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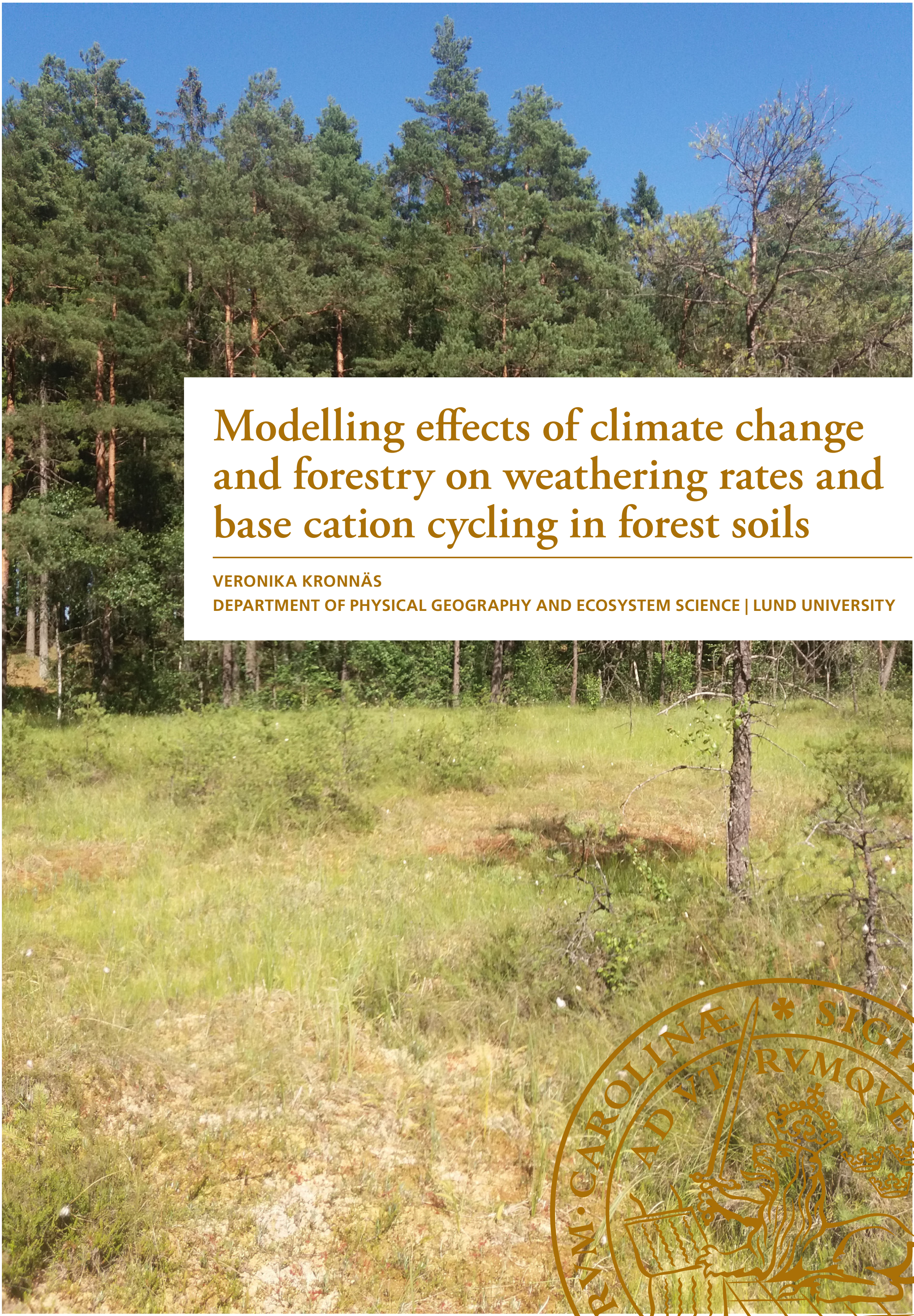

Modelling effects of climate change and forestry on weathering rates and base cation cycling in forest soils

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DOCTORAL DISSERTATION

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Abstract

Base cations are important in forest soils as nutrients for vegetation and protection against soil acidification. Today, base cations have been lost from soils through acidification and forest harvesting. With low levels in soils, temporary situations of deficiency could occur. One of the largest inputs of base cations to soils is minerals weathering, which makes it an important process. It is, however, hard to measure. By using process-based models, weathering rates can be calculated and used in estimates of the sustainability of forestry with regards to base cation stores. In a changing environment, dynamic process-based models can give important insights about changes in weathering and base cation cycling.

In this thesis, the two process-based models, PROFILE and ForSAFE, have been used to describe the effects of a changing climate, acidification and intensified forestry on weathering and base cation cycling. Based on the same weathering process descriptions, PROFILE is a simpler steady-state model, while ForSAFE is a more complex dynamic model with feedback, for example between vegetation and soil chemistry. These results contribute to the existing knowledge about sustainable forestry and add new knowledge about the seasonal effects of weathering.

The studies show that weathering rates modelled using PROFILE and long-term averages from ForSAFE are approximately the same size, if soil-moisture levels are equal. However, ForSAFE studies can provide results with high temporal resolution. Depending on research question and data availability, both models can be useful. ForSAFE can increase understanding of processes and dynamics in a changing environment at well-investigated sites. PROFILE can be used for areal upscaling and to assess long-term base cation sustainability of forestry.

ForSAFE results show that weathering rates have a strong seasonal dynamic, with low winter rates and high and highly variable summer rates, depending on temperature and soil moisture. Weathering increases temporarily after clear-cutting, as both concentrations of weathering products in the soil water and the soil moisture levels are affected. Removal of branches and tree-tops, in addition to stems, has only a small effect on weathering, much smaller than the loss of base cations through the harvesting.

Climate change increases weathering in the Swedish climate. In southern Sweden, rates increase throughout the year, though most in spring and summer. Recurrent low soil moisture during summers already has an inhibiting effect on weathering. This is projected to continue. In northern Sweden, winter temperatures will still mostly be below zero, and weathering rates will only increase in the warmer seasons. The increasing weathering in spring might become increasingly important for vegetation. Vegetation period lengthens in a warming climate and dry summers (especially in the south) might push growth and nutrient uptake to earlier in the year.

PROFILE-modelled weathering, with harvest removal of base cations, was used to calculate exceedance of critical biomass harvesting. This, together with acidification status of the soil, was used to assess if whole-tree harvesting would be detrimental to the acidification situation of the soils. Results show that soils generally are more at risk in southern Sweden, with a couple of exceptions where high weathering provides enough base cations.

These studies confirm that soil moisture is one of the bigger uncertainties in weathering calculations and an important factor in modelling future biomass growth. Soil moisture is not measured at the modelled sites. Future studies would be needed, to measure soil moisture and compare it with ForSAFE-modelled soil moisture to reduce uncertainties related to soil moisture in PROFILE and ForSAFE. For ForSAFE, this is needed in areas with flat surroundings, where runoff is not as efficient as ForSAFE models it to be. For PROFILE, it is most relevant in sites with differing textures in different soil layers.

Key words weathering, forest, biogeochemistry, dynamic modelling, ForSAFE

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Author contributions

I	VK was involved in the design of the study. VK prepared the model inputs and model parameterizations supported by the other authors. VK performed the model simulation and data analysis. VK wrote the paper with contributions from the other authors.
II	VK conceptualised the study with guidance from the other authors. VK together with other authors developed the model version, prepared the model inputs and model parameterizations. VK performed the model simulation and data analysis. VK wrote the paper with contributions from the other authors.
III	VK was involved in preparing the model inputs for the sites and validating the results. VK contributed to writing the paper.
IV	VK was responsible for the weathering calculations. VK was involved in developing the methodology, validating the results, curating data and making the formal analysis and contributed to writing the paper.

Abstract

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Sammanfattning

Kemisk vittring kallas processen när mineralkorn löses upp och sönderdelas i molekyler eller atomer. Det är en viktig process, bland annat för att den frigör ämnen som skyddar marken från försurning och som fungerar som näringsämnen för växter. För de så kallade baskatjonerna kalium, kalcium, magnesium och natrium är vittringen i marken bland de största källorna. Framför allt de tre förstnämnda baskatjonerna är viktiga näringsämnen för växter och baskatjoner skyddar också marken från markförsurning. Svensk berggrund, och därmed svenskt jordtäcke, innehåller mest kiselrika långsamvittrade mineral, med ganska låga halter av baskatjoner. Det kan därför finnas ganska låga halter av tillgängliga baskatjoner i marken, särskilt i områden som har utsatts för försurning eller där skogen är mycket produktiv. Produktiv skog tar upp mycket baskatjoner, som förs bort från skogen vid skörd. Södra Sverige, särskilt sydvästra delen, är oftast både mycket produktiv och har varit utsatt för försurning.

Vittringshastigheten varierar mellan olika mineral och beroende på omgivning, framför allt temperatur och markfuktighet. I svenskt klimat är den vanligen mycket högre på sommaren än på vintern. Andra faktorer som påverkar är kornstorlek (med högre vittring i finkorniga jordar med hög sammanlagd mineralyta), samt tillgång på de ämnen som mineralkornen reagerar med under vittringsprocesserna: vattnet i sig, koldioxid, vätejoner, hydroxidjoner (fast de sistnämnda finns bara i väldigt låga koncentrationer i jordar under svenska förhållanden) och organiska syror.

Vittring är svårt att mäta utanför labbmiljö och därför finns i stället olika metoder och modeller för att beräkna vittringen. I den här studien har två modeller använts, som beräknar vittringen utifrån de kemiska reaktioner som bidrar till vittringen. Den ena, PROFILE, behöver mindre data och ger ett medelvärde av vittringen för en viss modellerad mark, utan hänsyn till variation i tid. Den andra, ForSAFE, använder mer data och inkluderar fler processer, såsom hur vatten från nederbörden infiltreras i marken och fuktar den, trädens tillväxt och nedbrytning av trädens förna. Den visar dessutom utvecklingen i tid av vittringen.

I avhandlingens första artikel jämförs vittringsresultat för de två modellerna, för två skogsytor i mellanskåne, Västra Torup och Hissmossa. I denna artikel visas också, med hjälp av ForSAFE-modellen, hur vittringen förändras av en framtida klimatförändring, modellerad efter ett klimatscenario från IPCC (Intergovernmental Panel on Climate Change), hur den påverkas av ett mer eller mindre intensivt skogsbruk och hur den påverkades av försurningen som ägde rum under andra halvan av 1900-talet. Studien visar att PROFILE och ForSAFE ger liknande medelvittring, om nivåerna av markfuktighet är lika mellan modellerna. PROFILE behöver få markfuktighetsnivån som indata, medan ForSAFE modellerar den.

Enligt modellen kan vittringen förväntas öka i ett framtida varmare klimat, särskilt om marken inte blir försurad och därför får höga halter av löst aluminium som minskar vittringen.

I den andra artikeln används ForSAFE-modellen på sju ytor, spridda i södra och norra Sverige, med ett klimatscenario från IPCC och ett scenario där det förutom klimatförändringen dessutom inträffar svår sommartorka fem år i rad. Resultaten visar att trots att temperaturen skiljer mycket från södra till norra Sverige och vittringen är så temperaturberoende, så finns det ingen gradient i vittring från söder till norr. Det beror på att skillnader i markens kornstorleksfördelning och vilka mineral som dominerar har större betydelse än temperaturen. Även i den här artikeln dras slutsatsen att vittringen ökar när temperaturen ökar i det framtida klimatscenariet, även om sommartorka tillfälligt kan göra att sommarvittringshastigheten blir lika låg som den brukar vara under vintrarna. I södra Sverige ökar vittringen året runt när temperaturen ökar, om än mest under vår och sommar. I norra Sverige ökar temperaturen mycket under vintern, men det har nästan ingen effekt på vittringen, eftersom temperaturen ändå oftast kommer att vara under noll och då är vittringen mycket låg. Därför sker ökningen i norra Sverige under de varma årstiderna. I torkscenariet är vittringen på helårsbasis bara 78 % av vad den skulle varit utan torkan, i den mest påverkade ytan.

Den tredje artikeln undersöker effekten av biobränsleuttag från skogen. Skillnaden i baskatjonhalter i markvatten jämförs för skogsavverkning med uttag av grenar och toppar till skogsbränsle (så kallat helträdsuttag) och för avverkning med bara stamskörd (där grenar och toppar får ligga kvar i skogen). ForSAFE-modellen används på sex av de ytor som undersöktes i artikel två. De första trettio åren efter avverkning (oavsett om grenar och toppar tas ut eller ej) är koncentrationerna av baskatjoner högre än i den ej avverkade skogen, eftersom kvarlämnat organiskt material bryts ner och frisläpper baskatjoner successivt, samtidigt som det inte finns några stora träd som tar upp stora mängder baskatjoner längre. I den helträdsavverkade skogen är baskatjonhalterna dock inte lika förhöjda som i skogen med stamuttag. Halterna av kväve, som är det viktigaste växtnäringsämnet, är också mycket lägre i markvattnet i den helträdsavverkade skogen och därför växer inte den nya planterade skogen lika snabbt, särskilt inte i de nordliga ytorna där det finns mindre kväve. Skillnaderna blir, enligt modellen, mindre med tiden och efter trettio år är koncentrationerna av baskatjoner och kväve i marken lika oavsett om grenar och toppar tas u teller ej. Träden planterade på ytorna med helträdsuttag växer också i kapp, trots att de i början växer något långsammare än träden planterade på ytor med bara stamskörd. Dessa resultat stämmer med resultaten från långliggande försök i Sverige, men skiljer sig från tidigare modelleringar med enklare dynamiska modeller. Trots att skillnaderna minskar i markvattnet och i trädtillväxten, kan det ändå på sikt finnas risk för brist på baskatjoner i helträdsskördade skogar, eftersom mycket baskatjoner har förts bort vid helträdsskörden och det inte kompenseras av ökande vittring.

I den fjärde artikeln används modellen PROFILE på 26 ytor, spridda i Sverige. De modellerade mängderna baskatjoner från vittringen jämförs med mängderna som tas ut med biomassa vid helträdsskörd. Markens försurningsstatus i ytan räknas också in. Denna modellering kan användas för att beräkna hur stort skördeuttag som är uthålligt, ur närings- och försurningssynpunkt. Resultaten visar att med några få undantag är ytor i södra Sverige mest försurade, samtidigt som de också har högst upptag av baskatjoner till träden och högst uttag vid helträdsskörd och därför är mest känsliga för uttag. Vittringen är däremot mer jämn över landet och räcker oftast inte för att få ett hållbart uttag på en högproduktiv yta i södra Sverige.

PROFILE och ForSAFE beräknar ungefär lika stor vittring och kan båda användas för att undersöka skogsbrukets uthållighet när det gäller baskatjoner och försurning. Skillnader i indatabehov och dynamik gör att de har lite olika användningsområden. PROFILE går att använda på ytor där man inte har lika mycket indata och kan med fördel användas i beräkningar med god geografisk täckning, där ingen tidsupplösning behövs, till exempel beräkningar av kritisk belastning av försurande ämnen eller, som i denna studie, kritiskt biomassauttag. ForSAFE ger mer detaljerade resultat, som kan användas för att öka processförståelsen och dynamiken i klimatförändringens effekter på vittring, markens innehåll av baskatjoner och hur det svarar mot dynamiken i trädens näringsupptag.

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1 Introduction

1.1 Weathering and base cation cycling in different scales

Weathering is when mineral particles lose mass and/or have their chemical structure altered. Physical weathering is when rocks are fragmented into smaller pieces by physical forces, that can be further altered by chemical weathering. Chemical weathering leads to chemical altering or dissolution of minerals and release of their constituents and is therefore of great importance in many biogeochemical cycles, including that of carbon. Weathering consumes and binds carbon dioxide in secondary minerals, and the amount of weathering on continents and in oceans, together with the amount of carbon emitted in tectonic processes, are regulating the long-term equilibrium level of carbon dioxide in the atmosphere. Thus, weathering is of great importance for the climate of the earth (Kump et al., 2000; van der Meer et al., 2014).

Plant nutrients such as potassium, calcium, magnesium and sodium, sometimes called base cations, are released through weathering processes. This is very important for ecosystem functioning, since these nutrients are easily leached and lost from the land ecosystems. Although they are seldom the limiting nutrients for growth, they are necessary plant nutrients and vital to counteract acidification of soils and waters (Brantley et al., 2008).

Chemical weathering happens when minerals in bedrock, rocks and soil at the surface are exposed to air, water, carbon dioxide and acids. It is highly temperature dependent, and occurs mostly on the surface of the minerals. This means that the total exposed area is of great importance for the weathering, and that a fine-grained material, with more total exposed area, has a much higher weathering rate than a coarse material (Tarbuck & Lutgens, 1993).

During the last three million years, the global climate has become cooler, with permanent glaciers in Antarctica and Greenland and recurring glaciers in Northern Europe and North America. The ice sheets have gradually removed the sedimentary rock covers and the sediments and deposited them outside of the glaciated areas (Willeit et al. 2019). Scandinavia and other regions that once were covered by inland ice, have in many regions granitic and gneissic bedrock at the surface, with a very shallow soil layer, consisting mostly of primary minerals from the bedrock. Sedimentary bedrock has mostly been eroded away and covers only very small areas. Soil particles from such bedrock are uncommon. Parts of these regions have a so called cnoc-and-lochan landscape, with an irregular bedrock surface with many lakes and ridges. According to Krabbendam & Bradwell (2014), this landscape has been formed by deep chemical weathering in fractures and faults in granitic bedrock, during a time when the region was situated in a warm and moist climate with much higher weathering rates than in today's temperate or boreal climate. This earlier weathering produced deep and wide zones around the faults of rocks, filled with rock altered by weathering, called saprolite. The saprolite was later eroded by ice and water during and between the ca 50 ice ages, producing the irregular cnoc-and-lochan surface.

Variation in weathering in Scandinavia during Holocene

In Scandinavia and other parts of the continents that were covered by ice during the latest glacial period, the soils are young and less weathered than in many other areas. The soils consist mostly of till deposited by the latest ice sheet, or sedimentary soils deposited at the melting, or in an underwater setting during the Holocene. In some locations there are soils from older glaciations or interglacials beneath the newer soils. In places in northernmost Scandinavia, where the glacial ice was frozen to the ground and not as abrasive, surface soils from older glaciations still exist (Lagerbäck, 1988).

Thus, most soils in Scandinavia have only been exposed to soil forming processes and weathering during the Holocene, which is a short time for geological processes, especially in a cool climate. Signs of chemical weathering are often only seen in the uppermost decimetres of soil (Snäll & Ek, 2000).

Newly deposited soils, deposited by ice sheets or water, should have a higher weathering rate than older soils, since the surfaces of the soil particles are newer and not as covered by organic materials, secondary minerals formed by chemical weathering and oxides. This was confirmed by Starr & Lindroos (2006), in a study of weathering rates in a profile of sedimentary soils in southern Finland, successively raised above sea level during a period of 5000 years. Weathering rates were calculated using the depletion method and were shown to be higher the more recently the soils were lifted above sea level. The youngest soil, about 300 years old, had weathering rates for calcium and magnesium of 5 and 2.5 times that of the oldest soil.

This does not mean that the weathering rates have always decreased with time since the ice age, since weathering rates also depend on temperature, soil moisture and soil water chemistry. Soil moisture and chemistry in turn depend on e.g. vegetation and human activities. During the Holocene, temperature and precipitation have varied, vegetation has changed a lot, and human activities in the form of e.g. slash burning, agriculture and harvesting have had a large influence on soil chemistry (Ford, 1990; Warfvinge et al., 1995).

Variation in weathering rates at landscape level

The mineralogy of the bedrock, as well as the mineralogy of the soil cover, varies geographically. The variation in the soil does not fully follow that of the bedrock, since the ice sheets and water have moved soil particles and rocks of all sizes, sometimes over vast distances (Snäll & Ek, 2000; Akselsson et al., 2004; Górska-Zabielska & Wachecka-Kotkowska, 2014).

Akselsson et al. (2004) used the weathering model PROFILE on more than 25 000 sites with measured soil geochemistry in Sweden. The authors found that the weathering varies from site to site, that there existed regions with higher or lower weathering, but that these regions were not connected to regions of different bedrock mineralogy. An exception was the few regions in Sweden that has calcite rich bedrock, around which there were regions with calcite containing soils of higher weathering. Thus, the more well chartered bedrock mineralogy cannot be used to estimate weathering where soil mineralogy is unknown.

Snäll & Ek (2000) investigated geographical variation in mineralogy and weathering rates on 13 sites, of which six were located in the mineralogically very diverse region of Dalarna. Texture and total chemistry of the mineral soil were analysed for several soil depths. Mineralogy of the soil samples was analysed using the X-ray diffraction method and using mineral stoichiometry and total chemistry. Weathering rates were calculated using the PROFILE model. The authors found that part of the mineral mass was derived from the bedrock at the site, especially for the larger grain sizes, but a large part had been transported from areas with other bedrock. Some of the transported particles had been transported far. The C horizons had higher levels of silica and quartz than the bedrock had, which indicates that more of the easily weatherable, less silica rich minerals had been weathered away before the soil was deposited, or sorted and deposited elsewhere. This depletion of base cation richer minerals should have occurred before the till was deposited, as the C horizon is the bottom soil of uniform regolith, with no weathering gradients or horizons. The silica concentrations in the A horizon was even higher. The mineralogy differed between particle sizes, with more of the easily weatherable minerals in the silt fraction compared to the coarser fractions. The clay sized particles consisted of primary minerals, clay minerals, and iron and aluminium oxides. In the clay fraction there were clay minerals that could have been formed by weathering during the Holocene (e.g. vermiculite), and sometimes minerals that have been formed by weathering in a much earlier, warmer climate (e.g. kaolinite and gibbsite). In two of the southern sites of the study, there were clay minerals that suggest Holocene weathering in the C horizon.

Weathering depends strongly on temperature (see below) and temperature is, on average, much higher in southern than in northern Sweden. Despite this, there is no strong weathering gradient from south to north (Olsson et al., 1993; Akselsson et al., 2004).

Variation in weathering rates on a catchment level

A catchment is a geographical area where all runoff from precipitation in the area, for topographical reasons, discharge in the same point or points.

Landscape sized catchments can have large variations in mineralogy, climate, land use, deposition of nutrients and pollutants within them, and there can therefore be large differences in weathering rates in different parts of the catchments. Chemistry at the outlet cannot be easily tied to processes in the catchment. Small and well-defined catchments with uniform mineralogy and land use, such as the catchments around lake Gårdsjön in Sweden (Moldan et al., 2012a) can, on the other hand, be ideal for biogeochemical research, e.g. calculation of weathering rates. Catchments usually have one outlet that collects runoff from the whole catchment, where measurements of runoff chemistry that are representative of the whole catchment can be conducted. It can be relatively easy to measure in- and outflows over the catchment boundaries, uptake in vegetation and release through litterfall and decomposition of relevant chemical species with reasonable accuracy (Olsson et al. 1985). If measurements are conducted long enough so that changes in internal pools of base cations are small in comparison to the other flows, average weathering in the catchment can be calculated. The calculated weathering is the total weathering of all soil the water in the outlet has come in contact with, including soil layers beneath the root zone of the vegetation (Velbel & Price, 2007).

Weathering varies with topography within smaller catchments and hillsides as well. Yoo et al. (2009) describe a hillside in California, where the upper part shows physical erosion and the lower part receives deposited soil from the upper. The chemical weathering in the upper part was 3.33 times higher than in the lower. Weathering was low in both parts, as the soil was old and the mineral grains were covered with oxides and secondary minerals. The soil in the lower part was more nutrient rich, with phosphorus and calcium enrichment, compared to the soil higher up.

When no visible soil erosion exists, weathering still varies with topography, since the soil moisture is higher in the lower parts of the topography than in the higher. The higher soil moisture over time leads to higher soil carbon, that can store more nutrients, leading to different vegetation and growth rates. All of these factors influence the weathering rates (Erlandsson et al., 2016).

Starr et al. (2014) investigated the weathering rates in ten soil profiles in podzolic soil in a small catchment, using the depletion method, and found no significant differences in weathering, despite differing origins of the soils in the catchment, where three profiles were situated on sediment and seven on till.

Weathering in lab experiments

In the lab, as opposed to in the field, experiments can be designed to actually measure weathering rates, for example by measuring the change with time in concentration of weathering products. Several studies have shown that weathering measured in lab experiments, often with clean and newly ground mineral grains, are several orders of magnitude larger than calculated or modelled weathering rates in the field. White & Brantley (2003) showed that the weathering rate of a sample of plagioclase decreases exponentially and that it would take thousands of years of decreasing weathering rate to reach the weathering rates usually calculated for plagioclase in the field. The difference in weathering rates between lab experiments and in the field have two causes, according to the authors. Firstly, the mineral surface changes over time, with it being successively covered by weathering products, oxides and organic molecules. Secondly, the conditions in the field is less optimal for weathering, with lower permeability of the soil, lower soil moisture and higher concentrations of weathering products in the soil solution.

1.2 Effects of separate environmental factors on weathering

Weathering rates have been of interest since the late 19th century. Already then it was known that the weathering is strongly affected by temperature and moisture and that dissolved carbon dioxide and organic and strong inorganic acids also has an influence on the weathering rates (Merrill, 1896). Merrill measured weathering rates in the lab and showed that different minerals have different weathering rates under the same conditions, with chalk having the fastest rate and magnetite the slowest of the tested minerals. He also made calculations of weathering rates in the field, using mass balance: 143.5 resp. 275 tons of mineral mile^{-2} year^{-1} (in the Appalachians resp. England and Wales).

Today, the chemical reactions of the weathering are relatively well known and formulated (Brantley et al., 2008). The main weathering reactions are with water, carbon dioxide, dissolved organic molecules (DOC), hydrogen ions, and, in basic environments, hydroxide ions. Temperature, moisture, mineralogy and exposed mineral surface have large influences on the reactions.

Temperature

Higher temperature speeds up weathering exponentially. Weathering rates of different minerals have different sensitivities to temperature (White & Brantley, 1995). For any one mineral, the different weathering reactions (with water, carbon dioxide, hydrogen ions, hydroxide ions and organic acids) also have different sensitivity to temperature. The dependence of temperature on the weathering rate can be described using Arrhenius equation:

$$k = \mathrm{A} * 10^{-E_a/RT} \quad (1)$$

where E_a is the apparent activation energy (which in its turn can depend on soil water pH), R is the molar gas constant and T is temperature in Kelvin (White & Brantley, 1995).

According to Warfvinge & Sverdrup (1995), the weathering rates for the different chemical reactions often are most similar at 8°C, and increasingly different at lower and higher temperatures. The reactions with water and hydrogen ions are the most temperature dependant, which means that they are the largest at warm temperatures, and lowest at temperatures lower than around 8 °C. This means that in the Swedish climate (where yearly average temperature ranges from -2 °C to 8 °C), the reaction rates of all the reactions are of similar size, whereas in hot humid climates, the weathering rates of the reactions with water and hydrogen ions are the largest. Which of the reactions that dominate also depend on other factors, like availability of hydrogen and hydroxide ions, carbon dioxide and organic acids.

Different minerals have different temperature dependency for the different reactions, but the average weathering rates for the different reactions often double when temperature increase by 10°C. The minerals calcite and dolomite differ from most other, as their reactions with hydrogen ions have a very weak temperature dependency.

Soil moisture

The weathering reaction with water is one of the most important weathering paths (Sverdrup & Warfwinge, 1993), and water is also needed as a transport medium for reactants and products of all the weathering reactions. Thus, the amount of soil water that can act as a transporter of dissolved chemical species to and from exposed mineral surfaces has a large impact on total weathering in the soil. Too little soil water means that some of the mineral grains, with their surface film of adhesive water, will be isolated from the main volume of soil water. The increasing concentration of weathering products and diminishing concentrations of reactants (other than the mineral itself) in the isolated water, means that the weathering will come to a halt. Too much water, on the other hand, can lead to anoxic conditions, with lower movement of soil water. Roots, which normally take up the weathering products potassium, phosphorus, calcium and magnesium, and thus stimulate weathering, need oxygen and do not normally grow in anoxic conditions (Tamm et al., 1974).

The effect of climate change on precipitation is less well known than the effect on temperature, and the effect on soil moisture is even more uncertain (Rousteenoja et al., 2017). In Sweden, especially in southern Sweden, many models predict less precipitation during summer than today, or at least a lower percentage of yearly precipitation during summer (Toreti et al., 2019). Together with higher temperatures, this means that soils might be drier during summers on average, which would counteract the effect of the warmer temperatures on weathering (Belyazid et al., 2022).

Texture, exposed mineral surface and mineralogy

Chemical weathering takes place on the surface of the mineral particle, in the thin film of soil moisture that adheres to it. The surfaces are not smooth, nor homogeneous with respect to mineral composition, and there are hot spots for weathering on the surface where more easily weathered minerals or even more easily weatherable parts of the mineral molecules are exposed (Brantley et al., 2008).

This variability of weathering rates at the micro scale can be large, but when weathering is calculated in all larger scales than atom by atom viewed by an electron microscope, the weathering rates are averaged for the whole exposed surface of each mineral. The total exposed area of the mineral grains can be measured or approximated from the texture, for example from the amount of clay, silt and sand particles in a soil sample. The relationship between total exposed surface and texture is different for different soils and mineralogies. Thus, the equation used should be tested for the region it is used in (Whitfield & Reid, 2013).

A certain mass of mineral has a larger exposed area the finer the texture is. This means that fine grained soil has larger weathering rates than coarse textured soil, if everything else is equal (White & Brantley, 1995).

Sverdrup et al. (2009) described a study of weathering along a transect with the same mineralogy, but successively finer texture, i.e. successively larger exposed mineral surface. The study showed that weathering rates are linearly dependent on the total exposed surface, if the mineralogy is the same. Holmqvist et al. (2003) studied the potassium balance in Scottish agricultural soils and saw the same linear relationship between exposed surface and potassium weathering.

There are only a limited number of minerals, but the composition of each can vary within certain limits. Different minerals have very different weathering rates, if everything else is equal, and their weathering rates react to various environmental factors differently. A few are so easily weatherable that a large part of the total weathering of an element from a soil sample comes from these minerals, even when they only are present as a small fraction. Knowledge of the mineralogy is very important when using process-based models to calculate weathering. Casetou-Gustafson et al. (2018 and 2019) used two different methods for calculation of mineralogy of soil samples. The first method, normative mineralogy, consists of analysis of the total amount of all common elements in the sample, which, together with the estimated composition of each mineral, is used to calculate all possible mineralogical compositions of the soil sample (given it is known which minerals are present in the sample) (Posch & Kurz, 2007). The second method was analysis of the mineral grains using x-ray powder diffraction (Casetou et al., 2018). The normative method gives many theoretically possible mineralogies, all having the exact same elemental composition, without giving any information on which of them is more likely. The x-ray powder diffraction method on the other hand, does not detect minerals present in smaller fractions than a percent of the total mass. For some minerals such a small fraction can mean much for the total weathering rate.

Texture and mineralogy at a certain site are usually not affected much by climate change in the short term, but on longer time scales, changing winds and changing occurrence of bare soils can move the finest fractions in large quantities and over long distances. The large areas of deep loess soils in Europe and China were formed when silt was transported by wind from bare, dry, cold areas close to the ice sheets and in inland Asia during the latest glacial period, forming deep soils of sorted silt (Tarbuck & Lutgens, 1993). Today, in Scandinavia, most of the bare soils are agricultural soils, many of which have a fine texture with a larger amount of the small fractions, compared to most forested areas (Fredén, 1994).

pH-value

The pH-value in the soil water affects activity and solubility of different chemical species, including CO_2, DOC and weathering products, not least aluminium. The pH-value therefore affects all the weathering reactions through availability of reactants, concentrations of inhibiting reaction products and through precipitation of covering salts and molecules on the mineral surfaces. According to White & Brantley (1995) pH also affects the activation energy for the weathering reactions and therefore the temperature dependency of the reactions.

The hydrogen ions also react with the minerals, in the weathering reaction that, for most minerals, has the largest rate at higher temperatures (and lowest rate at low temperatures). A decreasing pH-value thus leads to higher weathering, unless the low pH leads to too high concentrations of inhibiting weathering products such as aluminium (Warfvinge & Sverdrup, 1995).

Concentration of CO_2 in the atmosphere

An increasing concentration of CO_2 in the atmosphere act as a fertilizer to vegetation. If all else is equal this leads to increased growth, larger plant biomass and larger need for mineral nutrients. This can increase weathering rates, as inhibiting weathering products are removed continually from the soil solution (Rosenstock et al., 2019).

An increasing concentration of CO_2 in the atmosphere also increases acidity of the oceans, increases the solubility of calcium in sea water (Fox et al., 2020), which possibly could increase the amount of sea salt derived calcium deposited in coastal areas. In the long run, this could have an effect in calcium poor areas, where it might affect vegetation composition and soil acidity, amount of adsorbed and dissolved calcium in the soils, and thus the weathering of calcium.

The concentration of CO_2 in the soil depend on decomposition of organic material in the soil and not directly on the concentration of CO_2 in the atmosphere. The decomposition rate in the soil is affected by temperature, moisture, type of organic material, type of vegetation, nutrient status, and thus indirectly also on the concentration of CO_2 in the atmosphere (Aber & Federer, 1992).

Amount of organic material in the soil and DOC

The amount of organic carbon in the soil affect the concentration of DOC in soil water, pH and the hydraulic and water holding capacities of the soil, and therefore the weathering rates. The amount of organic carbon in the soil is in its turn affected by soil moisture, temperature, amount of nitrogen, type of vegetation, presence of herbivores and detritivores and disturbances, which means that it is very affected by climate and climate changes.

Changes in wind and precipitation in coastal areas can affect the amount of sea salt deposition. Increased sea salt deposition lead to decreased solubility of DOC (Moldan et al., 2012a), inhibiting weathering reactions with DOC. The base cations from the sea salt also act inhibiting on all weathering reactions. Sea salt episodes lead to temporarily lowered pH, which could lead to increased weathering, unless aluminium concentrations also increase due to low pH.

The reaction rate for weathering with DOC is the least, or second least (depending on mineral), temperature dependent reaction, which means that it is one of the largest weathering pathways at low temperatures (Sverdrup & Warfvinge, 1993).

Biology and land use

Different vegetation, other organisms and land use affect litter type, quantity and quality, soil chemistry, texture and other factors. Different vegetation uses different amounts of nutrients (Aber & Melillo, 1991). Weathering is affected by these factors and biology thus indirectly affects the weathering rates.

The different pH-values and litter properties favour some detritivores and other soil organisms over others. They in their turn affect the soil by mixing the organic and mineral soil at different depth and creating macro pores or not being able to do so, by preferring some organic fractions over others and by leaving a slowly decomposing pool of organic matter with a higher or lower nitrogen content. These behaviours affect soil pH, soil moisture and weathering.

In large parts of Sweden, the dominant tree species in forests are decided by humans, within the limitations of the climate. In most forests the dominant tree species is Norway spruce (*Picea abies*) or Scots pine (*Pinus sylvestris*). A changing climate can both affect the understory vegetation and make the chosen dominant tree species unfeasible at a forest stand (or a whole landscape), thus indirectly affecting soil biota, soil chemistry and weathering.

Removal of biomass at harvesting, especially branches, leaves or needles, removes base cations from the ecosystem. This can lead to lower concentrations of base cations in the soil water, which in theory and in process-based models can have the effect of enhancing weathering. Lower concentrations of base cations can also lead to soil acidification, which can lead to increased solubility and raised concentrations of aluminium (Olsson et al, 1993; Stendahl et al., 2013). This in turn can inhibit weathering (Kronnäs et al., 2019). Another effect of harvesting is increased soil moisture as the water uptake from the trees is halted. Higher soil moisture enhances weathering.

Herbivorous insects are affected by forestry and to a large extent by a changing climate. Kristensen et al. (2019) show that insects in turn have a great impact on the soil carbon and nitrogen metabolism and reduce the trees' supply of energy for mycorrhiza. This in turn affects soil water chemistry and thus weathering. In Scandinavia, many herbivorous insects are benefitted by a warmer climate, leading to larger numbers and expansion into new areas, which could also potentially have large effects on vegetation (Karlsson et al., 2018) and therefore on weathering in a future warmer climate.

Mineral grains sometimes have microscopic etched rhizome like patterns, which has been taken as a sign of weathering enhancing activity from the plants (via roots and mycorrhiza). This so-called biological weathering is the effect of organic substances released from plant roots, to enhance weathering in close proximity of the roots of plant nutrients specifically needed by the plants. The occurrence, functioning and size of this biological weathering has been discussed (Rosenstock, 2009; Sverdrup, 2009). Sverdrup (2009) argues that the abiotic processes already described and parametrised explains these patterns. Roots do exude organic acids (whether they are deficient of any specific mineral derived nutrient or not), and organic acids do react with minerals in one of the main weathering reactions, and they lower pH and enhance weathering with hydrogen ions. Rosenstock et al. (2016) conducted an experiment with mesh bags containing minerals in areas with deficiency of phosphorus, magnesium, or potassium. Trees deficient in phosphorus increased their exudates of carbohydrates for mycorrhiza where mesh bags containing weathered phosphorus rich minerals, which increased weathering of phosphorus. However, they did not react in this way when deficient in magnesium or potassium. Smits & Wallander (2017) investigated a gradient in naturally lead polluted soil, where neither mycorrhiza nor plants could survive at the most polluted end. The study showed that the weathering of phosphorus was at least as high in the vegetation free area, meaning that biological weathering did not have an effect (on phosphorus weathering) in this soil. Finlay et al. (2020) argues that the fine roots and mycorrhiza transports both organic carbon from trees to soil and weathering products from the soil, making the soil more connected, more homogenous in soil water concentrations. The weathering therefore becomes less inhibited by local high concentrations of weathering products. In lab experiments conducted by Rosenstock et al. (2019), soil with *Picea abies* plants weathered faster than soil with no plants, and soil with plants and mycorrhiza had a tendency of weathering even faster. The authors drew the conclusion that the effect on weathering that the plants and mycorrhiza had, was due to their uptake and removal of weathering products, that would otherwise inhibit weathering.

1.3 Methods and models for weathering calculations

During the end of the 19th century and the beginning of the 20th, methods of calculating the size of the weathering were of interest, as the importance of weathering as a supply of nutrients for productive forests was discovered (Merrill, 1896; Tamm, 1920). The first weathering calculations were mass balance calculations and calculations from lab experiments. Later, during the 1960s, wide spread acidification of lakes in parts of Scandinavia was discovered and understood to be an effect of long range acidifying atmospheric pollution, acidifying both soils and surface waters (Odén, 1968). Thus, methods for calculating weathering rates became subject to a renewed interest, as weathering rates were determining for how sensitive soils and surface waters were to acidification. In the 1980s, when computers became more common in research, geochemical models were developed to calculate the pathways of pollutants from emission, through the atmosphere, to land and lake ecosystems, where harmful effects from the pollution were quantified. These were used in international negotiations for abatement strategies (Sverdrup et al., 1992; Warfvinge & Sverdrup, 1995; Alveteg, 2004). The abatement measures were successful and emission of especially sulphur decreased dramatically. Despite these measures, ecosystem recovery was slow or non-existent. Due to this, modelling efforts were redirected from calculations of critical loads of acidification, to questions about how and when ecosystems can recover from acidification. This required more sophisticated dynamical models. Models and experiments demonstrated that some recovery could be seen within years from diminishing exposure of acidifying pollutants, but that full recovery could take many decades (Sverdrup et al., 2005; Moldan et al., 2012b).

During the latest decade, the deposition of acidifying pollution in Europe has become so low and the nitrogen enhanced forest growth so high, that the removal of nutrients via harvest, as an average over the whole rotation period, is of the same order of magnitude as the leaching of base cations caused by the acidifying deposition (Iwald et al., 2013). This means that the removal of nutrients with harvest, especially whole-tree harvest for bio fuels, can be a hindrance for recovery of soils and waters from acidification and even cause reacidification (Moldan et al., 2017), especially in productive *Picea abies* forests or plantations. Today, there is a renewed interest of increasing forest growth and harvest even more, as a means of substituting fossil fuels and materials (Rytter et al., 2016). Therefore, estimations of weathering rates with a higher resolution and certainty are important, while also taking the effects of climate change and different forest management strategies into account.

Mass balance methods

Mass balance methods use measurements of in- and outflows of elements into and out of a well-defined ecosystem (for example a catchment), to calculate the sources of the elements in the ecosystem. Weathering is calculated as the inflows (deposition) minus the outflows (leaching and net uptake in vegetation) plus/minus changes in internal temporary stores of the element (Velbel & Price, 2007). Temporary stores in the soils are base cations incorporated in organic materials and base cations adsorbed on mineral surfaces, which are usually negatively charged and therefor attract cations. Sometimes the ecosystem is assumed to be in steady state, which means that the size of the stores is not changing with time (or changing with season only, so that they are the same at the same time of the year for different years) and only measurements of deposition, leaching and net uptake to vegetation are needed.

In mass balance calculations, weathering rates are calculated as the difference between other, often larger, measured flows. Uncertainties in measurements can add up and cause larger uncertainties in the calculated weathering rates. Unless the soil depth is very small (as in Gårdsjön, Olsson et al., 1985), it can be difficult to assess how large soil volume is actively involved in producing base cations for the outlet stream, as the flows and weathering rates gradually slows down with depth.

Another uncertainty is that under some circumstances the shape and size of some catchments can temporarily change at high water levels or ground water levels. This can mean that adjacent areas become part of the catchment, or that temporary discharge points are created. In these instances, the catchment soil volume is not easily defined, nor constant, and the measured discharge is not necessarily representative of the whole catchment.

The depletion method

The depletion method, also known by other names (e.g. historical weathering, or the zirconium method, Casetou-Gustafson et al., 2020), is a weathering calculation method that compares the ratios of easily weatherable minerals or easily leachable elements and very slowly weathering minerals or very immobile elements, throughout a soil profile. A deep soil layer from the C horizon is relatively unaffected by weathering and has little enrichments of leached organic materials or elements from above (Tarbuck & Lutgens, 1993). The ratios in this soil layer should be close to the original ratios of the whole soil profile as it was at soil formation, assuming the soil was uniform from the beginning. The depletion method works best for relatively young soils, were the conservative mineral or element has not had time to weather or move much, such as many Scandinavian soils, formed at the end of the latest glacial period. The depletion method calculates how much of the more easily weathered minerals have been lost from the shallow soil layers since soil formation. It uses the assumption that all of the very slowly weathering mineral or immobile element is still there in the soil layer, in the same amounts as at soil formation, and that different ratios between the easily weatherable mineral and the slowly weathering mineral, in these layers above the C horizon, exist because part of the more easily weatherable minerals have weathered and leached away. An average weathering rate since the soil formation can be calculated if the age of the soil is known.

Quartz is sometimes used as reference mineral, since it weathers much slower than many other minerals (Starr & Lindroos, 2006). Zirconium is often used, since the element is rather immobile in the soils (Stendahl et al., 2013).

Ideally, the soil profile used should be deep enough to show a clear downward convergence to some ratio in the deep C horizon. If the ratio changes abruptly at depth, this indicates that the soil was not deposited uniformly at the same time period, and the method is not fitting.

Weathering generally decrease with time in a soil and the weathering during the beginning of this period should have been much higher than later, as the mineral grains were clean and had more unweathered surfaces with exposed easily weatherable minerals. The beginning was, however, very short in comparison to the rest of the time period up to today, and has not much influence on the average weathering. Therefore, weathering calculated using the depletion method can be higher or lower than weathering today. Land et al. (1999) for example, show twice as high weathering rates of base cations today than the average rates since the glaciation, in the Kalix river in northern Sweden.

Process-based models

There are several process-based models that can be used for calculating weathering rates, such as PROFILE (Sverdrup & Warfvinge, 1988; Sverdrup & Warfvinge, 1993), SAFE (Alveteg et al., 1995), ForSAFE (Wallman et al., 2005; Belyazid et al., 2006), MAGIC (Cosby et al., 2001), WITCH (Goddéris et al., 2006), HD-Minteq (Gustafsson et al., 2018; McGivney et al., 2019), Crunchflow (Maher et al., 2009) and GEM-CO_2 (Amiotte-Suchet et al., 2003). These models are based on flows of common anions and cations into and out of the modelled site, and chemical reactions, uptake to vegetation, litterfall and decomposition within the site. The foci of the models, the detailing of the reactions and the time scales are different between models. MAGIC, for example, focuses on chemical properties of the surface water in a small catchment, whereas PROFILE focuses on the weathering reactions and critical loads of acidity in a single soil profile in a theoretical steady state. SAFE is a dynamic development of PROFILE, that take the time dimension into account, and ForSAFE is a still more developed version, with sub models for tree growth, decomposition and hydrology. HD-MINTEQ is another dynamic development of PROFILE, using a more advanced description of aluminium complexes and their effects on weathering. WITCH is also a dynamic geochemical model, focusing on the formation of secondary minerals, which the other biogeochemical models usually do not include. GEM-CO_2 focus on the effects of the CO_2-consumption of weathering on the global climate.

Comparison of different methods

Different methods of weathering calculations use different system boundaries and the weathering represent different soil depths and time periods. Mass balance methods using catchments give weathering for the whole soil depth that supplies water to the outlet runoff, which means that less weathered soil at depths below the root zone of the trees might have an influence. Weathering rates calculated with these methods are valid for the time when measurements of in- and outflows were taken. Process-based models usually model a soil profile down to the soil depth relevant for the trees at the site (often 50 cm, where the majority of the roots are situated, Tamm, 1920; Rosengren & Stjernqvist, 2004) and a time period of up to around a century back and forward in time from today. The depletion method uses a soil profile and calculates the average weathering from the time of soil formation up to today. This is a long time period of variable conditions, especially at the beginning, with clean newly crushed mineral grains, and at the end, with fast changes in deposition of acidifying pollution and base cations, land use and climate. The resulting weathering rates from different methods are therefore not readily comparable and can differ for the same site even without any of them being wrong.

Futter et al. (2012) compiled weathering estimations from several methods and sites, from three continents. The methods usually ranked sites in the same order going from low to high weathering. However, the different methods differed in the absolute weathering levels, primarily depending on uncertainties in input data. Mass balance methods usually differed most from the other methods. The authors concluded that if weathering calculations are important to decide management on some specific site, at least three different methods should be used on the site to get a good estimate on weathering.

Despite the fact that weathering rates from the depletion method are averages over very long time spans, the rates from this method is often comparable in size with present weathering rates calculated using other methods (Futter et al., 2012; Stendahl et al. 2013). According to Stendahl et al. (2013), this is at least partly because the present Scandinavian soils never contained much easily weathered minerals and thus the weathering has not decreased as much through the ages as if they would have contained much easily weathered minerals at the beginning. Also, weathering rates today might be raised by increased intensity of forestry, acidification and climate change, compared to earlier times.

Stendahl et al. (2013) compared weathering rates from the depletion method and from PROFILE for 16 sites across Sweden and found that the rates were usually of similar size, but that both methods had one or two sites where the method seemed to fail. For PROFILE, one site had very compact fine-grained soil, that PROFILE modelled as having high weathering rate (as opposed to the depletion method), even though the weathering most likely was inhibited by low conductivity. PROFILE does not model hydrology, as opposed to the dynamical developments SAFE and ForSAFE. The dynamical models might have been more successful with the site. The depletion method had problems with two sites which might have had soil layers of different origin and mineralogy. Except for these three sites, correlation between weathering rates from the two methods were good for calcium and magnesium weathering, but low for potassium and sodium weathering. This might suggest that weathering rates of potassium and sodium are more affected by contemporary environmental factors, whereas weathering of calcium and magnesium is more affected by more constant factors such as presence or absence of certain minerals.

Akselsson et al. (2019) compared calculated weathering rates for sites where at least two methods had been used and where the same soil depth was used. For any one site, the difference between methods could be big, but the methods were usually in agreement about which sites had high and which sites had low weathering. The span of weathering rates for all sites were also similar. Weathering rates calculated using the mass balance method were the most dissimilar from the other methods and historical rates from the depletion method were often about half as large as PROFILE values.

Casetou-Gustafson et al. (2020) compared the depletion method, PROFILE and the mass balance method on two sites, and found, as above, that the methods agreed on the which site had the highest weathering, but differed in absolute numbers, with the mass balance method giving the most dissimilar rates.

1.4 Process-based modelling of weathering and base cation cycling in a changing climate

Weathering rates vary with temperature, soil moisture and soil water chemistry, and are thus affected by climate change, atmospheric deposition and different types of forest management. Today climate is changing, and in large parts of Sweden soils have been acidified and many are slowly recovering. Base cation concentrations have become low, compared to earlier periods in many soils (Pihl Karlsson et al., 2011). At the same time forests are growing faster than in earlier times because of increased nitrogen deposition and changed management practices, and there is a societal pressure to increase growth and harvest even more (Rytter et al., 2016).

Thus, the details of the dynamics of the base cation cycle in the soils are more important than earlier. The seasonal dynamic of base cation uptake and weathering might be important, as well as the effect on base cations of forest management.

Dynamic process-based models like ForSAFE are needed for studies of dynamics and details of the effects (Akselsson et al., 2016; Akselsson et al., 2019; Kronnäs et al. 2019). ForSAFE include the many feedbacks between weathering, soil water concentrations, vegetation uptake, soil carbon, decomposition, soil CO_2 concentration, soil acidification status, drought, nutrient removal through forest harvest, and more.

The simpler, but also process-based model PROFILE does not need time series of climate, deposition and forest management data, and can be used to study some long-term effects on weathering and acidification soil status of climate change or management choices. The advantage of this is that many more sites can be modelled, giving a more area covering view of the effects, albeit less detailed and dynamic.

2 Objective and research questions

The demand for timber and biofuel from forests is increasing, leading to an enhanced pressure on forest ecosystems. The on-going climate change is at the same time affecting processes in ecosystems, such as tree growth, decomposition and weathering. In order to assess long-term sustainability, it is important to be able to predict the combined effect of intensified forest management and climate change on nutrient cycles in forest ecosystems.

The main objective of this thesis is to increase knowledge about how weathering rates vary in Sweden, both spatially and temporally in a changing climate, and to assess what implications it has for base cation availability and acidification, by the use of process-based geochemical models.

The research questions in the thesis were:

- Does the dynamic biogeochemical ForSAFE model give similar weathering rates, as averages over time, as the simpler steady-state geochemical model PROFILE, and how does the different ways of handling hydrology affect the results? (Paper I)
- How does weathering vary within and between years according to ForSAFE, and how does it respond to different drivers (climate change, atmospheric deposition and forest management)? (Paper I)
- How will weathering rates react to future severe droughts, on top of the climate change, in different climate regions in Sweden, and what are the potential implications for nutrient availability? (Paper II)
- How does whole-tree harvesting, here defined as harvesting of stems and branches, affect weathering and base cation concentrations in soil solution in different climate regions in Sweden? (Paper III)
- How does the risk of negative effects after whole-tree harvesting vary geographically over Sweden, and what is the role of weathering? (Paper IV)

3 Methods

Two process-based models were used to meet the objective and answer the research questions: the steady-state model PROFILE (paper I and IV) and the dynamic model ForSAFE (paper I, II and III). All model simulations were performed on sites in the SWETHRO network (see below), where high quality input data as well as validation data is available. In all studies, the A2M model (Posch & Kurz, 2007) was used to estimate mineralogy from total elemental content, which is required as input in PROFILE and ForSAFE.

In paper I, weathering rates derived from PROFILE and ForSAFE were compared at two sites and the temporal variation in the ForSAFE derived weathering rates was investigated. In paper II, effects on weathering rates of climate change in general, and from droughts in particular, were studied by the use of ForSAFE at seven sites. In paper III and IV the potential effects of the base cation losses related to whole-tree harvesting were studied, and the role of weathering was investigated. In paper III, ForSAFE was used on six sites to study the temporal development of base cations in soil solution after whole-tree harvesting. In paper IV weathering rates were calculated with PROFILE on 26 sites, and used to estimate critical biomass harvesting, which were combined with soil water chemistry measurements in a risk classification.

The sites used for simulations, the models and the input data required for the models are described below.

3.1 The SWETHRO Monitoring Network, study sites

The sites in all the studies in this thesis (Table 1, Figure 1) are sites from the SWETHRO Monitoring Network (Pihl Karlsson et al., 2011). This program was started in the mid-1980s, with the aim of investigating effects of air pollution on productive forests in Sweden. Today it consists of a bit more than 60 sites in mature even-aged forest stands, mostly of *Picea abies* or *Pinus sylvestris*, and mostly in productive managed forest. Atmospheric deposition and soil water are sampled in 30 m by 30 m squares, and analysed chemically. Given that the sites usually are situated in managed forest, measurements are terminated after clear-cutting, and the site is usually replaced by a nearby site in similar but younger forest. Sometimes measurements of soil water chemistry have been carried on a couple of years after clear-cutting, to investigate the effect of clear-cutting on soil water chemistry. In total, over 350 sites have been part of the SWETHRO network since it started.

At all sites, soil water chemistry is measured using lysimeters at 50 cm depth, which are sampled three times a year: before growing season, during growing season and after growing season. The sampling dates differ for different sites and varies slightly from year to year for practical reasons. The soil water samples from different lysimeters at one site and measurement occasion are merged to one composite sample and pH and alkalinity are measured, as well as concentrations of dissolved sulphate, chloride, nitrate, ammonia, calcium, magnesium, sodium, potassium, manganese, iron, inorganic as well as organic aluminium, and dissolved organic carbon.

Throughfall precipitation and deposition of hydrogen ions, sulphate, nitrate, ammonia, chloride, calcium, magnesium, sodium, potassium and manganese are measured once a month. Throughfall deposition of substances with relatively low biological interaction with trees, i.e. chloride, sodium and sulphate, are used as a measure of the total (wet + dry) deposition to forests. At many sites the same measurements are made on a nearby open field, without the interference of the canopy. From the open field measurements, wet deposition can be assessed. At a couple of sites there are also measurements of dry deposition of the nitrogen species and the base cations calcium, magnesium and potassium (Karlsson et al., 2019), which can be used, together with wet deposition, to estimate the total deposition of those substances.

All sites included in this thesis were part of a measurement campaign, starting in 2010, where measurements of tree height and diameter for estimation of biomass were made. Soil samples were taken from 4-5 soil layers, down to about 50 cm soil depth, for analysis of grain size distribution, density, exchangeable base cations, carbon and nitrogen content, and total chemistry for mineralogy estimation. Stoniness and soil moisture class were visually assessed in these places. Some of the sites are also forest sites included in the Level II of ICP forests, which is part of the International Co-operative Programme on Assessment and Monitoring of Air Pollution Effects on Forests operating under the UNECE Convention on Long-range Transboundary Air Pollution, and for those there are additional tree measurements, soil chemistry, needle chemistry and defoliation data.

The sites are in productive, usually planted, 50-100 years old *Picea abies* forest. There are more sites in southern Sweden, where air pollution and acidification have been higher and where also generally biomass productivity is higher, and where fluxes of base cations are larger. Northern Sweden is also represented by a couple of sites.

Table 1. SWETHRO sites
SWETHRO sites used in the studies, which model have been used and in which of the papers.

ID	Name	Latitude	Longitude	Model	Paper
BD06A	Grankölen	66.06	21.47	PROFILE	IV
AC34A	Ammarnäs	65.95	16.31	PROFILE, ForSAFE daily	II, IV
AC04A	Högbränna	65.41	18.10	PROFILE, ForSAFE daily	II, III, IV
AC35B	Holmsvattnet new	64.54	21.09	PROFILE	IV
AC35A	Holmsvattnet	64.54	21.05	ForSAFE daily	II, III
Y07A	Storulvsjön	62.28	16.34	PROFILE	IV
U06A	Hyttskogen	59.94	16.53	PROFILE, ForSAFE daily	II, III, IV
S22A	Blåbärskullen	59.82	12.91	PROFILE	IV
S05A	Södra Averstad	59.01	13.11	PROFILE, ForSAFE daily	II, III, IV
R09A	Stora Ek	58.63	13.78	PROFILE	IV
O35A	Hensbacka	58.44	11.74	PROFILE	IV
E21A	Solltorp	58.15	15.43	PROFILE	IV
P95A	Storskogen	57.86	12.67	PROFILE	IV
F22A	Bordsjö	57.84	14.99	PROFILE, ForSAFE daily	II, III, IV
F12A	Värnvik	57.83	14.39	PROFILE	IV
F23A	Fagerhult	57.51	15.34	PROFILE	IV
F18A	Mellby	57.14	13.59	PROFILE	IV
G22A	Tagel	57.06	14.38	PROFILE	IV
N12A	Borgared	56.95	12.81	PROFILE	IV
H03B	Rockneby	56.86	16.32	PROFILE	IV
N13B	Timrilt	56.77	13.16	PROFILE	IV
H22A	Alsjö	56.64	15.62	PROFILE	IV
N19A	Kullahus	56.36	13.00	PROFILE	IV
K13A	Vång	56.27	15.44	PROFILE	IV
L18A	Hissmossa	56.18	13.51	PROFILE, ForSAFE monthly	I, IV
L07A	Västra Torup	56.14	13.51	PROFILE, ForSAFE monthly & daily	I, II, III
L15A	Maryd	55.62	14.09	PROFILE	IV
M16A	Stenshult	55.54	13.57	PROFILE	IV

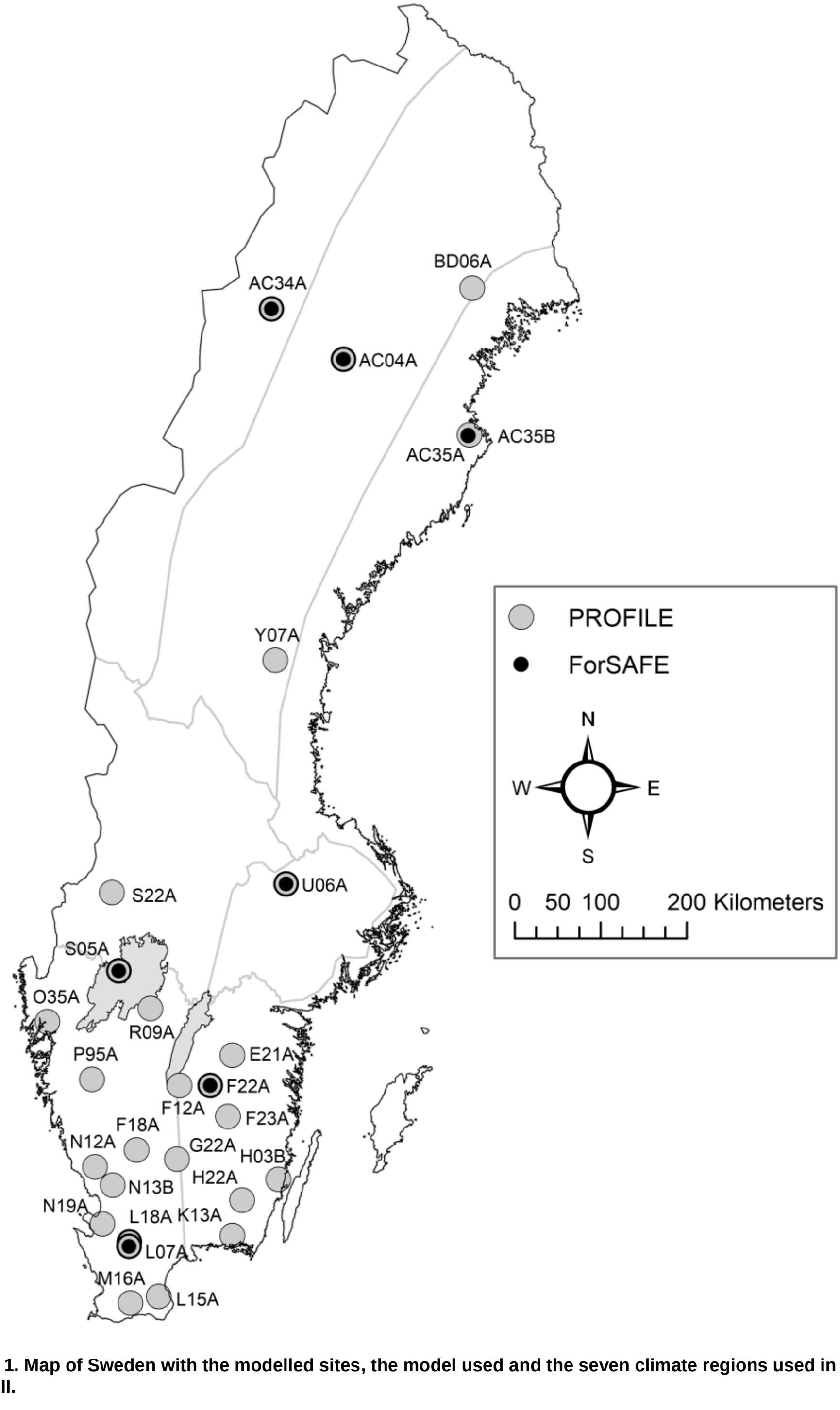

Figure 1. Map of Sweden with the modelled sites, the model used and the seven climate regions used in paper II and III.

3.2 The models

A2M

A2M is a mathematical model for calculation of possible soil mineralogy, based on soil data. Input data required are a list of possible minerals in the soil, chemical composition of those minerals and the total chemistry (an analysis of the elemental composition of all solid material of the soil after drying and combusting the sample) of the soil sample. The model calculates all possible mineralogical compositions given the input data (Posch & Kurz, 2007), for use as input data in geochemical models. It solves an equation of matrices and gives a matrix of all possible combinations of minerals in the sample. It cannot estimate which of all calculated possible mineralogies that is more likely to exist at a site, nor give a set of solutions where one or more of the input minerals is not present at all. Unless other information on which of the mathematical solutions from the model is more likely, and unless the whole range of possible mineralogies can be used, the average solution (which is in itself a possible solution) can be used as the mineralogy of the sample in further modelling. In these studies, the average mineralogy was used, except for a test using the PROFILE model (not published), where all solutions from A2M were used in separate model runs.

There are often solid or semi solid weathering products present in soils, consisting of aluminium or silica compounds (Brady & Weil, 1999). The aluminium and silica in these solids will be included in the total chemistry analysis. If no mineral consisting of aluminium without base cations is given in the input data to A2M, A2M will either not find a solution, or place the aluminium in silicate minerals together with base cations. This overestimates the amount of aluminium rich silicate minerals and underestimates the amount of base cation richer, and often more easily weatherable, minerals. Therefore, even if there usually are no fully mineralised aluminium oxide, such as gibbsite, in the Swedish soils (Snäll & Ek, 2000), the minerals gibbsite (aluminium oxide) or allophane (aluminium silicate) should be given as input data to A2M, to represent these not fully mineralised solids. In these studies, “gibbsite”, i.e. a solid consisting of aluminium, but no silica, was used as input data to A2M.

The uppermost layer from the SWETHRO sites is usually a mostly organic layer with just a small fraction of mineral soil. Total chemistry analysis has been conducted also on these layers, but the results are not reliable for mineralogy calculations by A2M, since the ashes of the organic materials are a big part of the total chemistry for those layers. Therefore, in these studies we have been using the mineralogy of the second layer also for the uppermost layer.

PROFILE

The PROFILE geochemical model (Sverdrup & Warfvinge, 1988; Sverdrup & Warfvinge, 1993) was created for calculation of weathering rates, leaching of base cations from soils and critical loads of acidity. Critical load of acidity (Grennfelt et al., 2001) is a measure of how large acidifying deposition that a particular site could be exposed to, and still have an acceptable quality of the water leaving the root zone, from an acidification point of view, in a steady-state situation. PROFILE has been used for calculations of critical loads of acidification and weathering rates in Europe, North America and East Asia on forests, open fields and agricultural land (Akselsson et al., 2016; Erlandsson et al., 2016; Fumoto et al., 2001; Holmqvist et al., 2003; Phelan et al., 2014; Stendahl et al., 2013).

The ecosystem is in PROFILE represented by a soil profile with a discrete number of soil layers, where soil and soil water chemistry are in equilibrium, where chemical reactions take place at steady-state rates and water and nutrients are taken up by vegetation or flows downward in pre-set proportions. Weathering calculations are based on transition state theory (Brantley et al., 2008). In addition to weathering, other sources or sinks of elements are atmospheric deposition, uptake to vegetation, litterfall and leaching at the bottom of the soil profile. As steady state is assumed in PROFILE, it cannot take changes in soil storage of base cations, carbon or nitrogen into account. In Warfvinge & Sverdrup (1995), the calculations in the PROFILE model are described in detail.

Input data needed for the model are: mineralogy, texture (in the form of exposed mineral surface) and soil density of the different layers, yearly averages of temperature, precipitation, evapotranspiration, soil moisture, deposition of strong acid anions, ammonia and base cations, vegetation uptake, litterfall and soil water concentration of DOC in the different layers.

The weathering reactions with hydrogen ions, water, organic acids and CO_2 are implemented in PROFILE and the weathering is inhibited by concentrations of weathering products, except dissolved silica. Exposed mineral surface, mineralogy and soil moisture have a great influence on weathering rates in PROFILE, as does temperature (Hodson et al., 1996).

ForSAFE

ForSAFE is a dynamic process-based biogeochemical model, developed to study the effect of atmospheric deposition, climate change and forest management on tree growth, soil and soil water chemistry and carbon and nitrogen cycling. It has dynamic feedbacks between soil chemistry, hydrology, forest growth and organic material in the soil. It consists of the dynamic development of the PROFILE model (Wallman et al., 2005; Belyazid et al., 2006), SAFE (Alveteg et al., 1995; Martinsson et al., 2005), integrated with the hydrological PULSE model (Lindström & Gardelin, 1992), the PnET model for tree growth (Aber & Federer, 1992) and the

DECOMP model for decomposition of soil organic matter (Wallman et al., 2006; Walse et al., 1998) (Figure 2).

Being a more complex model, many parameters that are given as input data to the PROFILE model are instead modelled by the ForSAFE model. These include runoff, soil moisture, decomposition of litter, and the uptake of nutrients by trees. ForSAFE is still being developed to answer new research questions (Belyazid et al., 2011; Erlandsson Lampa et al., 2020; Gaudio et al., 2015; Lucander et al., 2021; Phelan et al., 2016; Rizzetto et al., 2016; Yu et al., 2016; Yu et al., 2018; Zanchi et al., 2014; Zanchi et al., 2016; Zanchi et al., 2021). ForSAFE needs more data than the PROFILE model (since it models many more processes), but can in return show the dynamics in parameters interannually as well as on longer time frames. Thus, it is more suited for studies of effects of different scenarios. It can also show effects of the drivers on tree growth, decomposition and soil organic materials, as opposed to PROFILE, which has no such processes.

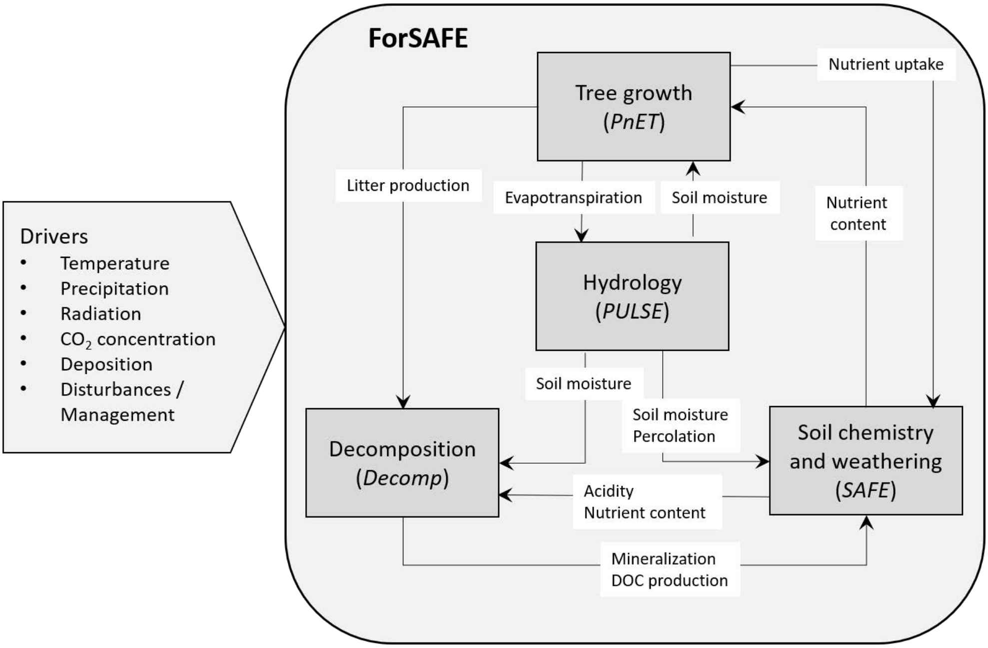

Figure 2 Schematic illustration of the ForSAFE model with drivers, submodels and feedbacks between them. From Zanchi & Brady (2019).

ForSAFE cannot properly be used on longer time scales than a couple of hundred years – usually it is used for the period 1900 – 2100 – because the minerals in the soil are not consumed by weathering, but remain in the same proportions while giving off weathering products. This is of no consequence on the time scales it is usually applied to, but would be on longer. Also, the layers in the soil are fixed and do not migrate downward, which would also be a problem on longer time scales.

Some soil parameters depend on the state of the soil in the beginning of the model period (most often the year 1900). Since these were not measured at the sites at that time, the model is calibrated using measured carbon, nitrogen and base cation content in a calibration year instead.

The three base cations calcium, magnesium and potassium are lumped together in the adsorption process and in the soil solution, but weathering and deposition are modelled for all base cations separately. This can be limiting for the modelling of tree nutrition, but not on weathering studies.

Three different ForSAFE versions, with different time step, were used in this thesis. The original version of ForSAFE had a monthly time step, which was the version used in paper I. It was later developed into a version with a daily time step, to better show the range of the dynamics, rather than monthly averages. This was used in paper III. For paper II, a ForSAFE version with an internal finer time step in the hydrology calculations was developed.

With a monthly time step, (monthly average) soil moisture will never be above the field capacity, as a month is more than enough time for the soil to drain. Months with higher water demand from plants than the precipitation provides will have a soil moisture below field capacity, but above the wilting point. With a daily time step, (daily average) soil moisture will, and should, sometimes be above field capacity. At moderate or heavy rainfall, it might even reach saturation for some layers, most often the uppermost, thin organic layer with small pore volume relative to the precipitation amount. In the daily ForSAFE version without the internal time step, the maximum amount of infiltration in the soil per day equals the pore volume of the uppermost thin organic layer. Sometimes the daily rainfall will exceed this volume and the rain that then cannot infiltrate will instead become surface runoff in the model. In the layer that is saturated, the conductivity will be high, as saturated conductivity is higher than conductivity of an unsaturated soil. Since the conductivity will be high for the whole timestep, the layer will drain down to field capacity during the time step.

In reality, the drainage from one layer to the next does not happen that fast, since the conductivity decreases as soon as the layer no longer is saturated, and the soil do not necessarily drain down to field capacity in one day (depending on texture and the amount of rainfall or infiltration from above). Also, in reality, more rain than the volume of the upper layer infiltrate per day, since the infiltration from above and the draining to the next layer happens simultaneously and there thus will be room for more water to pass through during a day than can fit at any one moment. At very heavy rainfalls some surface runoff does happen.

Both these issues can be a problem when modelling drought and rewetting of the soils in detail, even if they have small consequences usually. Both are also avoided if an internal smaller time step is introduced in the hydrology modelling. The extra time step was implemented in the hydrology sub model only, as it is not needed for the rest of the model. The effect of the extra time step was more realistic soil moisture dynamics at high rainfalls, diminishing levels of surface runoff, but small effects on soil chemistry or the rest of the outputs.

Critical biomass harvesting

In paper IV, weathering calculations were used together with base cation uptake and removal with whole-tree harvest to calculate critical biomass harvesting, which is the maximum amount of base cations that can be removed by harvest without harming the soil with regards to base cation status and acidification (Akselsson & Belyazid, 2018). This was coupled with ANC (acid neutralizing capacity, the concentration of base cations and ammonia minus that of strong acid anions) in soil water. An index was thus created for when whole-tree harvesting can be considered sustainable (ANC is positive and there is no exceedance of the critical biomass harvest), not sustainable (ANC is negative and there is an exceedance of the critical biomass harvest) or at risk of being unsustainable (ANC is negative or there is an exceedance).

3.3 Input data for the weathering models

The PROFILE and the ForSAFE models are different in complexity. PROFILE is a steady-state model, that does not model any changes with time, and also does not model tree growth, hydrology, decomposition of organic material et cetera. ForSAFE, on the other hand, includes the time dimension and models time series of the output data, as well as has whole sub models for processes that PROFILE lacks. Both PROFILE and ForSAFE require data on soil mineralogy, soil texture (ForSAFE in more detail) and density, as well as data on climate, atmospheric deposition and forest management. However, PROFILE needs some additional data that ForSAFE models internally, and ForSAFE requires more detailed data on climate, deposition and forest management (Table 2).

Table 2. Some differences in input data for the PROFILE and the ForSAFE models

Data	PROFILE	ForSAFE
Temperature	Long-term average	Monthly/daily minimum, average & maximum
Precipitation	Long-term average	Monthly/daily values
Atmospheric deposition	Long-term average	Yearly vales
Soil moisture	Long-term average	Internally modelled
Soil texture	Exposed mineral surface in soil layers	Fractions of organic material, rock, gravel, sand, silt & clay in the different soil layers
Soil chemistry	-	Carbon and nitrogen content, base saturation
Forest management	Tree species, average nutrient removal over a rotation	Tree species, planting year, year and extent of thinning and final felling

Climate scenarios

Climate scenarios in the model runs are IPCC (Intergovernmental Panel on Climate Change) scenarios from 2000 (Nakićenović et al., 2000; Figure 3), downscaled to Europe (Simpson et al., 2012). In paper I, the high emission A2 scenario is used. In A2, emissions continue to increase exponentially and CO_2 levels in the atmosphere increase to 850 ppm in 2100. At the sites of paper I, in southern Sweden, this equals a temperature increase of 3.8 °C between 1961-1990 and 2071-2100.

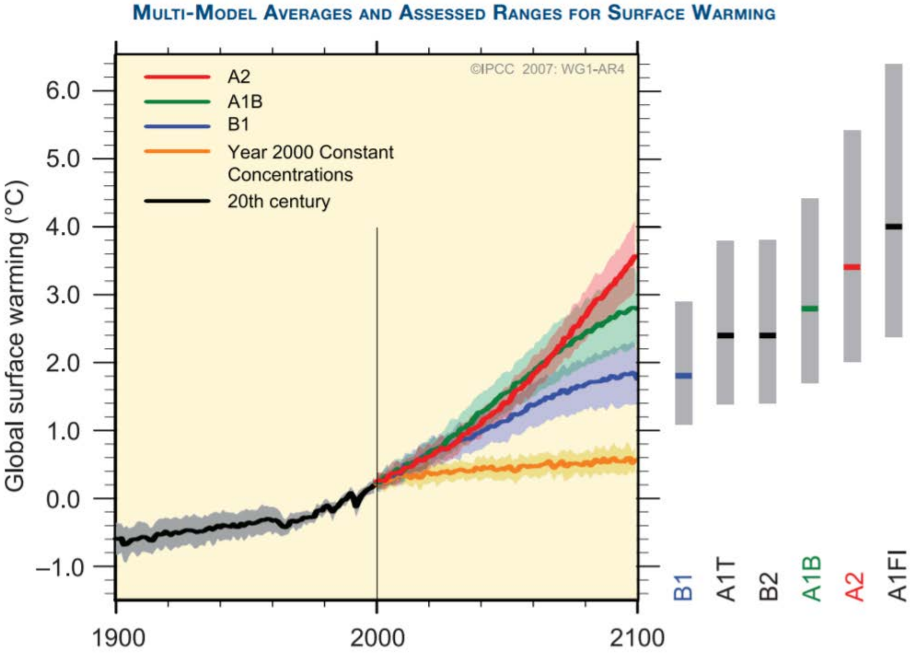

Figure 3. Global averages of surface warming (relative to 1980–1999) for the scenarios A2, A1b and B1, averages from multiple climate models. The orange line is for constant CO_2 concentrations after year 2000. From IPCC (2007), Figure SPM.5.

In papers II and III, the "balanced" scenario A1b is used. This is a slightly less severe scenario, where emissions increase up to 2050 and then decrease slowly. CO_2 levels in the atmosphere increase to 720 ppm in 2100. At the Swedish sites, the 30-year average temperature increases by 3.3 °C to 5.2 °C between 1961-1990 and 2071-2100. The temperature increase is largest in winter and especially large in the northern sites Högbränna and Holmsvattnet, where January at the end of the 21st century is projected to be more than 8 °C warmer than in 1961-1990. The smallest temperature increase for any month is 2.5 °C (Ammarnäs in July). Precipitation is more uncertain (Kjellström et al., 2018), but is projected to increase by 30-40 % in winter. In the warmer seasons, changes are mostly smaller and could go in both directions. Photosynthetically active radiation (PAR), i.e. the part of incoming solar radiation that can be used by plants for photosynthesising, is projected to decrease at all seasons and sites, due to increased cloudiness.

When preparing the scenario data for the A1b scenario for the daily ForSAFE version, it was noticed that the PAR values, as monthly averages, were significantly lower than the values used for the monthly ForSAFE runs with the A2 scenario. This earlier downscaling of the A2 scenario was done in a simpler way and seems to have underestimated the effect of clouds. The newer PAR values for the A1b scenario were on equal levels as measured values from the ICOS program (Carrara et al., 2018), as well as modelled values from SMHI (Swedish Meteorological and Hydrological Institute) (Landelius et al., 2001). The PAR values affect tree growth, and a parameter in ForSAFE for light use efficiency needed to be adjusted for the new PAR levels.

Deposition scenarios

Data on deposition of sulphur, nitrate, ammonia, chloride and base cations, used as input for PROFILE and ForSAFE, comes from measurements at the sites, or nearby SWETHRO sites, if measurements are missing. The ForSAFE model needs data for longer periods than SWETHROs current existence, as does PROFILE, if results representative of a forest generation are wanted (as in paper I, but not in paper IV). Therefore, measurements from SWETHRO are combined with time series of deposition evolution from the research programs ECLAIRE for 1900-1960 (ECLAIRE, 2021) and CLEO for 1961-2100 (Naturvårdsverket, 2016). In paper I, a scenario with constant deposition since 1900 was also used, to investigate the effect of the acidifying pollution by itself on weathering.

Forestry scenarios

The SWETHRO sites used in these studies are *Picea abies* plantations or forests, with mostly even-aged tree stands. In the future forestry scenarios, they are thinned and clear-cut at the same ages as was done previously at the sites, or, if this is unknown, according to Swedish forestry recommendations (which are different depending on site fertility and place in the country). In clear-cuttings in stem-only scenarios, 95 % of stems are harvested and branches and treetops are left on site. In clear-cuttings in whole-tree harvesting scenarios, used in papers I and III, branches and tree tops are also removed, together with 95% of the needles, but stumps and roots are left on site. In paper I, a scenario with no harvesting or thinning at all is also modelled.

4 Results and Discussion

4.1 Temporal variation of weathering rates (paper I)

The ForSAFE model has previously been used on several SWETHRO sites and it has been shown that it can recreate measured values well. Weathering, though, is not measured, and the ForSAFE weathering rates had not been thoroughly investigated or compared to other methods. PROFILE-modelled weathering rates, on the other hand, have been evaluated against other methods many times and have been shown to be comparable to other methods (Akselsson et al., 2019).

In paper I, ForSAFE modelled weathering rates were compared with PROFILE values for the two sites Västra Torup and Hissmossa in southern Sweden, and the dynamic of the weathering rates and how it reacted to different drivers was studied. PROFILE was used on values representing the forest rotation starting and ending in the 21st century (2010-2080 in Västra Torup and 2030-2100 in Hissmossa). The results were compared with average weathering rates for the same time periods from ForSAFE. For the ForSAFE runs, several scenarios were modelled, to understand the separate effects on weathering from different drivers, as both changes in climate, acidification and nutrient removal with harvest affect weathering rates.

The PROFILE weathering rates were close to the long-term average rates from the base scenario from ForSAFE (Figure 4). In Västra Torup the difference was very small, whereas in Hissmossa, weathering in layer four in the soil profile was smaller according to the ForSAFE model than according to PROFILE. This was due to the fact that the modelled ForSAFE soil moisture was usually lower there than what the input value to PROFILE was set to be.

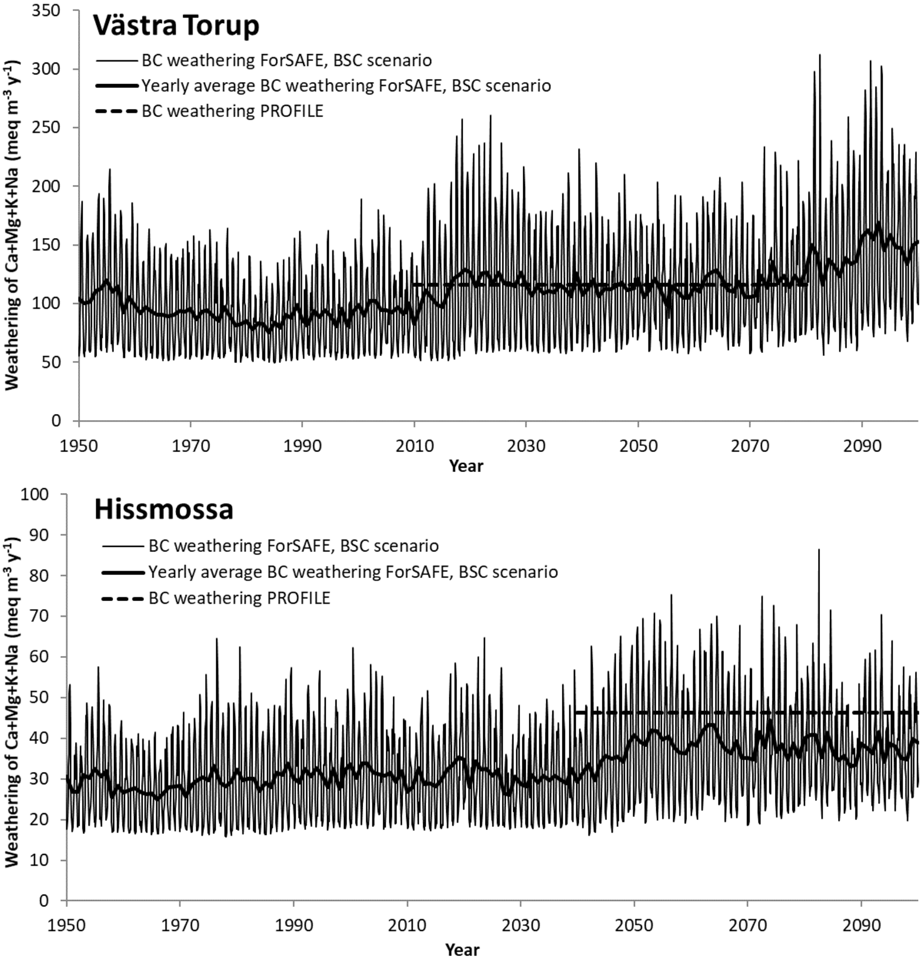

Figure 4. Weathering rates of base cations for Västra Torup and Hissmossa, monthly values and yearly averages of the base scenario from ForSAFE with a monthly time step. The PROFILE weathering rate is shown for the time period it represents, the forest rotation beginning in th 21st century.

According to the ForSAFE model, weathering varied a lot during a year, with much higher weathering during summer and lower during winter. It also varied on longer time scales, depending on clear-cuttings, age and size of trees, climate and the acidification status of the soil. The weathering of all base cation except calcium was supressed by acidified conditions in the soil, even though reaction with hydrogen ions is one of the main weathering pathways. This was because concentrations of dissolved aluminium were elevated in the acidified soils, and aluminium is a weathering product of silicates, but not of apatite (as it does not contain any aluminium at all). About half of the weathered calcium came from weathering of apatite in these two soils and acidification increased weathering of apatite. Climate change, on the other hand, increased weathering of all of the base cations, by

7 % °C^{-1}. This is equal to the average value from a Canadian study (Houle et al., 2020). Clear-cuttings increased weathering for about 30 years (the same time period as effects could be seen on base cation concentrations in the soil water in paper III), and clear-cuttings with whole-tree harvesting increased weathering with just one percent more than clear-cuttings with stem removal only. This is far less than the difference in base cation removal due to whole-tree harvest compared to harvest with stem-only removal. This shows that increased weathering because of whole-tree harvesting can in no way compensate for the losses of base cations in the removed branches and tree tops.

Västra Torup and Hissmossa are situated very close to each other and therefore experience almost the same climate and deposition. This means that any differences in weathering or base cation concentrations between them is not due to climate or deposition, but rather to soil properties. According to measurements, Hissmossa's soil was coarser than Västra Torup's, especially in the fourth soil layer in Hissmossa. This meant that it had a lower exposed mineral surface. Therefore, the weathering in Hissmossa was only a third of the weathering in Västra Torup, according to the models. The coarser texture in the fourth layer also led to lower modelled soil moisture there (as coarser soil holds less water), whereas the same soil moisture value was used in all layers in PROFILE. Therefore, total base cation weathering according to PROFILE was 18 % higher than the average weathering according to ForSAFE during the same time period. This shows how important texture and soil moisture is for weathering.

4.2 Climate effects on weathering and base cation cycling in different regions of Sweden – differences and similarities of future development (papers II and III)

In papers II and III, two ForSAFE versions with a daily time step were used on one SWETHRO site in each of the climate regions of Sweden in Figure 1. Paper II studied weathering dynamics in a warming climate and the effect of summer drought. Paper III studied the effect of whole-tree harvesting on base cation and nitrogen concentrations in soil solution, and on the tree growth of the next planted forest stand. The northernmost climate region in the mountains was excluded from this study, as whole-tree harvesting is not reasonable there.

In paper II, a ForSAFE version using an internal extra time step was used for the first time, to make sure large or moderate rainfall events would infiltrate the ground instead of running off on the soil surface in too large amounts.

In a future warmer climate, not only will average temperatures increase, but the risk of extreme events is also predicted to increase (Kellomäki et al., 2008). Periods of extremely dry, warm summer weather (such as the extremely dry summer of

2018, Toreti et al., 2019) might become more common, longer lasting and more extreme. Temperature and precipitation have large effects on soil weathering, and climate changes as well as dry episodes can therefore lead to negative effects on soil chemistry and plant nutrition. In paper II, two climate scenarios were run, one climate change base scenario according to the IPCC A1b scenario and one scenario with five consecutive years of summer drought, similar to the drought event in 2018, on top of the base scenario.

Like in paper I, the modelling in paper II showed that weathering rates have a strong seasonal dynamic, with much lower rates during the winters than during the summers. The daily ForSAFE version showed in more detail how variable summer weathering rates are, depending on soil moisture and temperature. Despite the strong dependence of temperature for the weathering and the strong gradient in temperature from southern Sweden to northern, there was no geographical gradient in yearly average weathering for these seven sites, as soil texture and mineralogy have an even stronger effect on weathering. The average seasonal dynamics looked a bit different across regions though, as summer weathering rates have a broader flatter peak, influenced by both temperature and lack of soil moisture, while the northern sites have seasonal dynamics reflecting the temperature curve, as soil moisture usually is sufficient.

The model results indicated that the weathering rates will increase with 5-17 % $°C^{-1}$ in the A1b scenario, with an average of 6 % $°C^{-1}$, which is very close to both the value in paper I and the average in Houle et al. (2020). The relative increase was projected to be largest in the two south eastern sites, with relatively coarse soils, already today dry summers and low total weathering rates.

Also changes in seasonal dynamics due to climate change differed between regions. At sites in southern Sweden, future weathering increase will occur throughout the year, though less in winter and more in spring and summer. In the north, the increase in weathering during winters will almost be negligible, even though the temperature increase during winter will be high. This is because the winter temperatures still will mostly be below zero. The increase in summer weathering rates were projected to be even more pronounced than in the south, as there will generally be less dry conditions during summers. In this modelling, summer soil moisture was not projected by ForSAFE to decrease further – sites in southern Sweden already have recurrent dry soils during summers and this will not accelerate. According to Rousteenoja et al. (2017), climate models disagree on whether summer soil moisture in Sweden will decrease or not, using the more severe RCP8.5 climate scenario, but the average projection is a small decrease.

The extreme drought scenario showed different results in different parts of Sweden and for different textured soils (Figure 5). Southern Sweden was affected most and here the weathering during drought summers can temporarily become as low as the winter weathering rates. Total yearly weathering during the drought years in the most affected site was only 78% of the weathering of the base scenario. In the north, the soils will not dry out as much despite the low precipitation, according to the model. In the northernmost site Ammarnäs, the clearer warmer weather of the drought scenario even gave higher weathering for some of the drought years than what the base scenario gave, though the overall effect of the drought was a slight decrease in weathering. In the sites with a coarser soil texture, soil moisture decreased rapidly to very low levels, but also increased quickly when the autumn rain started. In the less coarse sites, soil moisture did not respond as quickly either to the drought or the end of the drought. Weathering rates respond to both soil moisture and temperature, and thus varied a bit during the summer, but increased in the autumn when the soil moisture was restored.

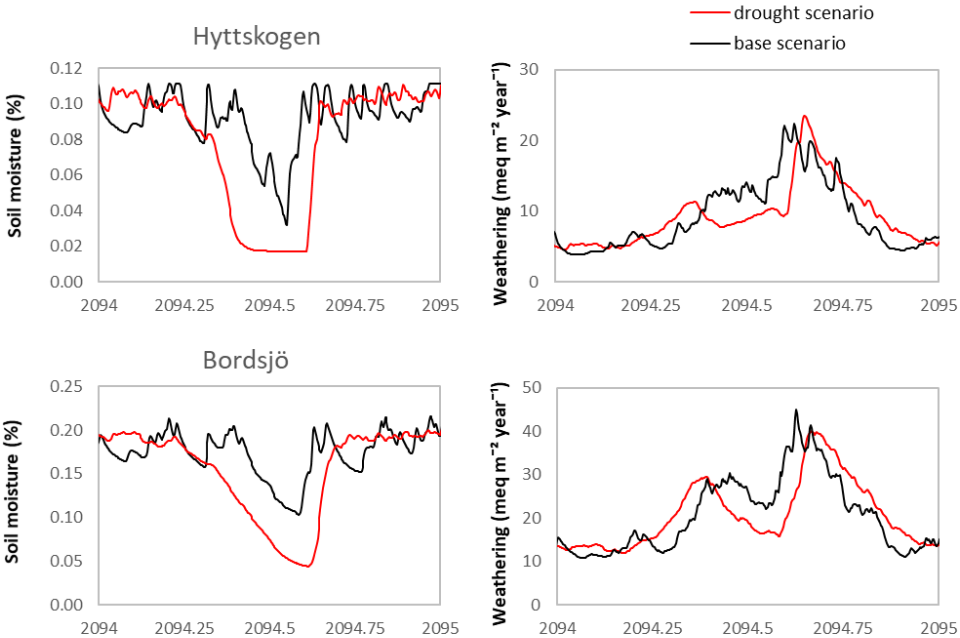

Figure 5. Effect of drought scenario and base scenario on soil moisture and weathering of base cations in Hyttskogen, a coarse textured site, and Bordsjö, a less coarse textured site, for one of the drought years.

The same sites, except the northernmost, were in paper III subjected to modelled whole-tree harvesting and stem-only harvesting scenarios. Experimental studies have shown that whole-tree harvesting has an effect on base cation concentrations in the soils, but that the effect diminish with time and cannot be seen 30 years after the harvest (Zetterberg et al., 2013). As ForSAFE is a process-based model, using it could show how the catch-up growth in the whole-tree harvest sites happens. This was done on four sites in two regions by Erlandsson Lampa et al. (2019), who showed that the pool of base cations incorporated in soil organic matter was important for the catch up of the whole-tree harvest sites. The study was here upscaled to climate regions covering most of Sweden. Modelling of the six sites showed the same pattern as the experimental studies: a large difference in base cation and nitrogen concentrations in soil solution in the first years after harvest, that gradually decreased and were gone after 30 years, or earlier in the northern sites. Clear-cuttings give rise to increased concentrations and leaching of nitrogen and potassium (which is one of the more mobile of the base cations), that gradually go back to normal concentrations as the new forest stand grows and take up more nutrients. In ForSAFE, which does not differentiate between calcium, magnesium and potassium in soil solution, this was seen as an increased leaching of these base cations. This increase after harvest happened in both the stem-only and the whole-tree harvesting scenarios, but the increase was much larger in the stem-only scenario, where nutrient rich branches and needles were left on site to decompose (Figure 6). Biomass growth was slightly slower after whole-tree harvesting than after stem-only harvesting, according to the model, especially at the northern, more strongly nitrogen limited sites. However, the growth gradually catched up. In none of the sites the growth was limited by base cations.

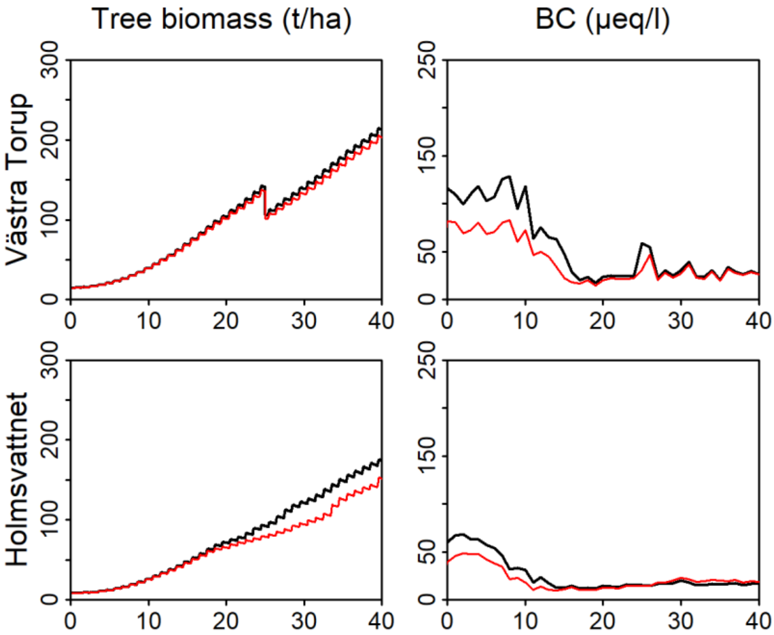

Figure 6. Effect of whole-tree harvesting (red line) and stem-only harvesting (black line) on biomass of the next generation and on base cation concentration in soil water, for one southern and one northern site in paper III. The time, in years, after harvesting is on the X axis.

In the whole-tree harvesting scenario, more base cations were removed by harvest, giving rise to the lower soil water concentrations of base cations. In the stem-only harvesting on the other hand, the higher soil water concentrations led to higher leaching of base cations from the soil with runoff water. This was part of the reason for the diminishing difference with time. In northern Sweden, another reason for the diminishing difference between treatments, was that tree uptake of base cations was larger after the stem-only harvesting, also removing base cations from the soil.

The results of this study are in line with experimental studies and the earlier ForSAFE study (Erlandsson Lampa et al. 2019), but differs from modelling studies using models that lack feedback between soil concentration and vegetation uptake, where the differences between treatments does not diminish with time in the same way (Zetterberg et al., 2016; de Jong et al., 2017). In earlier studies, the lack of dynamic weathering rates in the models has been proposed as a possible explanation for differences between the experiments and the model results (Zetterberg et al., 2016; Paré & Thiffault, 2016). However, both the studies in paper III and paper I showed very small effects of whole-tree harvesting on weathering. This indicates that increased weathering when needed by the trees cannot be expected to compensate for increased removal of base cations at harvesting.

4.3 Upscaling for policy - PROFILE in regions (paper IV)

In paper IV, time series for soil solution chemistry at 26 SWETRHO sites were combined with calculations of exceedance of critical biomass harvesting after whole-tree harvesting, as a basis for a risk assessment. Weathering, modelled with PROFILE, is an important part of these calculations. It is also the parameter that is the most demanding to estimate, and which limits how many sites can be included, since analyses of soil chemistry and soil texture from different layers in a soil pit are required.

The study showed that, although weathering potentially has a large effect on the risk for whole-tree harvesting not being sustainable, for most of the 26 sites, two other factors were more crucial (Figure 7). The current acidification situation, which is largely due to historical deposition, and the size of base cation losses by harvesting, which in its turn depends on the size of the trees, ended up determining the general picture. In two places in southern Sweden, weathering was relatively high and the soil conditions were good, and they were therefore classified in the lowest risk class. However, most sites in southern Sweden ended up in the worst risk class, despite the fact that the variation in weathering is quite large. This was because the base cation losses through whole-tree harvesting were generally high there, in relation to the weathering rates. Thus, in most soils in southern Sweden, weathering was not high enough for whole-tree harvesting to be long-term sustainable, especially not since the soil acidification status today is poor.

The dynamic modelling in the other papers showed that weathering rates can be affected indirectly and directly by climate change. The increasing effect from whole-tree harvesting is assessed to be very low in papers I and III. The direct effects from climate can go in both directions, depending on if there will be droughts or not (papers I and II). The studies indicate that even if the weathering rates increase due to higher temperatures, it will not be enough to compensate for losses with whole-tree harvesting, while contributing to recovery from acidification, which is in line with earlier studies (Akselsson et al., 2016).

PROFILE was used for this study, which means that seasonal variations in climate and thus also weathering rates were not considered. The study in paper I showed that the weathering levels from the steady-state model PROFILE and the dynamic model ForSAFE are relatively equal, when average ForSAFE values for several years are compared to PROFILE values. The disadvantages of using the steady-state model can therefore be considered to be small when calculating exceedance of critical biomass harvesting, where average values are required. PROFILE shows steady-state weathering, and the exceedance estimation indicates a direction on whether forestry is sustainable or not. It is similar to what has been done for a long time with critical loads calculations (Sverdrup & de Vries, 1994). To use steady-state calculations and only get a direction, without any information about the starting point, can be seen as a drawback in these kinds of assessments, which is one reason for dynamic models being developed (Grennfelt et al., 2001). Dynamic models, however, require greater efforts to set up and run on many sites. By combining steady-state calculations with measurements that show the starting point, the results are less limited.

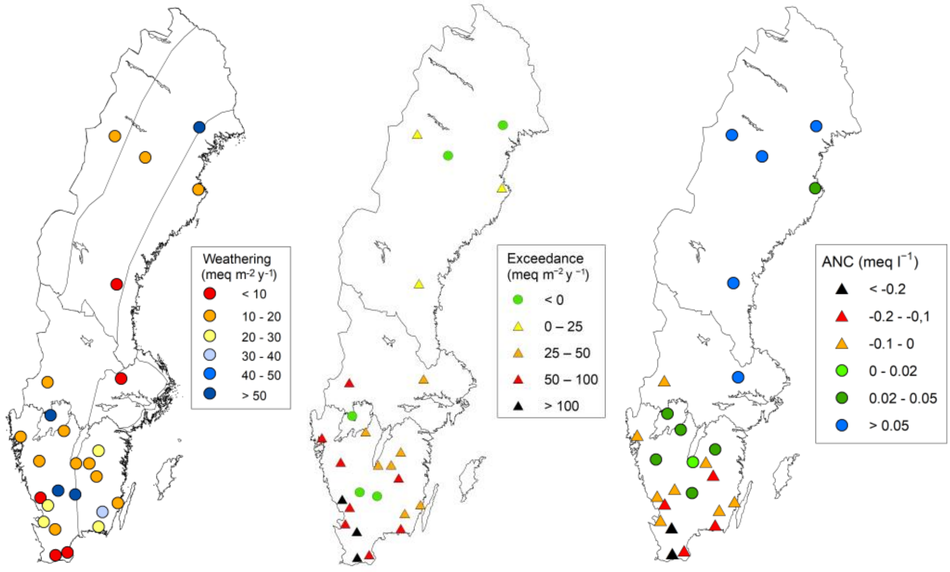

Figure 7. Maps of the sites, with PROFILE-calculated weathering rates (left), exceedance of critical biomass harvest if whole-tree harvesting is applied (middle) and ANC in soil solution (right).

4.4 Lessons learned from using different approaches

Several of the SWETHRO sites have been modelled using different models, different model versions, or with differences in input data, in different papers (Table 1). Comparisons between the results can help increase process understanding and highlight influence of input data.

In most cases, the dynamic biogeochemical ForSAFE model with a daily time step gives 50 % - 90 % higher weathering rates, as averages over time, as the simpler steady-state geochemical model PROFILE or ForSAFE with a monthly time step (Table 3). The rates are not entirely comparable between paper II and paper IV, since the time dimension is lacking in PROFILE. The uptake to vegetation used in paper IV is an average for a whole forest rotation (as that is what PROFILE needs) and temperature and precipitation averages are used, but the deposition data is from a much shorter period. In the ForSAFE modelling in paper II, the uptake to vegetation changes with forest age and year to year variation in weather and deposition have an effect on weathering, leading to rather large year to year differences in weathering (which is why more stable 30-year or longer averages are shown for ForSAFE in Table 3). The higher weathering rates from ForSAFE daily are mostly due to the variable soil moisture, with wetter days that contribute more to the weathering in a non-linear way. The ForSAFE version with a monthly time step, where soil moisture is never above field capacity, give more similar weathering rates to PROFILE in the calculations in paper I.

The biggest difference between PROFILE weathering rates in paper IV and ForSAFE average weathering rates in paper II occurs at the northernmost site Ammarnäs. It has been modelled using different assumptions on mineralogy in the two different model studies, and therefore has very different weathering rates. In the modelling using ForSAFE (paper II), Ammarnäs is modelled with more calcite rich mineralogy that can reproduce the average measured soil water chemistry. In contrast, the PROFILE study (paper IV) uses mineralogy estimated from measured total chemistry, that cannot reproduce measured soil water chemistry. The area around Ammarnäs has very varied mineralogy with calcite occurrences in some places (Greiling et al., 2018; Grimmer et al., 2016), and even though the soil sampled at the site did not have high calcium concentrations, it is likely that parts of the site or its immediate surroundings has higher calcium concentrations, giving rise to the high concentrations of calcium in the soil water. Therefore, it was modelled using a mineralogy containing calcite in paper II. The ForSAFE model gives 13 times higher modelled base cation weathering rate than the PROFILE model, but this is not an effect of model differences, in more than that ForSAFE is more equipped to show inconsistencies in input data, e.g. that the measured total chemistry cannot by itself give the measured soil water chemistry. Further investigations of mineralogical distribution and soil water flows in and around this site would be needed to explain the high calcium concentrations in soil water, and the high temporal variation. When the site is modelled with the ForSAFE model, using the same mineralogy as in paper IV, the average weathering rate is 89 % higher than the PROFILE weathering, like most of the other sites.

Table 3. Average weathering rates of base cations (meq m^{-2} y^{-1}) for the modelled sites.

Site name	Model	Time period	BC Weathering	Comment
Grankölen	PROFILE	2016-2018	52.6	paper IV
Ammarnäs	PROFILE	2014-2018	16.5	paper IV
	ForSAFE daily	1990-2019	31.2	paper IV mineralogy
	ForSAFE daily	1990-2019	228.9	paper II
Högbränna	PROFILE	2014-2018	16.9	paper IV
	ForSAFE daily	1990-2019	28.4	paper II
Holmsvattnet new	PROFILE	2014-2018	15.2	paper IV
Holmsvattnet	ForSAFE daily	1990-2019	36.0	paper II
Storulvsjön	PROFILE	2014-2018	7.7	paper IV
Hyttskogen	PROFILE	2014-2018	3.6	paper IV
	ForSAFE daily	1990-2019	6.8	paper II
Blåbärskullen	PROFILE	2014-2018	15.9	paper IV
Södra Averstad	PROFILE	2014-2016	54.1	paper IV
	ForSAFE daily	1990-2019	54.4	paper II
Stora Ek	PROFILE	2014-2018	18.4	paper IV
Hensbacka	PROFILE	2014-2017	12.9	paper IV
Solltorp	PROFILE	2014-2018	25.7	paper IV
Storskogen	PROFILE	2014-2018	19.3	paper IV
Bordsjö	PROFILE	2014-2018	10.8	paper IV
	ForSAFE daily	1990-2019	16.7	paper II
Värnvik	PROFILE	2014-2018	10.8	paper IV
Fagerhult	PROFILE	2014-2018	13.9	paper IV
Mellby	PROFILE	2014-2018	58.1	paper IV
Tagel	PROFILE	2014-2018	63.0	paper IV
Borgared	PROFILE	2014-2018	5.3	paper IV
Rockneby	PROFILE	2014-2018	17.0	paper IV
Timrilt	PROFILE	2014-2018	22.2	paper IV
Alsjö	PROFILE	2014-2018	30.2	paper IV
Kullahus	PROFILE	2014-2018	20.2	paper IV
Vång	PROFILE	2014-2016	24.0	paper IV
Hissmossa	PROFILE	2014-2018	12.2	paper IV
	PROFILE	2041-2100	21.5	paper I, A2 climate
	ForSAFE monthly	2041-2100	18.5	paper I, A2 climate
Västra Torup	PROFILE	2011-2080	52.5	paper I, A2 climate
	ForSAFE monthly	2011-2080	57.7	paper I, A2 climate
	ForSAFE monthly	2011-2080	60.3	not published, A1b climate
	ForSAFE daily	2011-2080	82.8	paper II, A1b climate
Maryd	PROFILE	2014-2018	4.1	paper IV
Stenshult	PROFILE	2014-2018	4.9	paper IV

The Hissmossa and the Södra Averstad sites show the importance of an accurate modelling of soil water content and an adequate amount of input data: the PROFILE model does not model soil water content at all, but instead uses an estimated soil moisture value based on observation in the field. In the Södra Averstad case, the observation is that the site is slightly wet, which is reasonable given its placement low in the landscape, very close to the shore of the large lake Vänern. ForSAFE, on the other hand, models soil water content dynamically, using water inputs, texture, soil depth and the assumption of drainage downwards. It does include the possibility of including slope along a transect (Zanchi et al., 2021), but in our studies (papers II and III) we did not have data for that, and settled for modelling a single soil profile for each site. For Södra Averstad, ForSAFE simulated recurrent summer droughts due to the combination of precipitation level, vegetation demand of water in this rather nutrient rich site and the texture of the soil. Recurrent summer droughts do not seem to be likely, given the observed moist conditions, the placement of the site and the fact that few of the soil chemistry measurements from SWETHRO are missing (which they often do at dry locations, because soil water cannot be extracted using lysimeters when the soil moisture is too low). Because of this, weathering rates as modelled by ForSAFE with a daily time step were almost exactly equal to those modelled by PROFILE, contrary to the other sites. Hissmossa has a similar issue, as described in chapter 4.1.

In Västra Torup, a test was made where 1622 different combinations of mineralogy solutions, all from the A2M model run of the site specific measured total chemistry, were used as input to PROFILE (data not published). Everything else was kept the same in all runs, to estimate the effect on weathering of mineralogy, given the measured total chemistry. The median base cation weathering of these test runs was very close to the weathering of the average mineralogy (the mineralogy that is usually used) and the weathering for all runs was between 84% and 121% of the median weathering for all of the 1622 runs (Figure 8). The relative variability in weathering was very small (about ±3%) for sodium, intermediate for silica, aluminium and potassium (about ±16%), larger for calcium (±33%) and largest for magnesium (-45% - 145%). This means that, at least at this site, regardless of which minerals the measured sodium in the total chemistry is allocated in, the weathering of sodium will be approximately the same. On the other hand, the allocation of magnesium has a much bigger effect on the magnesium weathering. Allocations to the easily weatherable mineral hornblende leads to the highest weathering rates. This could explain the observation in Stendahl et al. (2013), that weathering levels of calcium and magnesium are similar between the depletion method (very long-term average) and PROFILE (contemporary value), i.e. less variable in time and more decided by what minerals are present in the soil, whereas weathering levels of potassium and sodium differ between the depletion method and PROFILE (as they are less decided by mineralogy and instead varies in time due to changing environmental factors).

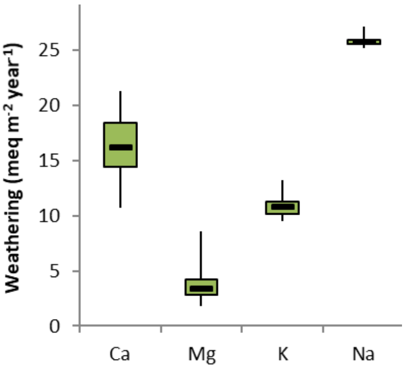

Figure 8. Variation in PROFILE-modelled weathering of calcium, magnesium, potassium and sodium using different possible A2M solutions of mineralogy in Västra Torup. Minimum, 25th percentile, median, 75th percentile and maximum values.

In another test using PROFILE in Västra Torup, one input parameter (base cation and nitrogen uptake to vegetation, the distribution between soil layers of the uptake, nutrient in litterfall, soil moisture and soil water DOC) at a time was changed between the value used for PROFILE in paper I, and a value derived from the ForSAFE modelling of the site (also in paper I). The effect on weathering of these differing input data was investigated (data not published). As in Hodson et al. (1996), the largest effect by far was from the soil moisture.

Västra Torup was also modelled using different ForSAFE versions and different future climate scenarios (Table 3). The IPCC scenarios A2 and A1b was modelled using the ForSAFE version with a monthly time step. The A1b future climate scenario was also modelled for Västra Torup using the two ForSAFE versions with a daily time step, with and without the internal smaller time step for infiltration of precipitation and percolation between soil layers. ForSAFE with a monthly time step predicted a very small increase in water stress and decrease in soil moisture in the future, both for the A2 and the A1b scenario, whereas ForSAFE with a daily time step predicted the same amount of water stress in the future as in the past forest rotation, using the A1b scenario. For the version with an internal hydrological time step, the soil did not dry out as severely during dry periods, and infiltrated more water during large rainfalls. The average soil moisture situation was very similar between versions, but the dynamic differs (Figure 9). A shorter time step should be able to capture the dynamic better than the longer, but as there is no measured soil moisture at the sites, this has not been confirmed.

Tree growth is an important parameter, both in itself and since it affects soil moisture and nutrient uptake. For Västra Torup, ForSAFE with a monthly time step, forest growth is predicted to decrease in the future forest rotation, compared to the 20th century forest stand (that was clear-cut in 2010), when the A2 scenario was applied. For the less severe A1b scenario, ForSAFE monthly predicts a small increase in growth in the future generation. ForSAFE daily tend to predict a larger increase in growth, both for the version with and the version without the extra internal hydrology time step.

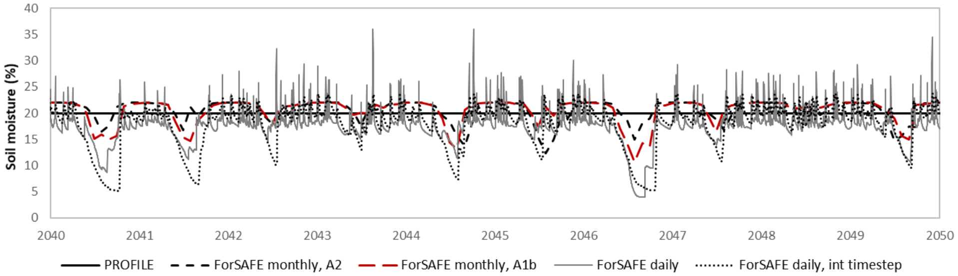

Figure 9. Soil moisture for PROFILE and different ForSAFE versions and future climate scenarios (PROFILE and ForSAFE monthly using the A2 scenario, ForSAFE monthly and ForSAFE daily with and without the internal time step for hydrology using the A1b scenario).

4.5 Future perspectives

The studies in this thesis emphasize the importance of seasonal variation of weathering rates for base cation cycling in a changing climate. Process-based models with high temporal resolution, like ForSAFE, have the potential to increase the accuracy in future predictions of nutrient availability, and should thus be further developed and used. Some processes in ForSAFE of great importance for weathering would need to be further investigated and validated:

The modelling of the Hissmossa and Södra Averstad sites shows how important a good knowledge of the hydrological situation of the sites are. ForSAFE is made for recharge areas, which all of the modelled sites are, but even recharge areas are affected by the topography of the immediate surroundings. Observations of vegetation on the sites Hissmossa and Södra Averstad show that they do not dry out as much and regularly as ForSAFE predicts. There are no measurements of soil moisture or ground water level at the SWETHRO sites, which makes it impossible to validate ForSAFE's modelling of soil moisture on these sites. In a future study to improve modelling, soil moisture as well as ground water level could be measured at the same site during different seasons and at different soil depths. The data series could then be compared to ForSAFE values, in order to find out in what type of settings ForSAFE does not succeed in predicting the soil moisture situation, and thus the weathering.

Zanchi et al. (2016) developed ForSAFE-2D, that can model a series of soil profiles along a hill slope. This ForSAFE version can have a saturated zone in the bottom of each soil profile, i.e. it can model both recharge and discharge areas. For ForSAFE-2D, soil data for several soil profiles in a row are needed. Most sites with available deposition data do not have that. Further development and use of

ForSAFE-2D has the potential to increase the accuracy of modelled soil moisture and thus weathering rates.

Measurements of soil moisture would also be useful for PROFILE modelling, since PROFILE needs average soil moisture in different layers as input data. Usually, this data comes from observation on site, which puts the site in one of six moisture classes (where the middle class is by far the most common in Swedish forests), associated with one value for soil moisture (Warfvinge & Sverdrup, 1995). Since texture influences the water holding capacities of the soil substantially and texture can vary between layers, more thorough knowledge on soil moisture levels in the actual sites would be useful to improve the input given to PROFILE.

Both PROFILE and ForSAFE tend to overestimate weathering in deep soil layers. This happens both for layers under the groundwater table (Erlandsson et al., 2016, Erlandsson Lampa et al., 2020) and for deep unsaturated soil layers. The modelled weathering rates there seem high, both compared to other methods for weathering calculations and considering that the weathering rates should be small in C horizons that have only small occurrences of secondary minerals, and no colour difference compared to the unweathered parent material of the soil (Tamm, 1920; Snäll & Ek, 2000). One possible explanation is that the inhibiting effect of soil solution silica is not implemented in neither ForSAFE nor PROFILE, since the concentrations of dissolved silica tend to be low in the surface soils. When this inhibiting effect was implemented in a test, it made the base cations concentrations in the runoff to the stream in Svartberget in northern Sweden more accurate (Erlandsson et al., 2016, Erlandsson Lampa et al., 2020). The process might be needed for the regular ForSAFE versions too, even though the model seldom is used for deeper soil layers than 50 cm depth.

When using A2M, knowledge of which minerals are present in the area is important. Small quantities of some easily weatherable minerals can influence weathering rates disproportionately. In the studies in this thesis, regions of different mineralogy from Warfvinge & Sverdrup (1995) were used. These regions are based on samples from about 100 soils across Sweden. Since both bedrock and soil mineralogy vary widely in some parts of the country and not as much in others, higher spatial resolution of the sampling in the more complex regions would reduce uncertainties.

ForSAFE, as used in paper II and III, can reproduce measured tree biomass very well for some of the sites and less well for others, for example the unusually dry and coarse textured site Hyttskogen. Future development of forest growth is also a bit unclear, at least in Västra Torup, where different model versions have been compared. Tree biomass affects evapotranspiration and thus soil moisture, so modelling the tree growth right is important also for weathering rates, and further refinement of tree growth processes in ForSAFE is therefore an important future field of research.

5 Conclusions

This thesis highlights the potential of dynamic modelling as a complement to steady-state modelling of weathering rates. Whereas the steady-state model PROFILE can be used in simplified sustainability assessments with a higher spatial resolution but no temporal resolution, such as critical biomass harvesting calculations (paper IV), the dynamic model ForSAFE can be used to simulate weathering and base cation cycling at high temporal resolution, to increase process understanding and predict effects of climate change, atmospheric deposition and forest management (papers I, II and III).

The weathering rates modelled using the geochemical steady-state model PROFILE and long-term averages from the more complex dynamic biogeochemical model ForSAFE are of the same size in most cases (paper I). However, in some cases they differ, and those cases provide important knowledge. Limitations in soil input data, e.g. due to large variation of mineralogy at a site, not covered by the measurements, can lead to inaccurate model results, which can be detected in ForSAFE when modelled and measured soil solution chemistry is compared. Differences between soil moisture from field assessments, used in PROFILE, and soil moisture modelled by ForSAFE can also lead to major differences in modelled weathering rates.

ForSAFE shows that weathering rates have a strong seasonal dynamic, with low winter rates and high and highly variable summer rates, that depend on temperature and soil moisture.

Weathering is temporarily affected by clear-cuts, as both concentrations of weathering products in the soil water and the soil moisture levels are affected. Removal of branches and treetops, in addition to stems, has only a small effect on weathering, increasing weathering rates by far less than the loss of base cations through the harvest. This leads to temporarily lowered concentrations in soil water for up to 30 years post-harvest.

Acidification of the soils can have an inhibiting effect on weathering rates, if concentrations of dissolved aluminium increase. In southern Sweden many forest soils have been acidified. Many have started to recover, but the recovery process can be halted or even reversed by unsustainably large harvests of base cations.

Climate change generally increase weathering rates in the Swedish climate. In southern Sweden rates are increased year round, though most in spring and summer, whereas northern Sweden’s winters will still mostly be below zero and have low weathering rates. In southern Sweden low levels of soil moisture during many summers already have an inhibiting effect on weathering rates and this is projected to continue. The increasing weathering rates in spring might become more important for vegetation in the future, as the vegetation period lengthens.

The risk of extreme events increases with a warming climate. Extreme summer droughts can become more common and would inhibit the weathering, especially in southern Sweden.

The soil moisture is highlighted as a key parameter for accurate calculations of weathering rates. It is also an important factor in the modelling of future biomass growth. It is also not measured at the modelled sites. Future studies would be needed, to measure soil moisture and compare it with ForSAFE-modelled soil moisture for different ForSAFE versions, to reduce uncertainties related to soil moisture in PROFILE and ForSAFE. For ForSAFE, this is needed especially in areas with flat surroundings, where runoff is not as efficient as ForSAFE models it to be. For PROFILE, it is most relevant in sites with differing textures in different soil layers.

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List of Papers

I. Kronnäs, V., Akselsson, C., and Belyazid, S.; Dynamic modelling of weathering rates – the benefit over steady-state modelling. *SOIL* 5, 33–47. doi.org/10.5194/soil-5-33-2019. 2019

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