , Sr <sup>2+</sup> and Ba <sup>2+</sup> using ion chromatography – Method for water and waste water (ISO 14911:1998).	0,1-36 mg/l	<b>Ion chromatography (IC)</b>
	(SS 28161) <sup>a,b</sup>	Atomic absorption spectrometry – atomization in flame – special guidelines for calcium and magnesium.	0,1-6 mg/l	<b>Atomic Absorption Spectroscopy (AAS)</b>
<b>Magnesium</b>	SS-EN ISO 14911, utg. 1	See calcium	0,05-18 mg/l	<b>Ion chromatography (IC)</b>
	(SS 28161) <sup>a,b</sup>	See calcium	0,05-1	<b>Atomic Absorption Spectroscopy (AAS)</b>
<b>Sodium</b>	SS-EN ISO 14911, utg. 1	See calcium	0,12-15 mg/l	<b>Ion chromatography (IC)</b>
<b>Potassium</b>	SS-EN ISO 14911, utg. 1	See calcium	0,1-35 mg/l	<b>Ion chromatography (IC)</b>
<b>Manganese</b>	SS-EN ISO 14911, utg. 1 <sup>a</sup>	See calcium	0.03-4.5 mg/l	<b>Ion chromatography (IC)</b>
<b>Ammonium (NH<sub>4</sub><sup>+</sup>-N)</b>	SS EN ISO 11732, utg. 1, mod.	Determination of ammonium nitrogen by flow analysis (CFA and FIA) and spectrometric detection (ISO 11732:1997).	0,02-10 mg/l	<b>Flow Injector Analysis (FIA)</b>
<b>Aluminium fractioning</b>	According to Driscoll (1984)	A cation exchange method whereby total aluminium is divided into organic and inorganic aluminium	-	<b>Ion exchange column</b>
<b>Total and organic aluminium</b>	SS-EN ISO 11885, utg. 1,	Determination of 33 elements by inductively coupled plasma atomic emission spectroscopy (ISO 11885:1996)	5-40 µg/l	<b>ICP AES</b>
<b>Iron</b>	SS-EN ISO 11885, utg. 1,	See total aluminium	5-40 µg/l	<b>ICP AES</b>
<b>Total organic carbon (TOC)</b>	SS-EN 1484, utg. 1,	Guidelines for the determination of total organic carbon (TOC) and dissolved organic carbon (DOC)	0,5-20	<b>Shimatzu TOC 5050 med provinjektor ASI-502.</b>

<sup>a</sup>Not accredited according to the Swedish Board for Accreditation and Conformity Assessment (SWEDAC).

<sup>b</sup>Between 2008-2009, analysis of calcium and manganese was carried out using an atomic absorption spectroscopy at pH≥5.5.

## Appendix 5. Activity documentation

Kosta			Tönnersjöheden			Lövliden		
Date	Measure	Season	Date	Measure	Season	Date	Measure	Season
2002-10-04	Installation A & B		2002-11-05	Installation A		2002-10-11	Installation A & B	
2003-06-02	Rinsing (A & B)		2003-06-05	Installation B		2003-05-29	Rinsing (A & B)	
2003-06-09	Rinsing (A & B)		2003-06-06	Rinsing (A & B)		2003-06-02	Rinsing (A & B)	
2003-06-12	Rinsing (A & B)		2003-06-10	Rinsing (A & B)				
2003-06-09	Sampling (A & B) <sup>a</sup>	Summer	2003-06-10	Sampling (A & B) <sup>a</sup>	Summer	2003-06-06	Sampling (A & B) <sup>a</sup>	Summer
						2003-06-23	Sampling (A & B) <sup>a</sup>	Summer
2003-11-14	Sampling (A & B) <sup>a</sup>	Autumn	2003-11-18	Sampling (A & B) <sup>a</sup>	Autumn	2003-10-20	Sampling (A & B) <sup>a</sup>	Autumn
			2003-12-16	Sampling (A & B) <sup>a</sup>	Winter			
2004-04-22	Sampling (A & B) <sup>a</sup>	Spring	2004-04-15	Sampling (A & B) <sup>a</sup>	Spring	2004-05-10	Sampling (A & B) <sup>a</sup>	Spring
2004-09-27	Sampling (A & B) <sup>a</sup>	Autumn	2004-10-04	Sampling (A & B) <sup>a</sup>	Autumn	2004-10-04	Sampling (A & B) <sup>a</sup>	Autumn
2005-05-11	Sampling (A & B) <sup>a</sup>	Spring	2005-05-09	Sampling (A & B) <sup>a</sup>	Spring	2005-05-23	Sampling (A & B) <sup>a</sup>	Spring
2005-08-15	Sampling (A & B) <sup>a</sup>	Summer	2005-08-17	Sampling (A & B) <sup>a</sup>	Summer	2005-08-16	Sampling (A & B) <sup>a</sup>	Summer
2005-10-27	Sampling (A & B) <sup>a</sup>	Autumn	2005-10-24	Sampling (A & B) <sup>a</sup>	Autumn	2005-10-17	Sampling (A & B) <sup>a</sup>	Autumn
2008-04-28	Installation C, D & E Rinsing (A-E)		2008-05-08	Installation C, D & E Rinsing (A-E)		2008-07-09	Installation C, D & E Rinsing (A-E)	
2008-09-08	Rinsing (A-E)		2008-09-16	Rinsing (A-E)		2008-09-15	Rinsing (A-E)	
			2008-10-07	Rinsing (A-E)		2008-10-02	Rinsing (A-E)	
2008-10-08	Sampling (A-E) <sup>b</sup>	Autumn	2008-10-23	Sampling (A-E) <sup>b</sup>	Autumn	2008-10-13	Sampling (A-E) <sup>b</sup>	Autumn
2009-05-11	Sampling (A-E) <sup>b</sup>	Spring	2009-05-25	Sampling (A-E) <sup>b</sup>	Spring	2009-05-25	Sampling (A-E) <sup>b</sup>	Spring
2009-07-22	Sampling (A-E) <sup>b</sup>	Summer	2009-07-20	Sampling (A-E) <sup>b</sup>	Summer	2009-07-20	Sampling (A-E) <sup>b</sup>	Summer
2009-10-21	Sampling (A-E) <sup>b</sup>	Autumn	2009-10-19	Sampling (A-E) <sup>b</sup>	Autumn	2009-10-12	Sampling (A-E) <sup>b</sup>	Autumn
2010-04-12	Sampling (A-E) <sup>b</sup>	Spring	2010-04-12	Sampling (A-E) <sup>b</sup>	Spring	2010-06-07	Sampling (A-E) <sup>b</sup>	Spring

<sup>a</sup>Composite sample of A+B

<sup>b</sup>Discrete samples

## Appendix 6. Differences of least square means for statistically significant interaction effects (site\*treatment)

Time period	Variable	Effect	Site	Treatment	p
2003-2005	ANC	site*treatment	KO	CH vs WTH	0.0572
		site*treatment	LL	CH vs WTH	0.0013
		site*treatment	TS	CH vs WTH	0.3736
2003-2005	K <sup>+</sup>	site*treatment	KO	CH vs WTH	0.9627
		site*treatment	LL	CH vs WTH	0.0128
		site*treatment	TS	CH vs WTH	0.2680
2008-2010	SO <sub>4</sub> <sup>2-</sup>	site*treatment	KO	CH vs WTH	0.2679
		site*treatment	LL	CH vs WTH	<.0001
		site*treatment	TS	CH vs WTH	0.8333
2008-2010	Ca <sup>2+</sup>	site*treatment	KO	CH vs WTH	0.8215
		site*treatment	LL	CH vs WTH	0.0022
		site*treatment	TS	CH vs WTH	0.8873
2008-2010	Al <sub>o</sub>	site*treatment	KO	CH vs WTH	0.0124
		site*treatment	LL	CH vs WTH	0.7625
		site*treatment	TS	CH vs WTH	0.2661

## Appendix 7. Soil water chemistry

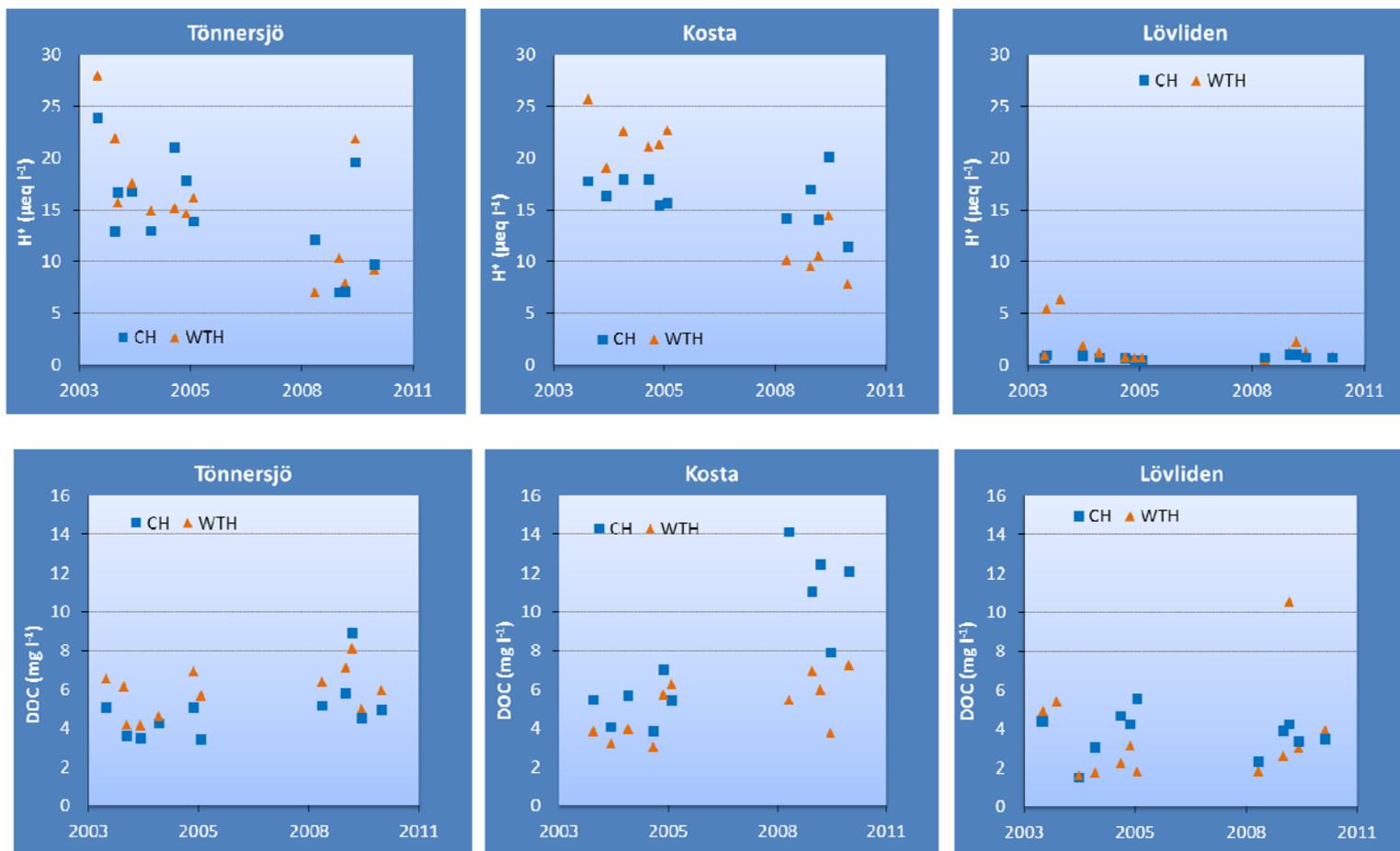


Figure 15. H<sup>+</sup> and DOC-concentrations (volume weighed means) in soil water at 50 cm depth (~B/C-horizon). Legends as in Figure 5.

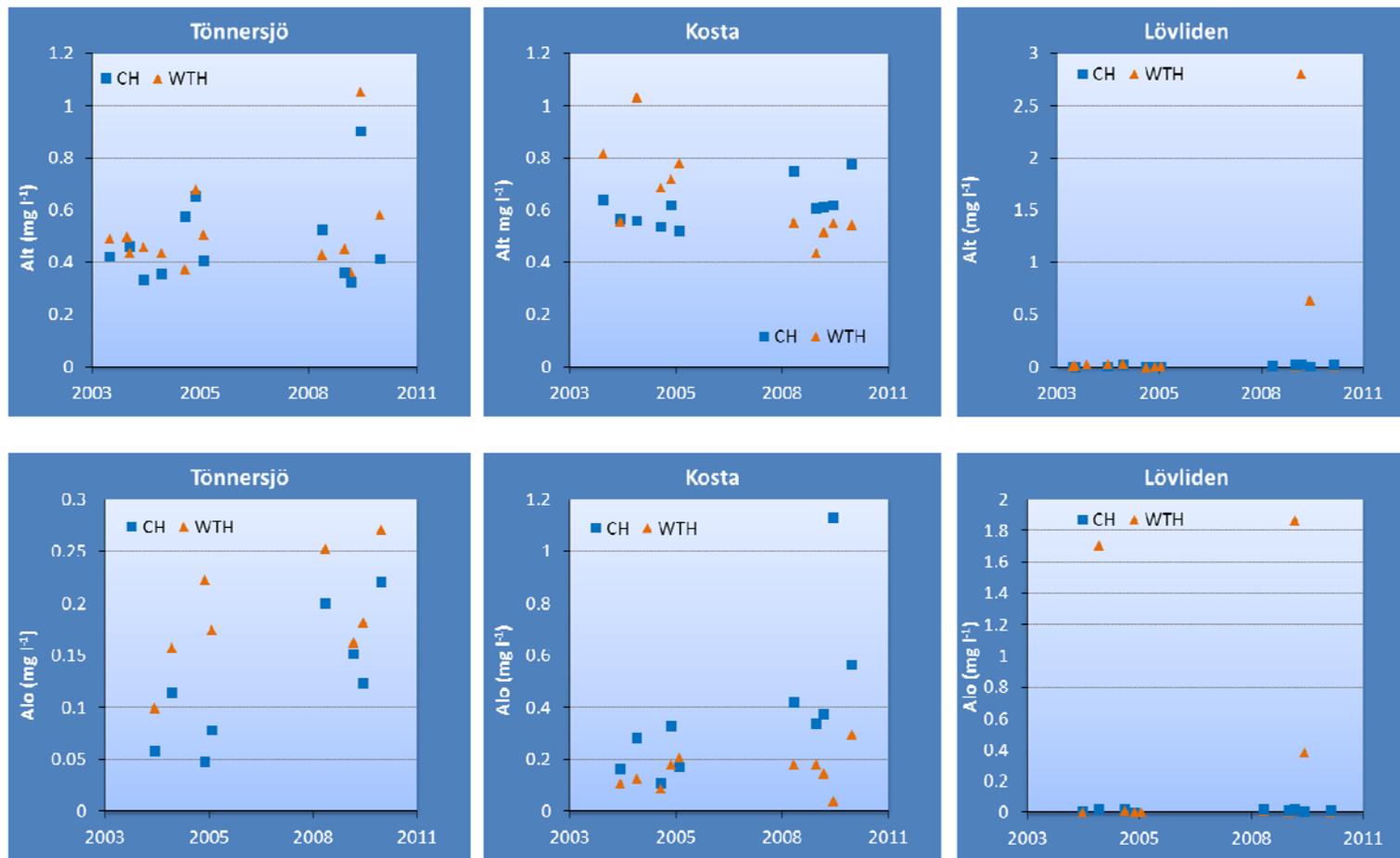


Figure 16. Alt- and Alo-concentrations (volume weighed means) in soil water at 50 cm depth (~B/C-horizon). Legends as in Figure 5.

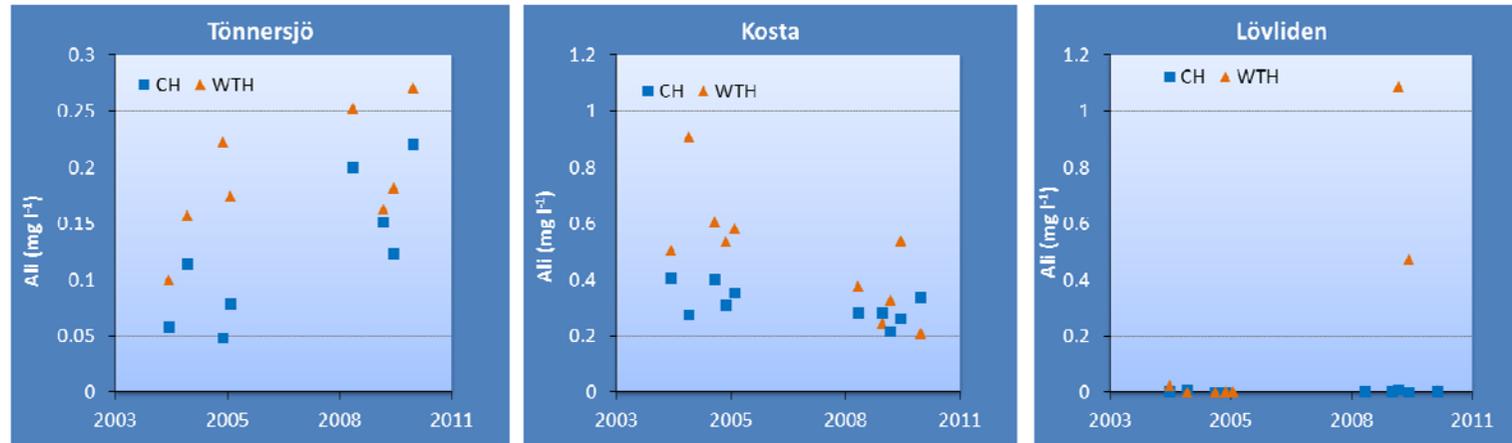


Figure 17. Ali-concentrations (volume weighed means) in soil water at 50 cm depth (~B/C-horizon). Legends as in Figure 5.