Answer to Referees and Editor, and description of how the comments were handled

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Associate Editor Suzanne Anderson asked for major revisions (10 May 2019), based on the comments from the two reviewers and on our answer to the comments. The manuscript has been thoroughly reworked, based on the comments, and in our opinion substantially improved. In this document, we describe how we answer the comments and describe how we have handled them (pages 1-33), and we also attach the manuscript with track changes (pages 34-98). We first answer the comments and describe the changes related to the comments from Referee 1 (pages 1-24), then Referee 2 (pages 25-27), the Editor (pages 28-31) and finally answers to the comments by the Editor that came along with the decision (pages 32-33). All answers are written in italics, in order to be able to separate between comments and answers. In the comments, the page and line numbers in the original manuscript are referred to, whereas in the answers, the page and line numbers in the revised version (without track changes) are referred to (akselsson_etal_aug2019.pdf)).

Answers to comments by Referee 1 (R1)

Comments from 19 March 2019

R1 starts off with a paragraph with general comments, followed by a long paragraph with specific comments.

General comments

“The authors present an overview or synthesis of base cation weathering studies carried out under the Swedish QWARTS project. The paper is clearly part of a special issue, as much of the text refers to other papers in ‘this issue’. Given the dependence on other papers, it is not a standalone paper but a very ‘Swedish’ view of soil weathering. Nonetheless, the paper has an important objective, to demonstrate that despite the variation in estimated soil base cation weathering rates at the site level, there is general agreement and these data can be used to support the assessment of sustainable forestry. However, the paper falls down in several areas:”

“1. The text is overly long, at times there is extensive repetition within and between sections,”

We have removed redundant and repetitive text and shortened parts that are described in other papers in the special issue. The paper (including first page and references but without Tables and Figures) is now 29 pages, compared to more than 37 pages before. The main changes are listed below:

We have changed to a more conventional structure with a Methods section and a Result and Discussion section. This eliminated much of the repetitions. This is further described in the answer to general comment #2 below.

We have removed descriptions of models not represented in the results, according to the answer to general comment #2 below.

We have reworked the chapter “Potential for biological weathering”, which has reduced the length from 3.5 pages to 1 2/3 pages. We have removed detailed descriptions and discussions that can be found in the paper Finlay et al (in review, this issue), and instead focused on the implications of the new in-sights from a modelling perspective.

We have shortened the chapter about “Future research”, according to the answer to general comment #3 below, and renamed it to “Prospects for method development”.

“2. The text had a tendency to loose focus, the manuscript jumps between project summary, scientific review, and comparison of specific results, and while the authors forewarn of the contents in the abstract, the conclusion more succinctly speaks to the true contribution of the paper, if there are other papers in this special issue, do the authors need to be so broad in their coverage?”

This comment, as well as general comment #3, questions the structure of the paper. Based on these comments, and recommendation ‘a’ (below), we have clarified the aims in the end of the introduction, restructured the text, removed parts that are not contributing to answering the aims, removed repetitious text and shortened parts that are described in other papers in the special issue. The aims now read: “The aims of this study were to (1) investigate the variation in weathering rates from *different approaches in Sweden, with consideration of the key uncertainties for each method, (2) assess the robustness of the results in relation to sustainable forestry and (3) discuss the results in relation to new insights from the QWARTS programme, and propose future research to further reduce uncertainties." The restructuring and shortening of the text is described more thoroughly below, and in answers to general comment #1 and #3.*

-We still want to be a bit broad in the coverage, e.g. including the part of biological weathering and the implications of higher resolution of chemical reactions (to answer aim 3), but we have reduced the length of those parts substantially.

-We have changed the structure, so that it follows a more conventional scheme, with a Methods section and a Results and Discussion section. We start the Methods section by describing how we have compiled the data for the weathering rate comparison based on literature studies, we continue with a brief description of each method (shortened versions of the old ones), including methods for regional applications. Finally we describe how we made the comparison with harvest losses. We removed the descriptions of the models not used for any of the sites in the paper, i.e. WHITCH and Crunchflow. By having a pure Methods chapter, we avoid some of the repetition that now exists, e.g. regarding model descriptions (in the chapter "Methods for estimating weathering rates" and the chapter "Weathering rate comparisons on a regional scale"). In the Results and discussion chapter we start with site level results, continue with regional results and then the comparison with harvest losses. Thereafter, sections about potential for biological weathering, more detailed chemical reactions and prospects for method development follow.

3. Section names and section contents are confusing, the section on future research seems to focus on limitations, while repeating text from previous sections, and generally has the feeling that much of the text could have been integrated into previous sections-

Regarding confusing section names and contents, see answer to general comment #2, where we describe how we have restructured the paper with new section names. We agree that the section about future research is too long and insufficiently focused. We have shortened it substantially, renamed it to "Prospects for method development", and removed or moved parts that do not fit in.

4. Unfortunately, much of the comparison between weathering estimates is too qualitative, there is no quantitative assessment, statements such as ‘they agree’, ‘do not agree’ or ‘estimates are similar’ need quantitative support.

We have added medians, maximum, minimum and “maximal deviation from median (%)” in Table 3, with site-level weathering rates, and we refer to those numbers in the text (sectin 3.1). For the regional-level comparison and the sustainability assessments, we have added numbers to quantify the difference. In the regional-level comparison we compare medians and the width of the intervals in the text (section 3.2). In the sustainability assessments, we give the difference between weathering rates and harvest losses in %, in the text (section 3).

I suggest the authors (a) step back from their manuscript and try to pinpoint their exact (unique) contribution, (b) they should remove repetitious text, and remove text that is described (reviewed) elsewhere in the special issue, and (c) add a stronger quantitative element to their comparison / assessment of weathering / sustainable forestry.

(a) See answer to question #2 above.

(b) See answer to question #1 above.

(c) See answer to question #4 above.

Specific comments

Page 2 L1. It was internationally recognised during the 1970s but regionally recognised long before that... 1 to 2 decades!

We changed from “first recognised” to “internationally recognized”. Moreover, we changed “during the 1970’s” to “the late 1960’s”, in accordance with what is written in the introduction. (Page 2, line 1)

L2. one could argue that the peak was a little later... 1980s to 1990s?

We have changed to “1980s and 1990s”. (Page 2, line 2)

L4. Reword / clarify ’more harvest’, more correctly you are referring to the use of forest residues for renewables!

We replaced “more harvest” with “forest residues”. (Page 2, line 4)

L7. lab –> laboratory

We have change accordingly. (Page 2, line 8)

L7. There was no intensive modelling? Perhaps ’extensive’ is superfluous?

We have removed "extensive" (Page 2, line 8)

L8. Simplify (here and throughout): ’This paper presents the state...’

We have clarified the aims (Page 2, lines 8-11) based on the general comments, as described in the answers to the comments above. By doing this, the text has been simplified.

L9. You jump too quickly into the specific of the results, give the reader a more guided introduction, ’Under the project, we found that... ’

We have now started the presentation of the results with "In this study we…". (Page 2, line 11)

L10. Variation from what? Data? Methods? Remember the international audience knows nothing of the project!

We refer to the variation in estimated weathering rates from different approaches. We have now clarified that. (Page 2, line 12)

L12. Important but the manuscript would greatly benefit from the ’word smiting’ of the native English-speaking co-authors, Finlay and Bishop?

The manuscript was corrected by a professional language editor before submission! Finlay and Bishop have participated in the revision of the manuscript.

L13. I think this is an important result but the term ’clear imbalances’ obscures the implications of the findings. The activities are unstainable.

We have clarified the text accordingly: "...showed sites where whole-tree harvesting was clearly not sustainable…". (Page 2, lines 14-15)

L16. Step back and provide greater support... approaches based on the weathering of (observed) mineralogy, such as PROFILE..., provide the most important fundamental understanding of the contribution of weathering to long-term availability of base cations to support forest growth, nonetheless, these approaches should be continually assessed against...’

We changed to "Based on the research findings in the QWARTS programme, it was concluded that the PROFILE/ForSAFE family of models provides the most important fundamental understanding of the *contribution of weathering to long-term availability of base cations to support forest growth. However, these approaches should be continually assessed against other approaches." (Page 2, lines 17-20).*

L19. this point needs further development / clarity

The two last sentences were replaced with one: "Uncertainties in the model approaches can be further reduced, mainly by finding ways to reduce uncertainties in input data on soil texture and associated hydrological parameters, but also by developing the models, e.g. to better represent biological feedbacks under the influence of climate change." (Page 2, lines 20-22).

Page 3. L1. change acid to acidic throughout

We have gone through the document and changed accordingly.

L3. remove one 'processes'

We have reformulated to: "…extensive research examined processes that acidifies and counteracts acidification….". (Page 3, lines 2-3)

L5. refer to SWAP first, it started before NAPAP (and is more important in a European context)

We have changed the order of SWAP and NAPAP. (Page 3, lines 3-6)

L7. You need to provide more context for critical loads; it is an effect-based approach for emission reductions, essentially a direct response to the recognition that emissions of sulphur dioxide were causing significant impacts. Notably it has nothing to do with SWAP or NAPAP!

We changed the first sentence about critical loads to: "In the end of the 1980s, the critical load concept was developed as an effects-based approach for emissions reductions (Nilsson and Grennfelt, 1988),….". By doing that, we add info about CL being an effect-based approach for emission reductions. By starting with "In the end of the 1980s…" we discouple this sentence from the sentence about SWAP and NAPAP. (Page 3, lines 6-7)

L9. 'A critical load...'

We have changed from "The.." to "A..". (Page 3, line 9)

L11. '... critical loads of acidity...'

We have added "..of acidity". (Page 3, line 11)

L15. Yes, very true but those of us interested in water barely consider weathering directly...?

We don´t understand what R1 suggests here. We have not done any changes.

L16. '... and as such a sink of acidity'.

We have revised as suggested. (Page 3, line 17)

L19. You need to differentiate between plot and catchment scale estimates, models such as MAGIC are process-based, and can be used to provide catchment-based estimates of weathering, however, they are fundamentally different to process-based estimates from PROFILE.

In the changes of the introduction, to meet other review comments, this paragraph has been changed. Now the main content can be found on page 3, line 30-: "Therefore, a number of indirect methods to quantify weathering rates have been developed, e.g. process-based modelling (Sverdrup and Warfvinge, 1993), soil measurements where the depletion of weathering products in different soil layers is determined in order to assess average weathering rates since soil formation (Olsson et al., 1993), and budget calculations where all other parameters except weathering are measured (Lundström, 1990; Jacks and Åberg, 1987; Wickman and Jacks, 1991; Sverdrup et al., 1998)." In the way it is written now, we don´t think it is necessary to mention scales here (we don´t want to make the Introduction longer). The differences between methods are described in the Methods sections.

L24. I do not completely agree with this. Many jurisdictions were faced with national scale modelling, and the application of simple approaches such as 'skokloster' provide a practical solution compared to the application of a process-based model that requires quantitative mineralogy on a high spatial resolution... more correctly, given the high loads at that time, the uncertainty in weathering was trivial.

We changed to "The uncertainties in weathering rates were, during the times of high deposition, less important. Therefore, the interest waned in further weathering research that might revise these weathering estimates." (page 4, lines 6-8).

L29. Was it severity, or a shift in policy to support mitigation of climate change impacts?

The policies changed when the severity of climate change became fully recongnized, and it was the policy shift which led to higher demand of renewable fuel. The sentence now reads: "As the severity of climate change became fully recognised, policies for mitigation of climate change led to increased demand for renewable fuels, thereby increasing the pressure on forests." (Page 4, lines 9-10).

Page 4. L1. increase from 25

Yes it increased again after 2016, it was a temporal dip. The sentence now reads: "Since 2000 in

Sweden, the proportion of clearcuts involving whole-tree harvesting has increased from around 15% to 25-35%, according to statistics from the Swedish Forest Agency, except for 2014-2016, when the proportion was temporarily 15-25% due to lower energy prices during that period." (Page 4, lines 11-13).

L3. Substantial

It now says: "a substantially increased removal". We have not changed since we think that it is correct as it is.

L5. Depletion methods needs more description... or just exclude such detail for the moment (estimates of base cation weathering...)

The description of the Depletion method comes later. We did not do any changes.

L11. 'Akselsson et al. (2007) used a mass balance approach (with weathering estimated using PROFILE) ...'

This part of the introduction has been shortened during the revision. However, we still mention this study: "Accordingly, mass balance calculations, with weathering and deposition as inputs and harvest losses and losses through leaching as outputs (Akselsson et al., 2007) (Fig. 1), as well as simplified calculations, where weathering rates are compared with base cation losses through harvesting (Olsson et al., 1993), have been used during the last decades, for forest sustainability assessments." (Page 4, Line 19-22).

L17. I suggest 'Similarly, the influence of whole-tree harvesting ...'

This part has been removed during the revisions.

L20. Yes, but only in a Scandinavian context, this has not spilled over into the rest of Europe or north America (yet).

OK, it is interesting to think about why. We have added "in Scandinavia". (Page 4, line 25).

L23. Was the conclusion valid? I would suggest the greatest uncertainty was derived from comparing approaches that should not have been compared?

We agree that it makes no sense to compare weathering rates estimated for different soil depths. That is also what Futter et al concluded, which we write about in the next paragraph. We have not done any changes here.

L30. It is okay to call out errors. Three approaches? In truth there are two. Mass balance approaches where you indirectly estimate weathering rate (there are also other indirect methods) OR mineralogy-based approaches, often if mineralogy is not available you have surrogate-based approaches but ’at least three approaches’ may verge on ridiculous? Weathering is the breakdown of minerals... so what does three independent approaches refer to?

We are also skeptical about the conclusion about three approaches. However, we have written about that in the discussion, and base it on the results from this study. We think that the discussion is a better place to “call out errors” than the introduction.

Page 5. L1. Provide background on the depletion method... total analysis regression... the reader need help.

To not make the introduction too long, we removed the names of the methods. They are presented in the Methods section.

L3-4. reference to other methods are difficult to navigate...

Methods names removed, see above.

L7-8. Combine sentences... reduce words...

We changed to: “Two other potential sources of uncertainties that have been explored, but that are still widely discussed, were revisited: (1) the role of biological weathering that might generate weathering not included in the current generation of biogeochemical models (Banfield et al., 1999; Finlay et al., 2009; Finlay et al., in review (this issue)) and (2) simplifications relating to base cation exchange and aluminium complexation (Tipping 2002; Gustafsson et al., 2018 (this issue); van der Heijden et al., 2018).” (Page 5, lines 10-14).

L17. ’... by revisiting older w...’

This part has been removed as part of the simplification suggested by the referees.

L21. Flows or removals?

This part has been removed as part of the simplification suggested by the referees.

L30. Replace flows with ’sources and sinks’

This part is now in the Introduction (Page 3, line 29). We changed according to the suggestion.

Page 6. L2. Cite ’Warfvinge and Sverdrup, 1992’ for PROFILE

We have changed accordingly. (Page 6, line 22)

L2. The work of Susan Brantley should be cited here

We added a sentence and included a Brantley reference. We also included references to other weathering models: “Due to the difficulties in measuring field weathering rates, weathering kinetic haves been frequently studied in laboratory environments (Brantley et al., 2008). Mechanistic modelling of weathering rates, based on laboratory-determined weathering kinetics, is one of the most widely used approaches for estimating field weathering rate (Warfvinge and Sverdrup, 1992; Godderis et al., 2006; Maher et al., 2009).” (Page 6, lines 19-22).

L7. The key point here, and what separates PROFILE from other approaches, it that weathering is derived from the breakdown of mineralogy (an essential input), the other inputs only estimate the amount of minerals that are being weathered.

We included that in the first sentence: “The PROFILE model (Warfvinge and Sverdrup, 1992) is a steady state soil chemistry model where weathering is derived from the breakdown of minerals…” (Page 6, lines 22-23).

L13. Again, it might be worth citing Brantley here...

We didn´t add a reference to Brantley also here, since this part is specifically about PROFILE.

L17. hydrological model...

We have changed “hydrology” to “hydrological” (Page 7, line 6).

L21. The discussion / details on SAFE and ForSAFE can be removed.

We want to include a description of ForSAFE, as for the other approaches. However, we have reduced the description about the models, in the restructuring of the paper. SAFE is mentioned since it is still more well-known in some contexts, in order to explain how SAFE and ForSAFE relate to each other. (Page 7, line 3-)

L30. Simplify to PROFILE

This section has been removed, as part of the shortening of the paper. See answer to general comment #2.

Page 7 L4. There is an application of SAFE to Hubbard Brook which models the catchment (compared with MAGIC, VSD, etc.)

Also this section has been removed we now focus on the methods used in this study, in order to get a shorter and more focused paper, according to the referee comments.

L7. Should MAGIC be cited here or under 'mass balance' approaches? PnET-BGC is another example of a model that uses a mass-balance approach

We actually thought a lot about that. It fits in both chapters. We have thought about it again have now decided to put it under "Budget calculations" (page 9, lines 11-18). We have´t mentioned PnET BGC, since we now only include models that have contributed to weathering rates in this paper (see above).

L17. Assumes that deeper soil is the parent material, so does not work for glaciofluvial soils, etc.

We will clarify that in assumption nr 2 (Page 8, line 2): "(2) the soil pedon consists of homogeneous, where the deep soil constitutes the parent material,…"

L26. Could add that the approach has been widely used and cite a few examples?

We have done that. (Page 7, line 26-27)

L27. Typically referred to as 'Catchment mass balance budgets' as they are widely estimated at the catchment scale, as such the estimates of weathering are an average of a larger landscape unit and can be highly influenced by localised geology.

The examples in this paper is site mass balance studies. Therefore, we will not change to 'Catchment mass balance budgets'.

L28. MAGIC should really be mentioned in this section!

See answer to comment on Page 7, L7 above.

Page 8 L10. Retitle to 'strontium isotope ratio'

This chapter is now part of the budget calculation chapter, so the title is removed.

L17. This really should be included under the depletion, as it is a derivative of that approach

Yes, the total analysis regression method is a derivative of the depletion method. However, we decided to keep them separated, but clarify in the beginning that this is the case. The reason is that, in the single site comparison, the total analysis regression methods are in many cases referred to, and in one case (Gårdsjön), weathering rates have been calculated both using the depletion method, and using the total *analysis correlations. We now start the chapter about the total analysis regression with "The total analysis regression method is a derivative from the depletion method, which requires much less soil data than the depletion method." (Page 8, lines 9-11).*

L24. Remind the reader that you are focused on QWARTZ, i.e., 'Under QWARTZ, weathering ...'

The restructuring of the paper, to the more conventional division into methods and results, makes this information superfluous. Moreover, we have used data from the QWARTS programme, but even more from earlier studies. Thus, we did not do any changes.

L30. 'profile 17-20cm deeper', this is a little unclear. I assume in simple terms the soil depth differs between estimates... Maybe present table on a 'weathering per cm'?

We have made major changes in this chapter, and this part is removed. We don´t want to add a new table, since we are trying to shorten the paper, and since the depths are the same for the same site, except for in Gårdsjön and Svartberget, where we have different rows for the different subsites (with different depths).

Page 9 L2. This is an important point and should perhaps be stated much earlier, homogeneity of soil and bedrock are important considerations for agreement / lack of agreement between approaches

We think it fits good here, in the very first part of the results.

L15. Why does the depletion indicate a lower weathering rate? This could suggest that the un-weathered layer did have weathering? In many of the studies in Table 1, the depletion method is lower. Why? However, it may also be argued that the range between methods is smaller than the uncertainty?

We have some potential explanations to this in page 11, lines 24-28. One of them is in accordance with the suggestion above about that the "original till" is actually weathered till from earlier glaciations.

L21. Do you mean 'soil bulk density'?

Yes, we have clarified that (page 11, line 11).

L23. Correct 'to to'

This sentence has been removed in the restructuring and shortening of the paper.

Page 10 L19. Disqualified? Excluded! However, just exclude, and note in a footnote to the Table. No need to explain

We changed to 'excluded' and refer to Stendahl et al. instead of explaining in detail. However, we keep *it here, not in a footnote in the Table, since the sites are not part of the Table, and a footnote about three other sites feels a bit out of place. (Page 5, line 30).*

L30. Till is the most obvious / likely explanation... remove other excludes / reasons and only present this one.

We have changed the order, so that the till explanation comes first. However, we keep the others as they also might be contributory explanations (Page 11, lines 24-28).

L34. What does 'conceptual limitations' mean? clarify

We agree that this is not a good expression. We have shortened this part, and removed the "explanation", and refer to the discussion in Stendahl et al.(Page 11, lines 30-33).

Page 11 L1. 'for Ca and Mg...'

We have added 'for' (page 11, line 32).

L3 to L10. This can be reduced to one sentence, state 'The majority of weathering rate estimates were classified as acidic or intermediate (cite Table, UNECE, etc.).

We shortened the paragraph. It now reads: "The intervals of all sites were compared with four reference weathering intervals, based on weathering rate approximations frequently used in the critical load work (Fig. 3; de Vries, 1994; Umweltsbundesamt, 1996). The majority of weathering rate estimates were within or close to the interval outlined for acidic/intermediate parent material with coarse texture." (Page 12, lines 7-9).

L4. Figure 2 essentially repeats Table 2; the classification could be added to the Table 2 instead.

We think that the figure referred to, which is now Figure 3, gives a good overview, much better than the table (which is now Table 3), but we also want to share the actual numbers in the paper, so we would like to keep both.

L10 to L14. This text repeats detail already discussed.

This part has been removed in the restructuring and shortening of the paper.

L20. This is a long section, and rather than go through each site (one by one), it would be more efficient to summarise and focus on the broad agreement, and disagree, but describe from the point-of-view of the factors that drive disagreement, e.g. ' ... there was slight disagreement between some estimates owing to difference in input data use by the different approaches, such as soil depth (give example) or soil moisture (give example). The table is provided, so the reader can evaluate the results, and there is no need to describe in detail.

We have shortened section 3.1 abut site-level comparisons substantially (from three pages to just over two). We have added quantitative measures and have removed many f the details.

L22. are they scaled-up or just regional applications (more sites)? How are they scaled?

It is more regional applications (more sites). However, this formulation was removed during the restructuring of the paper.

L25 to L33. The relationship between ForSAFE and PROFILE has already been described at length. There is no need to repeat again.

This has been solved by restructuring the paper to a more conventional structure with methods, results and discussion, see more detailed description in the answer to general comment #2. The difference is now described in Methods. (Page 7, lines 15-18)

Page 12 L7. Is the analysis really only based on 11 sites? Did the study use 11 sites to predict at 400+?

Yes, this is described in Olsson et al. (1993) which we refer to.

Page 13 L5. Is this the same approach as used with PROFILE? Did both use UPPSALA? Clarify

No, another normative model was used for PROFILE. However, we have shortened the method description according to earlier comments, and do not include this information in the paper anymore, but instead refer to the papers where it has been described thoroughly, so we don´t add anything based on this comment.

L14. Again, is this similar to MATCH used in PROFILE. Perhaps have a consistent description (and only described in one place in the text)

Yes, MATCH was used for both. However, due to the shortening of the method description (see above), we don´t include this info in the paper, instead we refer to the papers where it has been described thoroughly, so we don´t add anything based on this comment.

L15. Why compare PROFILE and ForSAFE. Are they dramatically different, or are you just comparing the effect of different hydrological data on estimates of weathering? The justification for this needs to be clearly stated under section 4. This is a very different comparison to total analysis (which is fundamentally a different estimate of weathering).

We agree that the comparison is very different than a comparison with to completely different approaches. PROFILE and ForSAFE are built on the same weathering reactions, but the dynamics in ForSAFE affects weathering rates. Also, the fact that e.g. hydrology is modelled in ForSAFE, whereas it is input data in PROFILE, may affect weathering rates. This is described thoroughly in Kronnäs et al. in the same issue. We have now clarified this in the Methods chapter, page 7, line 15-19: "Although the weathering module is the same in PROFILE and ForSAFE, some differences can be expected since ForSAFE includes dynamics, which means that weathering is affected by other processes over time, and that soil moisture, which is an input in PROFILE, is dynamically modelled in ForSAFE."

L16 to L18. This detail should be presented under the main part of section 4

This part has been completely changed and shortened in the restructuring of the paper. The information about methodology for regional runs can be found in Methods, on Page 7, lines 20-24.

L19. Why use climate regions?

We want to show regional differences, and those regions are logical in the way that they represent different climate and atmospheric deposition regimes. They have been used in one paper, just accepted for publication (Belyazid & Zanchi, 2019), and are used also in Belyazid et al (this issue).

L24. Above you have noted that the differences in estimates of weathering is often driven by differences in inputs (under different applications) for the same model. Here you add further confusion to that issue... What is the goal of the comparison?

The goal of the comparison was not to run different approaches on many sites with exactly the same input data. The goal was to collect published weathering estimates from different approaches, in many cases run independently of each other, and compare, to get a span covering both differences in approaches and differences in input data used. This represents the reality for e.g stakeholders. We show that, even though we do like this, we can draw conclusions about sustainability. This is clarified in our new first aim: "(1) investigate the variation in published weathering rates from different approaches in Sweden, under consideration of the key uncertainties for each method,"

L29. Above you state they are more-or-less the same, and since that statement we find that one is higher than the other, and so on... which is the truth?

We wrote that there were "no large systematic differences between the medians or ranges for the *different methods". And that "ForSAFE gave somewhat higher medians than PROFILE for all regions, especially in the northern regions." We don´t think that it is contradictory, it is still no large systematic differences. However, we agree that it could be better formulated Now we start the chapter about regional level comparisons with: "The weathering rates for the nutrient base cations Ca, Mg and K varied widely within the regions for all methods, but there were no large systematic differences between the medians or ranges for the different methods (Fig. 4-5). However, PROFILE gave generally somewhat lower weathering rates than ForSAFE and the Depletion method/Total analysis regression approach, with overall medians of 14.3 mekv* m^{-2} y^{-1} *(PROFILE) and 17.8 mekv* m^{-2} y^{-1} *(ForSAFE and the Depletion method/Total analysis regression approach). " (Page 12, lines 15-19).*

L33. Maximum weathering depth? This is confusing, why compare if they represent different depths / pools? This needs clarification, the text suggests that PROFILE covers a shallower depth compared with Total Analysis. They why compare? Normalise both to the same depth before comparing.

In this paper we have used results already published. We have decided not to try to normalize them, then we would add extra assumptions (e.g that weathering per cm is constant in the soil profile, which we know is not true). We know e.g. that the weathering rates according to the depletion method are much slower further down in the soil profile. We have thus decided to discuss the differences in depths instead of normalizing.

Page14 L6. Reword... what comparison between regions?

Here we mean the comparison of weathering rate intervals between regions. The sentence now reads: "In most cases, the width of the weathering rate intervals showed no major differences between the regions." (Page 13, line 5)

L8-L9. This statement, and similarly many of the statements in the previous section, are very qualitative. There is no quantitative element to the comparison at the site or regional level. Statements such as broadly agree, similar / non-similar are fine IF they are also supported by a quantitative assessment. This is missing.

We have now quantified differences in the chapter 3.2 Weathering rate comparisons on a regional level. For example, we compare medians between the approaches and regions. See also answer to general comment #4, regarding similar changes in other parts of the paper.

L10+. This has been already stated, and is obvious, it should be noted in the main part of section 4, and not included as part of the comparison (it was known before starting the comparison). However, it would be useful to know the purpose for such a comparison?

See answer to comment regarding page 13, L15.

L15. I think (more-or-less) the results of this assessment are stated here, as such, perhaps the whole assessment could be collapsed to one paragraph?

We have substantially shortened chapter 3.2, as part of the restructuring and shortening of the paper.

L27. If it is already described, then why repeat here?

We have substantially shortened the chapter about biological weathering, focusing on implications of insufficient process descriptions for modelling results, in line with the comments from Referees and Editor.

L30. wording ’dependent’

Chapter 3.4 about Biological weathering has been very much shortened and this formulation has been removed.

Page 15. L1-L5. Citations?

We added the reference Sverdrup and Warfvinge, 1993, where this is gone through more thoroughly (Page 15, line 11).

L11. The weathering process in safe is not directly affected by biological processes, it is only affected in as far as the recognition that some of the processes are likely influenced by biology.

Biological processes do affect weathering in PROFILE, SAFE and ForSAFE through the following pathways: 1- uptake reduces cation concentrations, directly alleviating the brakes on weathering, 2- transpiration reduces water availability, thus limiting one of the four weathering kinetics (dissolution in water), and increasing concentrations, which can have positive (in case of acids) or negative (in case of cations) effects on weathering, 3- cation uptake produces more protons in the soil solution, lowering pH and promoting weathering, and 4- the production of dissolved organic radicals through litterfall decomposition and in the case of ForSAFE exudation contribute directly to one of the four weathering kinetics. Through these four pathways, the models do directly modify weathering rates based on the outcome of biological activity as described here.

L12. PROFILE has some biological feedback? Really?

See answer to L11.

L33. ’There is extensive literature...’

This part has been removed when the chapter about biological weathering was shortened.

Page 16. L1 to L34. The entire page (and some of the previous) presents a good review of the ’state of knowledge’ but it can be much reduced... and the benefit / objective of such a review should be considered... why cover so much text if this if not part of the work under QWARTS described by the authors.

We have shortened this chapter substantially, see answer to general comment ¤1.

Page 17. L24. ’We found’? Which ’we’?

This part has been removed when the chapter about biological weathering was shortened.

Page 18. L4. Was this stated already?

We have shortened the chapter substantially and got rid of repetitious text.

L10. What does the section title mean?

We changed the title to “Implications of improved model descriptions of base cation exchange and aluminium complexation”, which is similar to how it was phrased in the Introduction (Page 16, line 19).

L17.Why ’state-of-the-art’. WHAM has been around for decades?

We removed ‘state of the art’. (Page 16, lines 26-27).

L22. Replace ’former’ with specific term.

We did that: “…but for a long time, the simpler ion-exchange equations have been more widely used…” (Page 16, lines 31-32).

Page 19 L10. I wonder if this is some of the context for this manuscript that would be better to present at the start?

In the restructuring, the chapter about Weathering in a sustainability perspective is focused on the results from the sustainability assessments. The first part where it was introduced has now been moved to the Introduction. (See e.g Page 4, line 16-).

L20. ’In regions where weathering rates...’

This part has been removed in the restructuring and shortening of the paper.

L23. Clarify or reword ’data on site index could be found’

We changed the word “site index” to “site quality” This part has been reformulated in the restructuring and shortening of the text. (Page 9, lines 30-34).

Page 20 L7. Differences in weathering has already been discussed?

We have shortened the chapter about Weathering in a sustainability perspective, but we need to mention the differences also here, since the sites with large differences are the ones where it is more difficult to draw conclusions about sustainability.

L10. This sounds like it was stated already?

No, this is the first time we compare weathering and WTH in the north. However, after the restructuring and shortening of the text, it is hopefully more easy to follow.

L21. The preceding text could be summarised much more succinctly.

The text has been restructured, so this chapter now only includes results, whereas the methods have been moved to the “Methods” chapter, and the first part to the introduction.

L31. This is more-or-less the summary of the results (if a further quantitative description was added) then this would be sufficient.

We have shortened this chapter and added quantitative measures.

Page 21. L12 to L18. This has nothing to do with future, It is mostly repetitious text.

See answer to general comment# 3.

L22. This section seems odd... we have just been presented with a 2+ page section that covered biological weathering. It is difficult to justify this additional text! I believe this section should be removed, and any ’fresh’ text be included above.

We have shortened the chapter about biological weathering substantially and we have replaced the chapter “future research”, with “Prospects for method development”. The first subchapter in “Prospects for method development” is about “PROFILE/ForSAFE – Development of model descriptions” There we included one sentence about improvements related to biological weathering (Page 18, line 2-7).

L23. Yet despite the previous lengthy discussion on biological weathering, we were not introduced to the term ’EPS’??

We have now introduced the term extracellular polysaccharides (EPS) (Page 15, lines 23-24)

L23 to L4 Page 22. This text can be deleted.

This chapter has been merged with the chapter about Potential for biological weathering, and the text in those chapters has been very much shortened.

Page 22 L5 to L11. Is this model development or uncertainties?

In the restructuring about the Biological weathering part, this chapter has been removed, and we now only include one model development sentence about biological weathering in the chapter “Prospects for method development”, that has replaced “Future research”.

L24. Again, it is difficult to justify such an extension section, shortly after we have already been presented with a discussion on the topic.

In the restructuring of the paper, this chapter was removed, and we now only include one short paragraph about this in the chapter “Prospects for method development”.

L25 to L32. This is not model development...just repetitious text

See answer to Page 22, L24 above.

Page 23 L1. This is a trivial point, with the right implementation the sped can improve. Just because it takes an hour to cycle to work, does not mean that everyone must cycle!

The organic complexation does not have an arithmetic solution, the only way to do it without compromising its purpose is by optimization (of multiple complexation parameters) and a whole set of assumptions (including that the Al pool is constant for example), i.e. iterations until near-equilibrium. In HD-Minteq this is feasible because most state variables and fluxes (soil organic matter, decomposition, uptake…) are given as input and therefore not dependent on changes in soil solution chemistry. While in ForSAFE these processes are internally simulated, meaning that each iteration of Al complexation will require an entire simulation of all fluxes and equilibria, exponentially increasing computation time or even requiring a new optimization of exchange parameters, which in the word of the original author “is impossible” (see Gustafsson 2001, Journal of Colloid and Interface Science 244, 102-112. doi:10.1006/jcis.2001.7871).

L8 to L11. I am sure this is repetitious text.

This text was removed.

L14. ’ overestimated estimates of weathering rates’ –> ’ overestimated rates of weathering’

This was changed accordingly (page 18, line 15)

L19.So it was not tested? Is this future research? It seems to be more of ’ongoing’ research?

New brakes have not been implemented and tested in PROFILE/ForSAFE. This is future work. In Erlandsson et al., we implemented and tested silicate release through weathering and the feedback brakes from Si concentrations on the four weathering kinetic equations, while dictating other inputs such as uptake, water flow and so on. This has produce the expected results (constraining weathering in the saturated zone). Implementation in PROFILE/ForSAFE has not yet been carried out. The chapter “Future research” is now replaced with the chapter “Prospects for method development”.

L20. Is this catchment scale weathering?

Yes, however this heading has been removed.

L21 to L27. This is not future research, this is improvements needed in the application of weathering models (and better linkages with hydrological models). As such, uncertainties or limitations is a more appropriate section.

We thought about, but didn´t think that “Uncertainties” was suitable. We want this chapter to include advices based on the QWARTS program, about how to continue to reduce uncertainties. We have renamed the chapter: “Prospects for method development”, which we think fits with what we want to include. We have also removed or moved parts that do not fit in.

L28 to L4 Page 24. Not future... ongoing /current research?

See answer to comment L21-L27 above.

Page 24 L5. See comments above... much of the text presented so far under ’future research’ speaks more to limitations in application or uncertainties.

See answer to comment L21-L27 above.

L6. This is PROFILE specific text... not the depletion method, or others...

The chapter is now called: “PROFILE/ForSAFE – Improving input data quality”

L8. Often?

This sentence has been removed in the shortening and restructuring of the paper.

L9. ’Not only are...’

We corrected this (Page 19, line 5).

L13 to L20. This is a very uncertain uncertainty... why so much space for something that is not ’very uncertain’?

It is part of the QWARTS work and refers to one of the papers in the special issue. So we want to refer to it, but we have shortened the paragraph (Page 19, lines 6-15).

L24. Unless that span is used to estimate the uncertainty...

It is used for that in one of the papers of the special issue (Casetou Gustafson et al.). But still, often one value is used.

L26. This does not make sense. Is it possible to contain the solution space?

The reviewer is right that this is not a problem if we have information on which minerals are present, their respective stoichimetries and in what proportions (See Posch and Kurz, 2007). This however is most often missing, and that is the purpose of using A2M. It would, as indicated, be possible to narrow the space in more information is available, on for example the fraction of light primary minerals in the soil. So yes, it is possible to constrain the space if a considerably more information is available.

L28. This is not true. However, many users make that assumption. However, others do not.

In the cases we have seen, where one of the mineralogies is chosen from A2M, it has been the centre point, although everyone who uses A2M should know that all solutions are as likely. We think that we have been clear about that all have the same probability, but that many choose the center point, since they want one value.

Page 25. L1. BET may still be the best technology?

We removed “using modern technology”. Even if the BET technology hasn´t developed since the 80/90:ies (which we don´t know), the regressions could be revised based on a larger soil material (Page 19, lines 26-28).

L2. Are the ’current uncertainties (?)’ quantified? Are there uncertainties?

The regression graphs in Warfvinge and Sverdrup (1995) indicate large uncertainties. On page 19, *lines 27-28, we now write: "The regressions reveal that the uncertainties are large. Revisions of the regressions, based on a larger data material, could reduce the uncertainties."*

L5 to L9. Repetitious text.

This is now removed from here, instead we focus on the need for better soil moisture quantification in the models: "The soil moisture is one of the most important factors that introduces large uncertainties in the results, both in PROFILE where it is an input (Rapp and Bishop, 2003), and in ForSAFE where it is modelled based on hydrological parameters (Kronnäs et al., 2019). Improved input data quality for soil moisture would substantially reduce uncertainties in PROFILE and, even more importantly, soil moisture modelled by ForSAFE needs to be evaluated, and the sensitivity to soil input data needs to be examined." (Page 19, lines 29-33).

L10. Improved soil moisture should come with improved soil hydrological modelling...

Yes, and since this chapter is about improved input data we mention soil data which is very important for the hydrology modelling in ForSAFE. We don´t understand what R1 suggests here.

L12. All estimates are modelled? What are the other?

This chapter was removed in the restricting and shortening of the text.

L13 to L17 Page 26. This whole section can be deleted. Any useful should be moved to the section on comparisons. This is not 'future research'

This chapter was removed in the restricting and shortening of the text.

L23. Manual? This is a bit trivial... delete and move to personal 'to do' list. I suggest you write a paper on this.

We don´t mention to propose a manual any more.

L28. They were not outliers. They represent measurements for different compartments. This is well understood.

Mass balance methods are still used for weathering rate calculations and advocated by many. We included it here and came to the conclusion that it was problematic in the cases we had. We write "In the compilation of weathering rates in this paper, the most extreme outliers came from the budget method, which can be explained by the fact that the sources and sinks are not described in a completely accurate way (Rosenstock et al., in review (this issue)). For a fair comparison between weathering rates *from the budget method and from other methods, ways to distinguish between different sources and sinks need to be further developed. We think that this is a rather strong recommendation, which we would like to keep as it is.*

Page 26. L10. This is wrong. It is okay to state that. There independent methods? How many methods are there (truly independent)? More correctly, Futter et al. (2012) should have recommended that a method incorporating soil mineralogy be used (all other approaches are surrogates for weathering).

We think that we are rather clear when we conclude that it is unrealistic. We think that it is stronger to say that, than to say that is absurd or wrong, which are more "value" words.

L16. Good but you can more clearly call it out as an absurd suggestion.

We think it is enough to say that it is unrealistic in practice.

L22. Was some of this difference on single sites driven by differences in depths / inputs?

Yes. We have changed to "Although the variation in weathering estimates was large on single sites…". By doing that we include methods as well as input data and to some extent depths. (Page 20, lines 30-31).

L21 to L30. I agree that these are the primary conclusions from this work; I would urge the authors to reflect on this when revising the manuscript. Much of the text can be reduced and streamline to better present this issue (conclusion).

We agree and our revisions have been done with this in mind, see further answer to general comment # 1-3.

Page 27. L10. Other approaches?

Yes, the last sentence in the Conclusions now reads: "However, it is also important to continue to compare with results from the Depletion method and the Budget approach."

Answers to comments by Referee 2 (R2)

Comments from 19 March 2019

R2 starts off with a paragraph with general comments, describing the paper and its strengths. In this paragraph there are no questions or suggestions of changes. After the general comments, a few specific comments are listed.

Specific comments

1a. "The consistency of the weathering rates estimated by different methods at the same sites in this study was remarkable. Older studies frequently reported one or two order of magnitude differences in estimates of weathering by depletion versus budgets………… Have weathering rates changed substantially in the last 30 years, or were the differences always muted in Swedish soils?"

The main aim of this study was to investigate the variation in weathering rates from different approaches in Sweden, and assess the results from a sustainability perspective. To do that, we searched for Swedish sites where weathering rates had been estimated with more than one method, to the same depth, and where the method and data was well described. We didn´t exclude any sites where those criteria was met, but included all old and new studies that we could find in literature. We did not find any sites with differences of one or two order of magnitudes between weathering rates estimated with the depletion method and the budget method, which according to R2 was frequently reported in older studies. We didn´t find those differences when comparing other methods either. We pointed that out in our first answer to the review comments, and then got another interpretation of the question than our own from the Associate editor. We answer her comments under the heading "Answer to Associate Editor Decision". Regarding the question from R2 about whether weathering rates have changed substantially in the last 30 year, we don´t think that, and the dynamic modelling with the ForSAFE model by Kronnäs et al. (this issue), does not indicate any large changes.

1b. "Following upon the previous comment, have the authors tried plotting the estimated weathering rates against one another? For example, PROFILE vs. Depletion, etc.? Perhaps this is in one of the other papers in the series?"

In Stendahl et al. (2013) weathering rates from the depletion method was plotted against PROFILE weathering rates on 16 sites. The number of sites in that study is enough to be able to draw general conclusions about differences in weathering rates from the two different methods. In the present study, those sites are included, but also other sites, based on different combinations of methods. Based on the material in this paper, it is not as straight forward to do complementary pairwise comparisons, since most of the approaches (all except PROFILE and the depletion method that are already compared) are performed only on a few sites. Instead we have focused on presenting the weathering rate intervals for each site, to illustrate how much they differ.

3. "When comparing weathering rates to harvesting removals, what rotation length was assumed to calculate a meq/m2/yr value?"

We use site quality to calculate harvesting removals, in the same way as in Akselsson et al. (2007: 2016) and Stendahl et al. (2013), which we refer to. Site quality (in Swedish "bonitet") is the average yearly biomass growth on a specific place during a forest rotation, if the forest is managed optimally. In our calculations we assume, like in earlier studies, that the actual growth is 80% of the optimal growth. The average growth in the site quality concept "assumes " that the forest is harvested at the optimal point in time, which varies between north and south and between stands, from about 70 years in the south to 120 years in the north. We have now explained more thoroughly how the harvesting losses were estimated (Page 9, lines 32-34).

4. "An awful lot of confidence is placed in this paper in modeling approaches in general and in the PROFILE/ForSAFE family of models in particular They are good models conceptually and they produce results that appear to track the results from other methods (but see comment 1b above). The problem is that there are no "measured" values of weathering flux to use to validate these (or any) weathering models. So, just as budget-based approaches may be contaminated by non-weathering fluxes like net losses from exchange sites, and depletion approaches may suffer from invalid assumptions,

model results almost certainly contain a host of errors. This is addressed somewhat in the paper. Depletion methods and budget methods are based on field observations and data. With all their flaws, they are, at least in my view, fundamentally stronger than model results. To compare them as equivalent approaches is problematic."

We agree that the depletion method and the budget method, that are based on measurements, are very important in framing weathering rates, which we also point out in the paper. However, we don´t think that the results from those methods are fundamentally stronger than the model results (which actually also are based on measurements to a large extent, e.g. laboratory measurements). On the contrary, we think that further studies are required to get more robust results from those methods – something that we point to in the "Prospects for method development" chapter. In this paper we have tried to show how far we can go with the available publications. In the revised version, we have a chapter called "Prospects for method development" on page 17-20 (modified from the chapter "Future research" in the old version). There we discuss uncertainties for all methods, and how to reduce those uncertainties.

Regarding budget calculations our results show that the most extreme outliers came from the budget method. We explain that by the fact that other sources than weathering are included as discussed in Rosenstock et al. (in review, this issue), as well as the documented uncertainty in terms used in the mass balances. We conclude that "for a fair comparison between weathering rates from the budget method and from other methods, ways to distinguish between different sources and sinks need to be further developed."

The depletion method gave unrealistically low weathering rates in some cases. On one of the sites, Asa, that was studied in detail within another paper in this special issue (Casetou Gustafson et al., in review, this issue), the analysis of the soil profile indicated that the soil profile has been disturbed, introducing errors in the weathering estimates. We highlight the importance of excluding sites with disturbed profiles as well as to perform studies where the average weathering rate since the last glaciation is related to the present weathering rates are required, to be able to make necessary adjustments of the historical rates.

Answers to comments by the Editor, Professor Suzanne Anderson

Comments from 19 March 2019

General comments

"Overall, the manuscript is well written and clear, although several sections are longer and a bit more tedious than others. In particular section 5 on "Potential for biological weathering", and section 8 on "Future research" could be streamlined."

We have reworked the chapter "Potential for biological weathering", according to the descriptions in the "Answer to comments by Referee 1". The chapter "Future research" overlapped to some extent with the chapters "Potential for biological weathering" and "Implication of higher resolution of chemical reactions in weathering modelling" and has now been restructured, renamed ("Prospects for method development"), and shortened from more than 5 to just above 3 pages. The paper has also been restructured to follow a more traditional outline (with methods, results and discussion), see further Answers to comments by Referee 1.

"The challenge addressed in the manuscript is whether the losses of nutrients due to forest harvest can sustainably be balanced by release of nutrients by chemical weathering. This simple mass balance could be illustrated with a conceptual diagram showing the relevant fluxes at the beginning of the manuscript. Such a diagram might help focus on the important sources of uncertainty in long-term predictions of sustainability."

We have added a figure (Fig. 1) in the Introduction, showing the relevant fluxes in the simple mass balance of base cations in the forest ecosystem (weathering, deposition, harvest losses and leaching), to put our weathering results and the sustainability assessments in a context. We introduce the Figure in the Introduction.

"The most important question the manuscript, and indeed the QWARTS program addressed is if weathering rates, as computed by different models, are greater than or less than rates of harvest loss. The range of weathering values from different models gave a sense of uncertainty in weathering rates, but harvest losses were presented as a single value at each site. There must be uncertainty in export losses due to harvesting, and it seems important to convey what these uncertainties are."

We agree that the sustainability of harvesting is an important part of this paper, and that the uncertainties in the harvesting estimates should be mentioned. Research related to those uncertainties were not included in QWARTS, which focused on the actual weathering rates, but we have added a paragraph about the uncertainties in the chapter "Weathering in a sustainability perspective", based on results from other studies, e.g. Zetterberg et al. (2014) in Science of the Total Environment. (Page 14, lines 14-28).

"Two aspects of weathering that I would have expected to be important (and that might be highlighted in the conceptual diagram I suggest above) are rock type and depth of weathering. The first of these is perhaps simpler to consider. Rock type or soil substrate was not described anywhere or for any site. I could not tell if this was because all of Sweden has the same rock type, and so is dismissed as playing any role in variations in weathering rate or nutrient release, or if something else was going on. I would expect rock type to be the very first control on weathering rate, as nutrient availability on basalt vs limestone vs serpentinite vs : : :. is quite different. The depth of weathering is a more difficult problem to address, yet could also be important. Weathering often occurs at depths below the top 30-50 cm."

Yes parent material is of key importance for weathering rates. We have included a table (Table 2) describing the mineralogy used as input in the modelling for each sites, as a measure of the parent material. For the sites in which some of the authors have been involved in the weathering estimations, we have thorough databases from which we can get the information. For the other sites, we have extracted information from published papers. We refer to Table 2 at Page 5, line 29.

We have focused on the root zone in this study, since we are interested in the sustainability of forestry (clarified on Page 3, lines 23-24). Therefore, for this study we see the depth of the rooting zone as more interesting that the weathering depth, therefore we haven´t discussed the weathering depth.

In Swedish applications the depth 50 cm is often used for the rooting zone, based on studies in Sweden. In this paper, where one important aim is to compare different approaches, we have included sites where weathering rates have been calculated to deeper depths, up to 1 m, but we have only accepted sites where the different approaches have been run to the same depths. In the sustainability calculations only sites with depths<0.7 m are included. This is described in the Methods part (Page 9, lines 28-30).

We address the uncertainties related to a fixed root depth, with reference e.g. to Hodge, as suggested in the Associate editor decision (Page 14, lines 17-19).

A few detailed comments:

Acronyms should be defined at their first use. Several are not defined at all, or only after their first use (this list may not be exhaustive): UNECE, CLTRAP, EMF, A2M

We defined UNECE and CLRTAP the first time it was mentioned. We skipped the acronym EMF, and instead wrote Ectomycorrhizal fungi on the places were the acronym was used before. For A2M we did not do any changes. It is the model name, and when we introduce it we explain what it does and give a reference (just like for PROFILE and ForSAFE).

Whole-tree harvesting is defined on p3 as being “harvesting of branches”. Does this imply that stems are not included in whole-tree harvesting? This seems contradictory.

Stems are included in whole-tree harvesting. We have gone through the paper and clarified that, where required.

Total-regression analysis: what is the temperature sum?

The temperature sum is a measure often used in forestry. It is the daily mean temperature above a threshold value, in Sweden often +5ºC, summed during the growing season. Before we only referred to Morén and Perttu, now we added an explanation (Page 8, lines 13-14).

Figure 5: The boxes in the key are so small that they are indistinguishable. The gray patterns all look very similar.

This figure (which is now Fig. 6) has been improved, by increasing the size of the legend and changing the patterns so that they can be more easily distinguished from each others. Fig. 7 has been changed in the same way.

p 8, line 5: Casetou-Gustafson’s name is incomplete.

Corrected, on all places in the document.

p10, line 4: cation, not carion

Corrected, in the revised document it is on page 10, line 23.

p 12, line 9: Ca, Mg and K at 640 sites, not Ca, Mg and K on 640 sites

Corrected (although the exact formulation has changes in the revised version), page 8, line 22.

p. 17, line 2: to sparse grass, not via sparse grass

The whole paragraph has been removed, to shorten the chapter about biological weathering.

p. 17, line 2: naturally lead-contaminated, not natural, lead-contaminated.

The whole paragraph has been removed, to shorten the chapter about biological weathering.

p. 18, line 23: However, in 1996..., not However, already in 1996..

This has been changed according to the suggestion, see p. 16, line 32.

Answer to Decision by Associate Editor (Professor Suzanne Anderson)

Decision from 10 May 2019

Along with her decision about major revisions, added a few comments, which we reply to here.

1. "The proposed strategy of reorganizing the manuscript along more traditional "methods" and "results" lines seems useful, especially as it will reduce redundancies in the text. I look forward to seeing a new conceptual diagram in the introduction to further aid in this streamlining."
We reorganized the paper accordingly, which reduced redundancies substantially shortened the text substantially, from 37 pages to 29 pages (including first page and references, but not figures and tables). We also added a new conceptual picture, Figure 1, based on the suggestion, and referred to it in the Introduction, to create a better basis for the sustainability assessments.

"Referee 2's comment 1, expressing surprise at the remarkable consistency in weathering rates between methods reported in this manuscript is I believe a comment aimed at what is commonly referred to as the "field-lab discrepancy" in weathering rates. For a review of methods of measuring weathering rates, and a short discussion of this controversy, I recommend reading **Riebe et al. (2017)** Earth Surface Processes and Landforms (doi: 10.1002/esp.4052), in particular p. 133-135. I do suggest that this comment be addressed, as it is relevant. This topic also applies to Referee 2's comment 4."

We interpreted Referee 2´s comment 1 about "one or two order of magnitude differences in estimates of weathering by depletion versus budgets" in "older studies" as a comment directed towards the depletion method and the budget method. Here it is instead suggested that it addresses the "field-lab discrepancy", discussed e.g. in Riebe et al. (2017). We find it difficult to go in to a discussion about that in this paper, since the aim with the paper was to investigate the variation in weathering rates from different approaches in Sweden, and assess the results from a sustainability perspective, not to describe the development of each method and discuss them in detail. We included all Swedish sites that we could find in literature, old and new, that met our criteria (that at least two approaches for estimating *weathering rates have been applied, that the estimates have been done to the same depths, and that data and methods are well described). We don't think that discussions about things not related to the results from the analysis of weathering rates from those sites fit in the paper.*

Regarding the inclusion of lab data in PROFILE: It is described in books and papers, e.g. "Sverdrup (1990): The Kinetics of Chemical Weathering. Lund University Press, Lund, Sweden and Chartwell-Bratt Ltd, London, ISBN 0-86238-247-5" and "Sverdrup, H., Warfvinge, P. 1993. Calculating field weathering rates using a mechanistic geochemical model–PROFILE. Journal of Applied Geochemistry, 8:273–283." In the first, the experiments for different minerals are reviewed, evaluated and discussed, and the use of them in PROFILE is described. In the latter, the field-lab discrepancy is highlighted, starting with the sentence: "The rate coefficients published here are sometimes different from rate coefficient values found in the literature due to the division of the rate between different reactions, and the consistent determination of the exposed surface area." We think that a discussion about that is outside the scope of the paper.

"Finally, on the focus on a narrow rooting zone in this study (one of my comments), I think a comment on the assumption inherent in this focus is worthwhile. Root systems respond to nutrient availability, so if the 30-50 cm depth is depleted, they may grow deeper. The question is to what extent root systems are static versus plastic. You might want to at least acknowledge that thinking of the depth of nutrient access by roots as a static parameter is an assumption. See Hodge in doi:10.1016/B978-0-12-409548-9.05232-5

We clarified that we work with the rooting zone in the introduction (Page 3, lines 23-24), and we added a discussion about uncertainties related to the assumption of a specific rooting depth, with references, e.g. to Hodge, in the chapter "Weathering in a sustainability perspective" (Page 14, lines 16-19).

Weathering rates in Swedish forest soils

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Abstract. Soil and water acidification was ~~first~~ internationally recognised as a severe environmental problem in the ~~1970s~~late 1960’s. The interest in establishing ‘critical loads’ led to a peak in weathering research in the 1980s and 1990s, since base cation weathering is the long-term counterbalance to acidification pressure. Assessments of weathering rates and associated uncertainties have recently become an area of renewed research interest, this time due to demand for ~~more harvest~~forest residues to provide renewable bioenergy. Increased demand for forest fuels increases the risk of depleting the soils of base cations produced in situ by weathering. This is the background to the research programme ‘Quantifying Weathering Rates for Sustainable Forestry’ (QWARTS), which ran from 2012 to 2019. The programme involved research groups working at different scales, from laboratory experiments to ~~extensive~~ modelling. The aims of this study were to (1) investigate the variation in published weathering rates of base cations from different approaches in Sweden, with consideration of the key uncertainties for each method, (2) assess the robustness of the results in relation to sustainable forestry and (3) discuss the results in relation to new insights from the QWARTS programme, and propose ways to further reduce uncertainties~~The aims of this paper are to summarise the state of knowledge about weathering rates in Swedish forest soils at different scales, with an emphasis on the knowledge added by the QWARTS programme, to discuss the uncertainties in relation to sustainable forestry, and to highlight knowledge gaps where further research is needed~~. In the study we found that t~~T~~he variation in estimated weathering rates at single-site level was large, but still most sites could be placed reliably in broader classes of weathering rates. At the regional ~~to national~~ level, the results from the different approaches were in general agreement. Comparisons with~~of~~ base cation losses after stem-only and whole-tree harvesting showed sites ~~with clear imbalances between weathering supply and harvest losses~~where whole-tree harvesting was clearly not sustainable, and other sites where variation in weathering rates from different approaches obscured the overall balance. Clear imbalances appeared mainly after whole-tree harvesting in spruce forests in southern and central Sweden. Based on the r~~R~~esearch findings in the QWARTS programme, it was concluded that ~~support the continued use of the~~the PROFILE/ForSAFE family of models provides the most important fundamental understanding of the contribution of weathering to long-term availability of base cations to support forest growth. However, these approaches should be continually assessed against~~, but it is important to continue comparisons between these and~~ other approaches. Uncertainties in the model approaches can be further reduced, mainly by finding ways to reduce uncertainties in input data on soil texture and associated hydrological parameters, but also by developing the models, e.g., to better represent biological feedbacks under the influence of climate change~~Uncertainties in the model approaches can be further reduced, mainly by finding ways to reduce uncertainties in input data on soil texture and associated hydrological parameters. Another way to reduce uncertainties is by developing the models to better represent the delivery of weathering products to runoff waters and biological feedbacks under the influence of climate change~~.

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1 Introduction

Acidification of soils and water, caused by long-range transport of acidic compounds, was recognised as an environmental problem in Europe in the late 1960s (Odén, 1968). In subsequent decades, extensive research examined ~~acidifying~~ processes that acidifies and counteracts acidification~~processes and processes counteracting acidification~~ (Reuss and Johnson, 1986). Two key research programmes were the Surface Water Acidification Programme (1985-1990, Mason, 1990) funded by the UK (GBP 5 million) and the National Acid Precipitation Assessment Program (1980-90, Irving, 1991) funded by the US government (USD 17 million).~~ and~~ In the end of the 1980s, ~~the Surface Water Acidification Programme (1985-1990, Mason, 1990) funded by the UK (GBP 5 million, Mason, 1990).~~

t~~T~~he critical load concept~~, in which long-term weathering of base cations (Ca, Mg, Na and K) is a key parameter,~~ was developed as an effect-based approach for emission reductions (Nilsson and Grennfelt, 1988), and served as a link between science and policy within the framework of the UNECE - CLRTAP (The United Nations Economic Commission for Europe - ~~UNECE~~ Convention on Long-Range Transport of Air Pollutants~~-~~) (Lidskog and Sundqvist, 2002). ~~The~~ A critical load is defined as “a quantitative estimate of an exposure to one or more pollutants below which significant harmful effects on specified elements do not occur according to present knowledge” (UNECE, 1994). To calculate critical loads of acidity and their exceedance, mass balance calculations of acidity are used together with a critical limit for a chemical criterion, defining the maximum acidity of soil/runoff water that can be allowed without a risk of negative effects on a chosen biological indicator (Sverdrup and de Vries, 1994).

E~~Weathering e~~stimates of base cation weathering (Ca, Mg, K, Na) play a key role in all kinds of mass balance calculations related to acidity, e.g. the mentioned critical load calculations, as weathering is an important long-term natural source of base cations and as such a sink of acidity. The net accumulation or depletion of soil base cations in the soil is the result of the mass balance between inputs (atmospheric deposition and weathering) and outputs (losses through leaching and harvesting) of base cations (Fig. 1). Atmospheric deposition depends on external factors and can vary in time (Hedin et al., 1994). The leaching term is directly dependent on the mass balance as it mirrors the aqueous pool of base cations in the soil. Harvest losses are predefined following forest management. Finally, weathering is the long-term source of base cations, depending largely on soil mineral content and soil texture, which in areas that have been covered by ice, like Scandinavia, is the result of the composition of the parent material from bedrock and the glacial transport of material. When trees and forest soils are in focus, the weathering term refers to the weathering products in the rooting zone, i.e. the base cations available for trees. Due to its central role in mass balance and acidity calculations, w~~W~~eathering was ~~therefore~~ studied extensively, to enable accurate weathering quantifications. Weathering rates are, however, difficult to quantify through direct measurements in the field due to the complexity of base cation dynamics in soil. There are several different pools of base cations in soil, and also several different flows, e.g. decomposition, uptake, ion exchange and weathering, and it is difficult to distinguish between these different sources and sinks (Rosenstock et al., in review) and to define the pools accurately (van der Heijden et al.,

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2018). Therefore, a number of indirect methods to quantify weathering rates have been developed~~Various methods were used~~, e.g. process-based modelling (Sverdrup and Warfvinge, 1993), soil measurements where the depletion of weathering products in different soil layers is determined in order to assess average weathering rates since soil formation~~, i.e. the last ice age~~ (Olsson et al., 1993), and budget calculations where the~~all~~ flows in the mass balance,~~other parameters~~ except weathering, are measured (Lundström, 1990; Jacks and Åberg, 1987; Wickman and Jacks, 1991; Sverdrup et al., 1998).

The political and scientific agreement on the critical load concept as a basis for managing acidic deposition was a major factor in subsequent policy success on limiting acidifying emissions (Lidskog and Sundqvist, 2002). Major acidic deposition reductions occurred, and some recovery of soils and surface waters has been noted by the monitoring operations put in place by the UNECE CLRTAP (Graf Pannatier et al., 2011; Pihl Karlsson et al., 2011; Akselsson et al., 2013). The uncertainties in weathering rates were, during the times of high deposition, less important. Therefore~~However, one side-effect of the success in agreeing on specific weathering model estimates as the basis for long-term policy agreements to reduce atmospheric emissions~~, the ~~was that~~ interest waned in further weathering research that might revise these weathering estimates.

~~Since critical loads were aimed at establishing long-term balances, those agreed reductions did not address specifically the recovery of soils and waters already acidified by acid deposition. Nonetheless, major acid deposition reductions occurred, and some recovery of soils and surface waters has been noted by the monitoring operations put in place by the UNECE CLTRAP (Graf Pannatier et al., 2011; Pihl Karlsson et al., 2011; Akselsson et al., 2013).~~

As the severity of climate change became ~~more~~ fully recognised, policies for mitigation of climate change led to increased~~the~~ demand for renewable fuels ~~increased rapidly~~, thereby increasing the pressure on forests. Whole-tree harvesting, here defined as harvesting of stems and branches, was seen as an important source of renewable fuel. Since 2000 in Sweden, the proportion of clearcuts involving whole-tree harvesting has increased from around 15% to 25-35%, according to statistics from the Swedish Forest Agency, except for 2014-2016, when ~~a drop in energy prices reduced demand and~~ the proportion was temporarily between ~~declined to~~ 15-25% due to lower energy prices.~~.~~ Demand is likely to increase in the future (Börjesson et al., 2017). Harvesting of branches and the nutrient-rich needles also means a substantially increased removal of base cations compared to conventional stem harvesting, which ~~. Several studies have shown that this~~ may counteract the recovery from acidification. Thus, whereas the weathering rates in the past mainly were compared with deposition, which at that time (1970-1980) was much greater than estimated weathering and critical loads of acidic deposition, the interesting comparison in today´s policy context is between base cation weathering rates and base cation losses through harvesting, to assess sustainable forest management. Accordingly, mass balance calculations, with weathering and deposition as inputs and harvest losses and losses through leaching as outputs (Akselsson et al., 2007) (Fig. 1), as well as simplified calculations, where weathering rates are compared with base cation losses through harvesting (Olsson et al., 1993), have been used during the last decades, for forest sustainability assessments. The conclusions from these studies were that base cations may be depleted in spruce forests in ~~Olsson et al. (1993) compared Ca, Mg and K weathering rates when applying the depletion method to estimate losses of those base cations at different harvesting intensities. They found that harvest losses of Ca generally exceeded the Ca weathering rates already at stem-only harvesting, and this pattern was even~~

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~~more noticeable at whole-tree harvesting. For Mg and K, the weathering rates were generally higher than the losses at stem-only harvesting, but for whole-tree harvesting the Mg and K losses exceeded the weathering rates over large areas, especially in southern Sweden. They concluded that Ca in particular, but also Mg and K in~~ certain regions, ~~will be depleted in soils~~ if whole-tree harvesting is applied.

~~Akselsson et al. (2007) used the PROFILE model, along with mass balances, including deposition and leaching, and drew similar conclusions. Both stem-only and whole-tree harvesting gave negative balances in most areas in Sweden for all three elements in spruce forests, and for Ca and Mg in pine forests. Whole-tree harvesting increased the net losses substantially, especially for Ca and K. Iwald et al. (2013) compared the acidification effect of biomass removal with the effect of acid deposition for 1996-2009, and concluded that the acidifying effect of harvesting of stems, branches and stumps in spruce forests was 110-260% of that of acid deposition. For pine, the corresponding interval was estimated to be 60-110%. The importance of whole-tree harvesting was also demonstrated in a study from Finland, where it was shown that whole-tree harvesting doubled the removal of base cations (Aherne et al., 2012).~~

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The prospect of increasing demand for forest biomass, and particularly concerns about the effects of whole-tree harvesting on nutrient sustainability, renewed the interest in weathering in Scandinavia, for new forest policy issues. ~~However, t~~The accuracy of the weathering calculations, and the conclusions about the long-term sustainability of forests, were questioned by Klaminder et al. (2011), who compared estimations of ~~the~~ weathering rates for Ca and K from different approaches, at a site in northern Sweden~~, using different approaches~~. These estimates differed widely, and the study concluded that nutrient budgets, based on calculations including weathering rates, are too uncertain to be useful in shaping forest policies regarding harvest practices.

Futter et al. (2012) suggested that some of this variation is due to differences in boundary conditions, for example the depth to which weathering ~~has~~ had been calculated. They examined weathering estimates from 82 sites, with up to eight different weathering estimates per site, and found considerable variability in weathering rates estimated with different methods, often with results differing on the same site by several hundred percent. They identified uncertainties in input data as the largest contributor to the variability, but differences in the soil depth for which weathering was calculated also contributed, in the same way as in Klaminder et al. (2011). Futter et al. (2012) concluded that the uncertainties are large, and that at least three independent methods should be used when making management decisions.

In 2012, a~~n~~ SEK 25-million research programme, ‘Quantifying Weathering Rates for Sustainable Forestry’ (QWARTS) was started in Sweden. The programme, which focused on weathering rates for the base cations Ca, Mg, Na and K in Sweden, included approaches covering the whole spectra from laboratory-scale experiments, through plot and catchment scale experiments in the field, to extensive weathering modelling. Different approaches~~, including modelling, the depletion method, mass balance calculations~~ ~~and total analysis regression~~ were ~~tested~~ compared at different scales and were in some cases refined (Stendahl et al., 2013; Casetou-Gustafson et al., 2018~~in review a (this issue)~~; Casetou-Gustafson et al., 2019;

Casetou-Gustafson et al. (~~.,~~in review~~b) (this issue)~~; Akselsson et al., 2016; Belyazid et al., (in review~~in review (this issue~~); Kronnäs et al., 2019~~in press (this issue)~~; Erlandsson et al., 2016).

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The input data was~~ere~~ also examined for uncertainties relating to the generalisations made when estimating normative mineralogy based on total chemical analyses (Casetou-Gustafson et al., 2018; Casetou-Gustafson et al., ~~in review (this issue)~~2019). Two other potential sources of uncertainties that have been explored, but that are still widely discussed, were revisited~~. One source was~~: (1) the role of biological weathering that might generate weathering not included in the current generation of biogeochemical models (Banfield et al., 1999; Finlay et al., 2009; Finlay et al. (~~,~~in review~~ (this issue)~~) and~~. The other source concerned~~ (2) model simplifications related~~ing~~ to base cation exchange and aluminium complexation (~~Tipping 2002; ~~Gustafsson et al., 2018~~ (this issue)~~; van der Heijden et al., 2018). Furthermore, the weathering kinetics used in models were revisited (Sverdrup et al., in review), and weathering rates representing not only the rooting zone, but the full catchment scale, were studied, to assess the export of weathering products to surface waters (Ameli et al., 2017; Erlandsson et al. (~~,~~in review)~~ (this issue))~~.

The aims of this study were to (1) investigate the variation in published weathering rates from different approaches in Sweden, with consideration of the key uncertainties for each method, (2) assess the robustness of the results in relation to sustainable forestry and (3) discuss the results in relation to new insights from the QWARTS programme, and propose ways to further reduce uncertainties. ~~While weathering is important for understanding the acidification of both soils and surface waters, this paper focuses on summarising the work of QWARTS on quantifying base cation weathering rates in the rooting zone (approximately 50 cm) of Swedish forest soils.~~ While weathering is important for understanding the acidification of both soils and surface waters, this paper focuses on soils, and specifically the rooting zone (approximately 50 cm).

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~~Findings from single, well-investigated sites up to the regional and national scales are synthesized and compared. Weathering rates from new weathering studies, a few based on new datasets, have been complemented with a revisiting of older weathering studies, in the cases where methods and assumptions have been thoroughly described. Several different approaches are included, and weathering rates from the ForSAFE model are included in weathering rate comparisons for the first time (Kronnäs et al., in press (this issue); Belyazid et al., in review (this issue)). The results are compared with the base cation flows from forest harvesting, and are discussed from a forest sustainability perspective. Possible implications of new insights regarding effects of biological weathering (Finlay et al., in review (this issue)) and a more accurate description of base cation exchange and aluminium complexation (Gustafsson et al., 2018 (this issue)) are discussed. Finally, future research directions, aiming to reduce uncertainties, are outlined, based on the overall progress achieved in the QWARTS programme.~~

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2 Methods~~ for estimating weathering rates~~

Weathering rates from different approaches were compared on a site-level and on a regional level. For the comparison of weathering rates on single sites, weathering estimates of base cations (Ca, Mg, K and Na) from Swedish forest sites, where at least two well-described approaches had been applied to the same soil depth, were compiled from literature and compared (Table 1 & 3; Fig. 2). The 23 sites found were located on till soils, with a mineralogical composition characterized by granitic and gneissic bedrock, i.e with mainly qwartz, orthoclase and plagioclase, and small amounts of dark minerals such as amphibole and epidote (Table 2). Thirteen of the sites were taken from Stendahl et al. (2013). Of the originally 16 sites in that study, three were excluded, since site conditions were not appropriate for using the Depletion method (two of the sites) or PROFILE (one of the sites) (Stendahl et al., 2013). For each of the 23 sites found, medians of the different approaches were calculated along with maximum deviation from the median (Table 3). The different approaches are described in sections 2.1-2.4.

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For the regional level comparison, three published approaches for calculating weathering rates of the nutrient base cations (Ca, Mg, K) on a regional level in Sweden were revisited, harmonized and compared: PROFILE, ForSAFE and the Depletion method combined with the Total analysis regression approach (section 2.1-2.3). Na was not included, since there were no estimates for Na from the latter approach. Weathering rates were compared in 346 sites in the SAFE database (Alveteg, 2004), which were the sites of the in total 640 sites in the database for which all methods could be applied successfully, and where data on stones and boulders were available (Stendahl et al., 2009). For a regional comparison, the sites were divided into seven climate regions, simplified from 19 weather forecast regions used by the Swedish Meteorological and Hydrological Institute (SMHI) (Fig. 4). One of the regions, northwestern Sweden, only contained one site, and was therefore excluded from the analysis. The regional approaches are further in section 2.1 and 2.3. ~~Weathering rates have been estimated on a number of well-investigated sites in Sweden, using the methods described above.~~ To put the weathering estimates in a sustainability perspective, simplified base cation mass balance calculations were performed for the single sites, where weathering rates, the most important natural long-term source of base cations, were compared with harvest losses of base cations, which is the one of the outputs that can be anthropogenically controlled (Fig. 1; Section 2.5). ~~Here the weathering rates are presented for sites where at least two methods have been used for estimations to the same soil depth, and for which method descriptions have been found.~~ Finally, the results from the studies in QWARTS, on biological weathering and on the representation of base cation exchange and aluminium complexation in the models, were synthesized, main uncertainties were highlighted and ways to reduce them were proposed.

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~~The release of the base cations Ca, Mg, Na and K through weathering is difficult to quantify through direct measurements in the field due to the complexity of base cation dynamics in soil. There are several different pools of base cations in soil, and also several different flows, e.g. decomposition, uptake, ion exchange and weathering, and it is difficult to distinguish between these different flows (Rosenstock et al., in review (this issue)), or even to define the pools accurately (van der Heijden et al., 2018). Therefore, a number of indirect methods to quantify weathering rates have been developed.~~

2.1 Modelling based on weathering kinetics

Due to the difficulties in measuring field weathering rates, weathering kinetics have been frequently studied in laboratory environments ~~Mechanistic modelling, based on laboratory experiments, describing the kinetics~~(Brantley et al., 2008). Mechanistic modelling of weathering rates, based on laboratory-determined weathering kinetics, is one of the most widely used approaches for estimating field weathering rates (Warfvinge and Sverdrup, 1992; Godderis et al., 2006; Maher et al., 2009) ~~is one way to get around the difficulties of measuring~~ .~~weathering in the field.~~ The PROFILE model (~~Sverdrup and~~ Warfvinge and Sverdrup, 1992~~3~~) is a steady state soil chemistry model where weathering is derived from the breakdown of minerals, based on ~~with~~ process-oriented descriptions of ~~-~~chemical weathering and solution equilibrium reactions. ~~, and has been used widely for estimating weathering in Europe (Akselsson et al., 2004; 2016; Stendahl et al., 2013; Holmqvist et al., 2003; Koptsik et al., 1999; Langan et al., 1995), the US (Phelan et al., 2014) and Asia (Fumoto et al., 2001).~~

Weathering rates are modelled calculated for different layers, with different soil properties, using transition state theory and the geochemical properties of the soil system, such as soil wetness, temperature, mineral surface area and mineral composition, and organic acid concentrations. Deposition of sulphur, nitrogen and base cations, as well as net losses of base cations and nitrogen through harvesting, are used as input for modelling of pH and base cation concentrations in soil water, which is required for the weathering modelling. Weathering rates are calculated for each mineral separately, using rate coefficients from laboratory studies for four reactions: with ~~hydrogen ions~~H^+, water, ~~carbon dioxide~~CO_2 and ~~organic ligands~~dissolved organic carbon (DOC) (Sverdrup and Warfvinge, 1993). PROFILE has been used widely for estimating weathering in Europe (Akselsson et al., 2004; 2016; Stendahl et al., 2013; Holmqvist et al., 2003; Koptsik et al., 1999; Langan et al., 1995), the US (Phelan et al., 2014) and Asia (Fumoto et al., 2001). At all 23 sites in the single-site comparison in this paper, weathering estimates from PROFILE were available (Table 3).

The weathering submodel in PROFILE was later built in to the dynamic version ~~of PROFILE,~~ SAFE (Alveteg et al., 1995), which was mainly used for acidification assessments, but has also been used for studying the dynamics of weathering rates (Warfvinge et al., 1995). Later, the SAFE model was coupled with the tree growth model PnET (Aber and Federer, 1992), the decomposition model DECOMP (Walse et al., 1998; Wallman et al., 2004) and the hydrological model PULSE (Lindström and Gardelin, 1992), resulting in ~~and in~~ the forest ecosystem model ForSAFE (Wallman et al., 2005; Belyazid et al., 2006). The ForSAFE model simulates the integrated biogeochemical processes of a forest ecosystem. It covers the processes of photosynthesis, allocation and growth, water and nutrient uptake, litterfall, organic matter decomposition and mineralisation, ion exchange, chemical speciation of and reactions between different elements, as well as hydrological transport.~~), where the SAFE model has been coupled with the tree growth model PnET (Aber and Federer, 1992), the decomposition model DECOMP (Walse et al., 1998; Wallman et al., 2004) and the hydrology model PULSE (Lindström and Gardelin, 1992). SAFE is used mainly for acidification assessments, and~~ All process rates are internally regulated by microenvironmental conditions such as acidity, water availability, temperature and element concentrations. The model

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requires inputs of external drivers in the form of climate, atmospheric deposition and forest management, and inputs on the properties of the forest ecosystem, such as soil texture, mineralogy and tree species. ForSAFE is used for studying effects of climate change, atmospheric deposition and forest management on tree growth, soil chemistry and runoff water quality. Although the weathering module is the same in PROFILE and ForSAFE, some differences can be expected since ForSAFE includes dynamics, which means that weathering is affected by other processes over time, and that soil moisture, which is an input in PROFILE, is dynamically modelled in ForSAFE. ~~.~~In the single-site comparisons in this paper, weathering estimates from ForSAFE were available at two sites (Table 3).

For the regional comparison of weathering rates of nutrient base cations (Ca, Mg, K), regional runs from both PROFILE and ForSAFE where included. Regional PROFILE weathering estimations for Sweden were taken from Akselsson et al. (2016), where weathering rates for the upper 50 cm of the soil (including the organic layer) has been modelled based on data from 17,333 Swedish National Forest Inventory (NFI) sites (Fridman et al., 2014).

Within QWARTS, ~~The weathering submodel in PROFILE was later built into the dynamic version of PROFILE, SAFE (Alveteg et al., 1995) and in the forest ecosystem model ForSAFE (Wallman et al., 2005; Belyazid et al., 2006), where the SAFE model has been coupled with the tree growth model PnET (Aber and Federer, 1992), the decomposition model DECOMP (Walse et al., 1998; Wallman et al., 2004) and the hydrology model PULSE (Lindström and Gardelin, 1992). SAFE is used mainly for acidification assessments, and ForSAFE is used for studying effects of climate change, atmospheric deposition and forest management on tree growth, soil chemistry and runoff water quality.~~

~~The models have also been used to explicitly study weathering rates (Warfvinge et al., 1995; Kronnäs et al., in press (this issue); Belyazid et al., in review (this issue)). The ForSAFE model simulates the integrated biogeochemical processes of a forest ecosystem. It covers the processes of photosynthesis, allocation and growth, water and nutrient uptake, litterfall, organic matter decomposition and mineralisation, ion exchange, chemical speciation of and reactions between different elements, as well as hydrological transport (Wallman et al., 2005). All process rates are internally regulated by microenvironmental conditions such as acidity, water availability, temperature and element concentrations. The model requires inputs of external drivers in the form of climate, atmospheric deposition and forest management, and inputs on the properties of the forest ecosystem, such as soil texture, mineralogy and tree species.~~w~~W~~eathering rates to 50 cm depth (including the organic layer) were modelled also with the ForSAFE model (Belyazid et al., in review) on ~~the 640~~ sites ~~in the SAFE database~~in the SAFE database~~ (Alveteg, 2004)~~. The SAFE database is a subset, consisting of 640 sites, of the NFI sites used for PROFILE modelling. ~~. Information on soil texture and mineralogy were derived from the database of the Research Infrastructure National Forest Inventory of Sweden (RINFI, Hägglund, 1985). Mineralogy was derived from total elemental analysis translated through the UPPSALA model (Sverdrup et al., 2002). Stones and boulders were taken into account using the same data on contents of stones and boulders as for PROFILE and the depletion method.~~

~~The climatic input data were derived from simulations of historical and future trends, based on the IPCC's A2 story line of emissions (David Rayner, pers. comm.) and downscaled to historical records from the Swedish Meteorological Institute~~

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~~(SMHI). The climate data include monthly information on air temperature, precipitation, photosynthetically active radiation and atmospheric CO_2 concentrations.~~

~~Atmospheric deposition data for sulphate, nitrate and ammonium were derived from simulations by the EMEP model (Simpson et al., 2012) according to the emissions history in Schöpp et al. (2003) and the projected emissions following the current legislation of the UN Convention on Long Range Transboundary Air Pollution (LRTAP). Atmospheric deposition data for chloride, calcium, sodium, potassium and magnesium were derived from the MATCH model (Persson et al., 1996).~~

~~Internationally, some other weathering models such as WITCH (Godderis et al., 2006) or CrunchFlow (Maher et al., 2009) employ transition-state theory rate laws for the kinetic descriptions of mineral dissolution (Aagaard and Helgeson, 1982), just like in PROFILE, SAFE and ForSAFE. However, there is a difference from the kinetic equations of PROFILE, SAFE and ForSAFE in terms of response to pH and aluminium concentrations (Erlandsson et al., 2016). Nonetheless, PROFILE has previously been demonstrated to replicate field weathering rates as determined by independent methods for a wide range of soils without calibration (Sverdrup and Warfvinge, 1993).~~

~~With a few exceptions, all process-based weathering models have been restricted to the 1D plot scale and unsaturated flow. Model estimates of the weathering flux from larger volumes of soil, such as hillslopes or catchments, are rare. Exceptions include the use of PROFILE for surface waters (Rapp and Bishop, 2003), the WITCH model to calculate the weathering rates in the Vosges catchment in Luxemburg (Godderis et al., 2006), and Erlandsson et al. (in review, this issue)), who used a mixing model, where the water transit time distribution and weathering rates were modelled separately to estimate the weathering flux from a hillslope in the Vindeln research park in Northern Sweden.~~

~~The MAGIC model (Cosby et al., 2001) was developed to predict effects of acidic deposition on surface water acidification. Weathering is not mechanistically modelled as in the models described above; instead, weathering rates are calculated internally using mass balances (Maxe, 1995; Köhler et al., 2011). MAGIC uses input fluxes from atmospheric deposition and weathering and output fluxes through net uptake and loss in biomass and to runoff. These fluxes govern processes in the soil, e.g. cation exchange, with the pool of exchangeable base cations in the soils at the centre. When the fluxes change over time, it affects the chemical equilibria between soil and soil solution, which has an impact on surface water chemistry. Observed values of surface water and soil chemistry are used to calibrate the model.~~

2.2 The ~~depletion~~ Depletion method

Another widely used approach for estimating weathering rates is the ~~depletion~~ Depletion method (e.g. Olsson et al., 1993; Starr et al., 1988; Stendahl et al., 2013). The method estimates historical weathering, i.e. the average weathering rate since the last deglaciation, of mobile (weatherable) elements, based on element concentrations in weathered soil horizons as compared to~~to~~ unweathered parent material. The method accounts for the general losses of soil material in a horizon by including an immobile (inert) element in the estimation. Concentrations of mobile elements will decrease as a result of weathering, while the immobile element will be enriched towards the soil surface. The concept has a long history (Marshall and Haseman 1942), while the theoretical framework was later formalised by Brimhall and Dietrich (1987) and Brimhall et al. (1991). The most commonly used immobile element is zirconium, which is found in the resistant mineral zircon (ZrSiO4) with negligible weathering (Hodson, 2002). The assumptions for the ~~depletion~~ Depletion method are: (1) there is no weathering of the immobile element, (2) the soil pedon consists of homogeneous soil, where the deep soil constitutes the parent material ~~parent material~~, and (3) no weathering occurs beyond a certain depth. The average annual weathering rate is calculated from the soil age, i.e. the time since deglaciation or since the land rose from the sea due to glacio-isostatic uplift. The average rate may deviate from current levels depending on the variation in weathering rates over time (Taylor and Blum, 1995). Weathering rates from the Depletion method were available for 18 of the 23 sites in the single-site comparison (Table 3).The Depletion method has been used, in combination with the Total analysis regression approach, for regional applications (see section 2.3).

2.3~~5~~ Total ~~a~~analysis r~~r~~egression approach~~s~~

The ~~t~~Total analysis regression approach~~n method~~ is a derivative from the Depletion method, which requires much less soil data than the Depletion method. It is based on the fact that weathering rates of different elements have been found to correlate with total content of the elements in soil and temperature (Olsson and Melkerud, 1990). Based on weathering estima~~tions~~ tes from the ~~d~~Depletion method, linear regressions containing total chemical contents for base cations in the C horizon (either separately or lumped together), and temperatures or temperature sums (i.e. daily mean temperature above a threshold value, summarized for the growing season), have been produced for a number of sites, and the regressions have then been applied to other sites (Sverdrup et al., 1998; Maxe, 1995; Olsson et al., 1993). In the single-site comparison, estimates based on the Total analysis regression approach were available for three sites (Table 3).

On a regional level in Sweden, weathering rates for Ca, Mg and K have been calculated based on the Depletion method in combination with the Total analysis regression approach, as a basis for assessments of nutrient sustainability after whole-tree harvesting (Olsson et al., 1993). In the study from 1993, regressions between weathering rates calculated with the Depletion method and different site factors were analysed on 11 sites. The strongest relationships were found between weathering rates of an element and the product of the concentration of the element in the C horizon and the temperature sum. In the present study, the regression functions in Olsson et al. (1993) were used to calculate weathering rates of Ca, Mg and K at the sites from the SAFE database that were modelled with both PROFILE and ForSAFE (see above), for a proper comparison. The temperature sum was calculated based on latitude and altitude according to Morén and Perttu (1994). Some of the calculations gave negative results for one of the base cations. This can be explained by the fact that the regressions are used for a new dataset, covering a broader range of temperature sums than the original dataset, which illustrates a limitation of the method. The estimations apply down to the weathering depth, which means different soil depths on different sites, but normally between 40 and 70 cm in the mineral soil (Mats Olsson, pers. comm).

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2.4~~3~~ Budget ~~calculations~~approach

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Weathering rates can also be estimated through the ~~mass balance~~Budget approach~~calculations~~ (Paces, 1986; Lundström, 1990; Sverdrup and Warfvinge, 1991; Sverdrup et al., 1998). In the Budget approach~~mass balance calculations~~, sources and sinks of base cations are considered, and weathering is calculated as the difference between sinks and sources. However, one difficulty is to distinguish between weathering, changes in the exchangeable pool and net mineralisation, so steady state is often assumed, and the weathering rates are calculated as leaching + net uptake – deposition. The Budget approach has been applied at five of the sites in the site-level comparison (Table 3). ~~This approach was applied i~~In Svartberget in northern Sweden the simplified approach assuming steady state was used (Lundström, 1990). ~~In~~ On two sites in Gårdsjön in southwestern Sweden, net mineralisation was estimated and considered in the weathering estimations, but changes in the exchangeable pool were disregarded (Sverdrup et al., 1998). Casetou-Gustafson et al. (in review) ~~Simonsson et al. (2015)~~ estimated weathering rates ~~over a 14-year period in Skogaby in southern Sweden,~~ based on measurements of atmospheric deposition, leaching, accumulation in biomass and changes in the soil exchangeable pool~~. The same approach was used by Casetou et al. (in review b, this issue)) in the mass balance estimates~~ for control plots of long-term fertilisation experiments in young Norway spruce forests in Asa and Flakaliden, in southern and northern Sweden, respectively. ~~In the latter two studies, leaching, soil change and accumulation in biomass were measured in control plots of long-term fertilisation experiments in young Norway spruce forests.~~ When using weathering estimates from ~~budget calculations~~the Budget approach, the assumptions used must be carefully evaluated (Sverdrup and Warfvinge, 1991; Rosenstock et al.~~, in review~~ ~~(this(~~ in review~~ssue)~~).

The Budget approach can also be applied by using t~~T~~he MAGIC model (Cosby et al., 2001), which was developed to predict effects of acidic deposition on surface water acidification. Weathering is not mechanistically modelled as in the models described above; instead, weathering rates are calculated internally using mass balances (Maxe, 1995; Köhler et al., 2011). MAGIC uses input fluxes from atmospheric deposition and weathering and output fluxes through net uptake and loss in biomass and to runoff. These fluxes govern processes in the soil, e.g. cation exchange, with the pool of exchangeable base cations in the soils at the centre. When the fluxes change over time, it affects the chemical equilibria between soil and soil solution, which has an impact on surface water chemistry. Observed values of surface water and soil chemistry are used to calibrate the model. Weathering rates from MAGIC were available at two of the sites in the site-level comparison (Table 3).

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~~2.4 Budget calculations using the strontium isotope ratio~~

Another way of applying the Budget approach is to use the strontium (Sr) isotope ratio. In weathering estimations based on ~~strontium (Sr)~~Sr isotope ratios, the difference in the ratio ^{87}Sr/^{86}Sr in bedrock and in atmospheric deposition is used (Wickman and Jacks, 1991). Since soil water is a mixture of what comes from deposition and what comes from weathering, the weathering rate of Sr can be estimated. Ca and Sr follow each other closely in forests (Wickman and Jacks, 1991), so the weathering of Ca is assumed to be linearly correlated to the weathering of Sr in the calculations. This method was used at three sites in the site-level comparison ~~To estimate base cation weathering, as was done for Gårdsjön (Sverdrup and Warfvinge, 1993), and in Svartberget and Risfallet~~ (~~Sverdrup and Warfvinge, 1991),~~Table 3). ~~a constant base cation/Ca fraction is assumed.~~The base cation/Ca fraction was assumed to be constant in all three studies.

~~2.5 Total analysis regressions~~

~~The total analysis regression method is based on the fact that weathering rates of different elements have been found to correlate with total content of the elements in soil and temperature (Olsson and Melkerud, 1990). Based on weathering estimations from the depletion method, linear regressions containing total chemical contents for base cations in the C horizon (either separately or lumped together), and temperature or temperature sums, have been produced for a number of sites, and the regressions have then been applied to other sites (Sverdrup et al., 1998; Maxe, 1995; Olsson et al., 1993).~~

~~Three of the 16 sites reported in Stendahl et al. (2013) were disqualified. One site in the north gave unrealistically high weathering values with PROFILE, which is probably at least partly due to a very high bulk density; the simplified hydrology routine in PROFILE, with only vertical flow, is unsatisfactory for these conditions. For the other two sites the profiles did not fulfil the assumptions for the depletion method.~~

2.5 Assessment of forest sustainability

Simplified budget calculations based only on weathering rates and harvest losses (Olsson et al., 1993; Klaminder et al., 2011; Stendahl et al., 2013), were performed for a selection of the sites in Table 3. The criteria that had to be fulfilled for inclusion of a site were: (1) availability of weathering rate assessments for the root zone which in Swedish forest soils often defined as 0.5 m (Rosengren and Stjernquist, 2004), but in this study included depths down to 0.7 m and (2) access to data on site quality. Calculations were made for sites in spruce and pine forest.

The calculations of harvest losses were based on the site quality of the forest (average growth rate per year during a forest rotation and optimal conditions), reduced by 20% to mimic actual conditions, and generalised densities and nutrient base cation concentrations in different tree parts. Two types of harvesting were considered, conventional stem-only harvesting and whole-tree harvesting, where in addition to stem, tops and branches are removed for biofuel. It was assumed that, in whole-tree harvesting, all stems and 60% of the branches were harvested, and that 75% of the needles were removed with the harvested branches. The methodology along with densities and base cation concentrations used is more thoroughly described in Akselsson et al. (2007).

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3. Results and Discussion

3.1 Weathering rate comparisons at site level

~~Weathering rates have been estimated on a number of well-investigated sites in Sweden, using the methods described above. Here the weathering rates are presented for sites where at least two methods have been used for estimations to the same soil depth, and for which method descriptions have been found. This~~ The single-site comparison enabled us to quantify the span of base cation weathering rates produced by the different approaches~~, as a way of “framing” the weathering rates on the sites~~ (Fig. ~~1-2~~3, Table ~~1~~3). The median weathering rates for the 23 sites spanned between 22 and 61 meq m^{-2} y^{-1} (Table 3). For two of the six sites where at least three approaches had been applied, the weathering rate spans were narrow, Gårdsjön B (49-62 meq m^{-2} y^{-1}, and a maximum deviation from the median of +15%) and Gårdsjön C (36-40 meq m^{-2} y^{-1}, and a maximum deviation from the median of ±5%). The span was somewhat wider in two of the other well-investigated sites, Stubbetorp (35-67 meq m^{-2} y^{-1}, and a maximum deviation from the median of +56%) and Flakaliden (34-61 meq m^{-2} y^{-1}, and a maximum deviation from the median of +42%). The weathering rate spans in Svartberget B and Asa, with four and three weathering estimates respectively, were remarkably wide: 32-79 meq m^{-2} y^{-1}, and a maximum deviation from the median of +121% in Svarberget B and 11-131 meq m^{-2} y^{-1} and a maximum deviation from the median of 254% in Asa. The wide spans in Asa and Svartberget B were mainly due to substantially higher weathering rates according to the Budget approach as compared with the other methods. Also in Flakaliden, the span was expanded by the Budget approach.

In Svartberget B, the weathering estimates from the Budget approach can be expected to be overestimated for several reasons (Lundström, 1990). The method does not distinguish between weathering, base cation exchange and base cation release through decomposition. The measurements were carried out in the 1980s when the acidification process was taking place, leading to base-cation release from the exchangeable pool, although this effect was much more pronounced in southern Sweden. Finally, dry deposition was not included in the calculations due to lack of data. In Flakaliden and Asa, base cation accumulation in biomass was the major sink in the mass balances. The high weathering rates produced by the mass balances, especially in Asa, were largely explained by the measured depletion of base cations in the soil being much lower than the accumulation in the young Norway spruce stands (Casetou-Gustafson et al., in review). The very high estimated weathering rates, especially in Asa, indicate that flows are described inadequately. Uptake occurring below the defined rooting zone could be one contributing factor (Casetou-Gustafson et al., in review). In Asa, very low weathering rates from the Depletion method contributed to the wide span. The low weathering rates originated from a fairly flat Zr depth gradient in the soil, which indicates that the soil had probably been disturbed, so the necessary assumptions for the Depletion method were not satisfied (Casetou-Gustafson et al., in review).

In Stubbetorp, the PROFILE estimates were substantially higher (67 meq m^{-2} y^{-1}), than the estimates from the Total analysis regression approach and MAGIC (30-51 meq m^{-2} y^{-1}). Maxe (1995) noted that the PROFILE weathering rate was higher than expected, given the soil properties in Stubbetorp, and argued that it may be due to unreasonably high specific surface area of

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the soil as input to PROFILE on the site. Specific surface area has been determined by BET analysis, and could, according to Maxe (1995), be overestimated due to a large occurrence of Al and Fe precipitates.

Whereas weathering rates in the different sites in Gårdsjön (A, B and C) and Svartberget (A and B) were generally on the same level, except for the Budget approach in Svartberget B as discussed above, the two sites in Risfallet (A and B) gave quite different weathering rates, 29-68 meq m^{-2} y^{-1} in Risfallet A (0.5 m) and 25-29 meq m^{-2} y^{-1} in Risfallet B (1 m). It could be expected that the weathering rates were higher in the deeper soil profile, but instead it was the other way around. The PROFILE modelled weathering rate in Risfallet A is one of the highest of all sites reported in Stendahl et al. (2013), which can be explained by a relatively high clay content (7%) and high soil bulk density. In contrast, Risfallet B (1 m) has a very low specific surface area (Sverdrup and Warfvinge, 1993), which can explain the low weathering rate produced by the model.

In five of the seventeen cases where only two methods per site were applied, in most cases PROFILE and the Depletion method, the maximum difference between the calculated median and the estimated weathering rates was less than ±10% (Table 3). In the other end, five sites showed a corresponding difference between ±40% and ±64%. The results show that the width of the span varies substantially between sites, and to explain these differences the sites and the methods need to be studied in detail.

The weathering rates calculated with the Depletion method for the 13 sites from Stendahl et al. (2013) were generally lower, than the PROFILE-modelled weathering rates (Table 3). The two sites with largest difference, Skånes Värsjö and Kloten, with a maximum difference between the median and the estimated weathering rate of around ±60%, distinguished themselves with very low rates estimated with the Depletion method, 8 and 11 meq m^{-2} y^{-1}. This is contrary to what was expected, since the weatherability of the soil is believed to decrease over time due to the depletion of more easily weathered minerals and formation of resistant coatings on the mineral surfaces (Taylor and Blum, 1995). The reasons are not fully known, but one reason could be that the original till partly comprises already weathered till from previous glaciations (Stendahl et al., 2013). Moreover, it is likely that declining weatherability over time is less pronounced in these young glacial till profiles, where the easily weathered minerals remain in the profile. Furthermore, some drivers of weathering, such as forest growth, are more prominent today, which may overshadow long-term decline in soil weatherability. For the total sum of base cations there was a tendency towards higher modelled rates when the rates from the Depletion method were higher, but the relationship was weak (r^2=0.20) on the 13 sites (Stendahl et al., 2013). However, for Ca and Mg there was a much stronger relationship, r^2=0.46 for Ca and r^2=0.64 for Mg. Thus, based on current knowledge and models, it seems possible to identify a narrow weathering rate interval for Ca and Mg weathering rates, but it seems difficult for K and Na, as discussed in Stendahl et al. (2013).

At Västra Torup and Hissmossa, weathering rates produced by PROFILE and ForSAFE were compared in detail for the first time. The results at Västra Torup showed that the maximum difference between the modelled weathering rates and the median was ±4%. At Hissmossa, the corresponding difference was ±9%. Much of the difference could be attributed to the difference between soil moisture input data in PROFILE and modelled soil moisture in ForSAFE. The sandy soil in

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Hissmossa gave substantially lower modelled soil moisture than the moisture estimates based on field observations used as inputs to PROFILE, resulting in lower weathering rates (Kronnäs et al., 2019).

~~For Svartberget B (0.8 m), three methods gave weathering rates within the interval 31-42 meq m^{-2} yr^{-1} whereas the fourth one, the budget study, produced a substantially higher weathering rate, 85 meq m^{-2} yr^{-1}. The aim of the budget study was to evaluate the importance of organic substances on weathering rates, which involved comparing two soil compartments, 0-20 cm and 30-80 cm, estimated as the difference between the sum of base cation deposition and base cation losses and the sum of net uptake and leaching (Lundström, 1990). The weathering estimates from the budget calculations can be expected to be overestimated for several reasons. The method does not distinguish between weathering, base cation exchange and base cation release through decomposition. The measurements were carried out in the 1980s when the acidification process was taking place, leading to base-cation release from the exchangeable pool, although this effect was much more pronounced in southern Sweden. Moreover, dry deposition was not included in the calculations due to lack of data.~~

~~For Flakaliden and Asa, three different weathering estimates were available, and the span was very large, particularly for Asa in southern Sweden. The mass balance method produced the highest weathering rates at both sites. However, weathering estimates for Flakaliden by the mass balance method and PROFILE were similar to the estimates produced by the same methods for the nearby (≈ 40 km) Svartberget B site. For Flakaliden, PROFILE produced a weathering rate twice as high as that produced by the depletion method, which is the same order of magnitude as many of the other sites. However, at Asa the estimated weathering rate with the depletion method was very low, 11 meq m^{-2} yr^{-1}, which is among the lowest estimated weathering rates found on all sites, with all methods, whereas PROFILE gave a weathering rate of the same size as many of the other sites. At both Asa and Flakaliden, base carion accumulation in biomass was the major sink in the mass balances. The high weathering rates produced by the mass balances were largely explained by the measured depletions of base cations in the soil being much lower than the accumulation in the young Norway spruce stands. The very high mass balance weathering rate at Asa was therefore mainly attributed to the higher growth rate at that site. The very low weathering rates produced by the depletion method at Asa originated from a fairly flat Zr depth gradient in the soil, which indicates that the soil had probably been disturbed, so the necessary assumptions for the Zr depletion method were not satisfied (Casetou Gustafson et al., in review b (this issue)).~~

~~The weathering rates calculated with the depletion method for the remaining 13 sites were generally lower, on average 50% lower, than the PROFILE-modelled weathering rates. This is contrary to what was expected, since the weatherability of the soil is believed to decrease over time due to the depletion of more easily weathered minerals and formation of resistant coatings on the mineral surfaces (Taylor and Blum, 1995). For Mg, the depletion method did indeed give higher values than PROFILE, in accordance with the theories, but for Ca and Na the reverse applied. The reasons are not fully known, but probably the declining weatherability over time is less pronounced in these young glacial till profiles, where the easily weathered minerals remain in the profile. Furthermore, some drivers of weathering, such as forest growth, are more prominent today, which may overshadow long-term decline in soil weatherability. Another plausible factor is that the original till probably partly comprises already weathered till from previous glaciations (Stendahl et al., 2013). For the total~~

~~sum of base cations there was a tendency towards higher modelled rates when the rates from the depletion method were higher, but the relationship was weak (r^2=0.20) on the 13 sites. However, for Ca and Mg there was a much stronger relationship, r^2=0.46 for Ca and r^2=0.64 for Mg. The relationship was weaker for K and Na, possibly due to conceptual limitations in the models. Based on current knowledge and models, it seems possible to identify a narrow weathering rate interval Ca and Mg weathering rates, but it seems difficult for K and Na, as discussed in Stendahl et al. (2013).At Västra Torup and Hissmossa, weathering rates produced by PROFILE and ForSAFE were compared in detail for the first time. Although the process descriptions of weathering are the same in the two models, the dynamics of ForSAFE were expected to produce differences in estimated weathering rates. The results at Västra Torup showed that ForSAFE produced weathering rates 8% higher than the rates produced by PROFILE. At Hissmossa, PROFILE gave 19% higher weathering rates than ForSAFE. Much of the difference could be attributed to the difference between soil moisture input data in PROFILE and modelled soil moisture in ForSAFE. The sandy soil in Hissmossa gave substantially lower modelled soil moisture than the moisture estimates based on field observations used as inputs to PROFILE, resulting in substantially lower weathering rates.~~

~~At Gårdsjön, the different approaches resulted in weathering rates in the interval 36-62 meq m^{-2} yr^{-1} (Table 1, Fig. 2). Some of the differences can be explained by the somewhat different depths; the highest rates in the interval are associated with a profile 17-20 cm deeper than the other ones. The similarities in weathering rates indicate that the framing of the weathering rates at Gårdsjön has been successful, i.e. that a limited range of weathering rates at Gårdsjön has been successfully defined. Moreover, it indicates that the soil conditions in the Gårdsjön area are rather homogeneous.~~

~~For Svartberget B (0.8 m), three methods gave weathering rates within the interval 31-42 meq m^{-2} yr^{-1} whereas the fourth one, the budget study, produced a substantially higher weathering rate, 85 meq m^{-2} yr^{-1}. The aim of the budget study was to evaluate the importance of organic substances on weathering rates, which involved comparing two soil compartments, 0-20 cm and 30-80 cm, estimated as the difference between the sum of base cation deposition and base cation losses and the sum of net uptake and leaching (Lundström, 1990). The weathering estimates from the budget calculations can be expected to be overestimated for several reasons. The method does not distinguish between weathering, base cation exchange and base cation release through decomposition. The measurements were carried out in the 1980s when the acidification process was taking place, leading to base-cation release from the exchangeable pool, although this effect was much more pronounced in southern Sweden. Moreover, dry deposition was not included in the calculations due to lack of data.~~

~~The nearby sites, Svartberget A (0.5 m) and Vindeln (0.5 m), were expected to give lower weathering rates than Svartberget B, due to the shallower depth, and this proved to be the case, 17-38 meq m^{-2} yr^{-1} for Svartberget A and 13-30 meq m^{-2} yr^{-1} for Vindeln. The lower values in the intervals are based on the depletion method, whereas the higher are from PROFILE. Although the results from Svartberget were more scattered than for Gårdsjön, it could be concluded that the weathering rate is probably lower at Svartberget than at Gårdsjön (Table 1, Fig. 2).~~

~~At Risfallet B (1 m), two methods were applied to the same depth, PROFILE and the Sr method. They gave similar results, 25-29 meq m^{-2} yr^{-1}. It is notable that the weathering rate for Risfallet A (0.5 m) was higher, in spite of the shallower depth,~~

~~with an interval of 29-68 meq m^{-2} yr^{-1}, where the higher value is from PROFILE. The weathering rate modelled by PROFILE in Risfallet A (0.5 m) is one of the highest of all sites reported in Stendahl et al. (2013), which can be explained by a relatively high clay content (7%) and high density. In contrast, Risfallet B (1 m) has a very low specific surface area (Sverdrup and Warfvinge, 1993), much lower than in Risfallet A (0.5 m), which can explain the low weathering rate produced by the model. Based on the approaches used so far at Risfallet, the weathering rate is hard to to define with such a high level of accuracy as for Gårdsjön and Svartberget (Table 1, Fig. 2), which can be at least partly explained by varying soil texture within short distances.~~

~~At Stubbetorp, total analysis regression and MAGIC gave weathering rates in the interval 30-51 meq m^{-2} yr^{-1}, whereas PROFILE gave a higher rate, 67 meq m^{-2} yr^{-1}. Maxe (1995) noted that the PROFILE weathering rate was higher than expected, given the soil properties in Stubbetorp, and argued that it may be due to unreasonably high specific surface area of the soil as input to PROFILE on the site. Specific surface area has been determined by BET analysis, and could, according to Maxe (1995), be overestimated due to a large occurrence of Al and Fe precipitates.~~

~~For Flakaliden and Asa, three different weathering estimates were available, and the span was very large, particularly for Asa in southern Sweden. The mass balance method produced the highest weathering rates at both sites. However, weathering estimates for Flakaliden by the mass balance method and PROFILE were similar to the estimates produced by the same methods for the nearby (≈ 40 km) Svartberget B site. For Flakaliden, PROFILE produced a weathering rate twice as high as that produced by the depletion method, which is the same order of magnitude as many of the other sites. However, at Asa the estimated weathering rate with the depletion method was very low, 11 meq m^{-2} yr^{-1}, which is among the lowest estimated weathering rates found on all sites, with all methods, whereas PROFILE gave a weathering rate of the same size as many of the other sites. At both Asa and Flakaliden, base carion accumulation in biomass was the major sink in the mass balances. The high weathering rates produced by the mass balances were largely explained by the measured depletions of base cations in the soil being much lower than the accumulation in the young Norway spruce stands. The very high mass balance weathering rate at Asa was therefore mainly attributed to the higher growth rate at that site. The very low weathering rates produced by the depletion method at Asa originated from a fairly flat Zr depth gradient in the soil, which indicates that the soil had probably been disturbed, so the necessary assumptions for the Zr depletion method were not satisfied (Casetou Gustafson et al., in review b (this issue)).~~

~~At Västra Torup and Hissmossa, weathering rates produced by PROFILE and ForSAFE were compared in detail for the first time. Although the process descriptions of weathering are the same in the two models, the dynamics of ForSAFE were expected to produce differences in estimated weathering rates. The results at Västra Torup showed that ForSAFE produced weathering rates 8% higher than the rates produced by PROFILE. At Hissmossa, PROFILE gave 19% higher weathering rates than ForSAFE. Much of the difference could be attributed to the difference between soil moisture input data in PROFILE and modelled soil moisture in ForSAFE. The sandy soil in Hissmossa gave substantially lower modelled soil moisture than the moisture estimates based on field observations used as inputs to PROFILE, resulting in substantially lower weathering rates.~~

~~Three of the 16 sites reported in Stendahl et al. (2013) were disqualified. One site in the north gave unrealistically high weathering values with PROFILE, which is probably at least partly due to a very high bulk density; the simplified hydrology routine in PROFILE, with only vertical flow, is unsatisfactory for these conditions. For the other two sites the profiles did not fulfil the assumptions for the depletion method. The weathering rates calculated with the depletion method for the remaining 13 sites were generally lower, on average 50% lower, than the PROFILE-modelled weathering rates. This is contrary to what was expected, since the weatherability of the soil is believed to decrease over time due to the depletion of more easily weathered minerals and formation of resistant coatings on the mineral surfaces (Taylor and Blum, 1995). For Mg, the depletion method did indeed give higher values than PROFILE, in accordance with the theories, but for Ca and Na the reverse applied. The reasons are not fully known, but probably the declining weatherability over time is less pronounced in these young glacial till profiles, where the easily weathered minerals remain in the profile. Furthermore, some drivers of weathering, such as forest growth, are more prominent today, which may overshadow long-term decline in soil weatherability. Another plausible factor is that the original till probably partly comprises already weathered till from previous glaciations (Stendahl et al., 2013). For the total sum of base cations there was a tendency towards higher modelled rates when the rates from the depletion method were higher, but the relationship was weak (r^2=0.20) on the 13 sites. However, for Ca and Mg there was a much stronger relationship, r^2=0.46 for Ca and r^2=0.64 for Mg. The relationship was weaker for K and Na, possibly due to conceptual limitations in the models. Based on current knowledge and models, it seems possible to identify a narrow weathering rate interval Ca and Mg weathering rates, but it seems difficult for K and Na, as discussed in Stendahl et al. (2013).~~

~~In Fig. 2 t~~The intervals of all sites we~~a~~re compared with four reference weathering intervals, based on weathering rate approximations frequently used in the critical load work (Fig. 3; de Vries, 1994; Umweltsbundesamt, 1996). The majority of weathering rate estimates was within or close to the interval outlined for acidic/intermediate parent material with coarse texture. ~~The four intervals are based on different combinations of parent material (acidic, intermediate and basic) and texture (coarse-, medium- and fine-grained). The coarse fraction, defined as soil with a clay content of less than 18 % (Umweltsbundesamt, 1996) is predominant in Sweden, and acidic parent material, defined as material composed of e.g. sandstone, granite and gneiss, is the most common parent material in Sweden.~~

~~The intervals for many of the sites were within or close to the interval outlined for acidic/intermediate parent material with coarse texture, corresponding to Swedish conditions (Fig. 2). Gårdsjön had small enough intervals to be able to define the intervals in the weathering rates with relatively high accuracy. This was also the case for Svartberget, if the outlier from the budget calculations was disregarded. Risfallet showed more contradictory results, which are hard to interpret, since the soil sampling was performed several decades ago. The weak correlation between the weathering rates from PROFILE and the depletion method in Stendahl et al. (2013) makes it difficult to, in detail, rank all the sites based on the weathering rates. However, some patterns are clear, e.g. that Gårdsjön seems to have higher weathering rates than most other sites, whereas Vindeln and Hjärtasjö lie at the lower end.~~

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The weathering rates for the sites with 0.5 m soil depth (including Gårdsjön G1 where the soil depth is 0.47 m) can roughly be divided into four different groups depending on the intervals, except for four sites with contradictory results (Table ~~2~~4). The assessment of whether the weathering rate intervals are accurate enough depends on their intended use. The weathering rates in relation to forest sustainability assessments are analysed in Sect. ~~7~~3.3.

~~4~~ 3.2 Weathering rate comparisons ~~on a~~at a regional scale

~~4.1 Total analysis regressions~~

~~Weathering rates for Ca, Mg and K has been calculated on a regional level in Sweden, based on the total analysis regression method, as a basis for assessments of nutrient sustainability after whole-tree harvesting (Olsson et al., 1993). In the study from 1993, regressions between weathering rates calculated with the depletion method and different site factors were analysed on 11 sites. The strongest relationships were found between weathering rates of an element and the product of the concentration of the element in the C horizon and the temperature sum. In the present study, the regression functions in Olsson et al. (1993) were used to calculate weathering rates of Ca, Mg and K on 640 sites in the SAFE database (Alveteg, 2004), which contains the data required for the regression calculations. The weathering rates estimated with the functions needed to be corrected for the fraction of stones and boulders, which was available on 512 of the sites (Stendahl et al., 2009). The temperature sum was calculated based on latitude and altitude according to Morén and Perttu (1994). Some of the calculations gave negative results for one of the base cations. This can be explained by the fact that the regressions are used for a new dataset, covering a broader range of temperature sums than the original dataset, which illustrates a limitation of the method. The sites where rates were negative were removed, which reduced the database to 445 sites. The estimations apply down to the weathering depth, which means different soil depths on different sites, but normally between 40 and 70 cm in the mineral soil (Mats Olsson, pers. comm).~~

~~4.2 PROFILE~~

~~The latest PROFILE weathering map for Sweden is presented in Akselsson et al. (2016), where weathering rates for the upper 50 cm of the soil (including the organic layer) were modelled based on data from 17,333 Swedish National Forest Inventory (NFI) sites (Fridman et al., 2014), of which the 640 SAFE sites are a subset. PROFILE requires soil input data, as well as climate and deposition data and information about net uptake of nutrients in trees. Mineralogy was derived from an earlier regional study (Akselsson et al., 2007), where total chemistry data from the Geochemical Atlas of Sweden (Andersson et al., 2014) has been used to calculate normative mineralogy for each site. Soil texture and moisture classifications are available on all NFI sites, and were transferred to specific surface area and volumetric water content using translation tables from Warfvinge and Sverdrup (1995). The same dataset as above was used to correct for stones and boulders. Net uptake of base cations and nitrogen was calculated based on tree growth data on the NFI sites. Deposition data from the MATCH model (an average for 2006-2008) was used (Langner et al., 1996). Temperature data from SMHI,~~

~~representing 1981-2010, were taken from Akselsson et al. (2016). The methodology is described more in detail in Akselsson et al. (2007; 2008; 2016).~~

~~4.3 ForSAFE~~

~~Weathering rates to 50 cm depth (including the organic layer) were modelled with the ForSAFE model on the 640 sites in the SAFE database (Alveteg, 2004). Information on soil texture and mineralogy were derived from the database of the Research Infrastructure National Forest Inventory of Sweden (RINFI, Hägglund, 1985). Mineralogy was derived from total elemental analysis translated through the UPPSALA model (Sverdrup et al., 2002). Stones and boulders were taken into account using the same data on contents of stones and boulders as for PROFILE and the depletion method.~~

~~The climatic input data were derived from simulations of historical and future trends, based on the IPCC's A2 story line of emissions (David Rayner, pers. comm.) and downscaled to historical records from the Swedish Meteorological Institute (SMHI). The climate data include monthly information on air temperature, precipitation, photosynthetically active radiation and atmospheric CO$_2$ concentrations.~~

~~Atmospheric deposition data for sulphate, nitrate and ammonium were derived from simulations by the EMEP model (Simpson et al., 2012) according to the emissions history in Schöpp et al. (2003) and the projected emissions following the current legislation of the UN Convention on Long Range Transboundary Air Pollution (LRTAP). Atmospheric deposition data for chloride, calcium, sodium, potassium and magnesium were derived from the MATCH model (Persson et al., 1996).~~

~~4.4 Comparison between regional estimates produced by the three methods~~

~~In order to compare the results from the different methods regionally, a subset of 346 sites where all three methods were applied was used. These were the sites for which all methods had been applied successfully, and where data on stones and boulders were available. For a regional comparison, the sites were divided into seven climate regions, simplified from 19 weather forecast regions used by the Swedish Meteorological and Hydrological Institute (SMHI) (Fig. 3-4). One of the regions, northwestern Sweden, only contained one site, and was therefore excluded from the analysis.~~

The weathering rates for the nutrient base cations Ca, Mg and K varied widely within the regions for all methods, but there were no large systematic differences between the medians or ranges for the different methods (Fig. 4-5). However, PROFILE gave generally somewhat lower weathering rates than ForSAFE and the Depletion method/Total analysis regression approach, with overall medians of 14.3 mekv m^{-2} y^{-1} (PROFILE) and 17.8 mekv m^{-2} y^{-1} (ForSAFE and the Depletion method/Total analysis regression approach).

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For ForSAFE, this difference was most distinct in the northern regions. ~~ForSAFE gave somewhat higher medians than PROFILE for all regions, especially in the northern regions.~~ One explanation regarding the difference between PROFILE and ForSAFE could be differences in the method for estimating mineralogy from total chemistry, where 'possible' minerals have to be set by the modeller. Since qualitative data on mineral contents in soils in most cases are not available at the modelling sites, ~~data must be~~the mineralogy has to be estimated based on a number of assumptions. In the ForSAFE database, limestone seems to have been set as a possible mineral more often than in the PROFILE database, due to differences in the assumptions made. Since even a small amount of limestone has a large effect on weathering rates, the modelled Ca weathering rates we~~a~~re substantially higher at some sites in the ForSAFE results~~database~~.

~~The total analysis regression method gave somewhat higher weathering rates than PROFILE for several of the regions.~~ The~~is~~ ~~was~~ difference between PROFILE and the Depletion method/Total Analysis regression approach was contradictory to the site-level comparisons, where PROFILE generally gave substantially higher weathering rates than the ~~depletion~~ Depletion method (Table 3). This can partly be explained by the site-level comparisons being made for the same depths (50 cm), whereas the regional calculations with the Depletion method/T~~t~~otal analysis regression ~~method~~ approach ~~gives~~ gave the weathering to the maximum weathering depth, which is often more than the 50 cm (including a 10 cm organic layer) used for PROFILE. Methodological differences between the old and the new calculations for the ~~depletion~~ Depletion method/Total analysis regression approach can also be part of the explanation, e.g. concerning how the weathering depth has been defined based on curves of the elemental variation with depth, which is partly a subjective operation. Finally, the fact that the regional calculations combine two methods (D~~d~~epletion method and T~~t~~otal analysis regression approach), whereas the site-level estimates are based only on the D~~d~~epletion method, may contribute to the differences.

In most cases, the width ~~comparison~~ of the weathering rate intervals ~~between regions~~ showed no major differences between the regions. The difference between the 25- and the 75-percentile was generally 10 to 20 meq m^{-2} y^{-1} (Fig. 5).~~.~~ An exception wa~~i~~s region 4, the eastern part of central Sweden, where the corresponding interval~~span~~ was up to 100 mekv m^{-2} y^{-1}~~wider, including much higher weathering rates than the other regions~~. In region 4, lime~~-~~rich soils are common, which can ~~may~~ explain this pattern. Other minor differences were that ~~Smaller differences could be distinguished between the other regions.~~ t~~T~~he weathering ~~ranges~~ rates in the southern regions ~~(5 and 6)~~ were generally on a somewhat higher level than the others (medians: 14-20 meq m^{-2}y^{-1}), whereas ~~the~~ the weathering rates in the western part of central Sweden were~~as~~ towards the lower end~~.~~ (medians: 9-14 meq m^{-2}y^{-1}).

~~Although PROFILE and ForSAFE were based on the same weathering module and the same input database, it is not self-evident that they give similar results. The dynamics of ForSAFE, which e.g. involves dynamic modelling of moisture content in soil instead of a fixed value based on rough field assessments, could lead to differences, as discussed in Kronnäs et al. (in press, this issue). Moreover, different methods for processing input data, e.g. estimating mineralogy from total chemistry, may cause differences (Casetou-Gustafson et al., in review a (this issue)). Despite those differences, the methods gave comparable results, with similar weathering rate levels and geographical patterns. The depletion method, based on a completely different concept, also gave similar results.~~

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3.3 Weathering in a sustainability perspective

Although the difference in weathering rates between methods is large on several sites, a number of general conclusions could be drawn from the comparison with harvest losses. For the stem-only harvesting scenario, the harvest losses were generally at the same level or lower than PROFILE weathering rates (Fig. 6-7). The Depletion method gave generally higher weathering rates than harvest losses at stem-only harvesting in the northern sites, but lower in the southern sites.

In five of the seven spruce forests in southern and central Sweden (the sites to the right in Fig. 6), the harvest losses in the whole-tree harvesting scenario were higher than the weathering rates (5-180%), regardless of the method used to calculate weathering rates (Table 5, Fig. 6). Exceptions were Asa, where the weathering rates from the Budget approach gave a many times higher weathering rate than the other methods, and Gårdsjön, where the weathering rates from most of the methods were of a similar size as the harvest losses. Despite the variation between methods, the results clearly indicate that whole-tree harvesting is not sustainable in the long term in spruce forests in southern and central Sweden, since the weathering rates generally are substantially lower than the base cation losses at whole-tree harvesting.

On the four spruce sites in northern Sweden (to the left in Fig. 6), PROFILE gave 20-130% higher weathering rates than harvest losses after whole-tree harvesting, whereas the Depletion method gave 20-50% lower weathering rates than harvest losses for three of the four sites (Table 5, Fig. 6). The Budget approach in Flakaliden gave 220 % higher weathering rates than the harvest losses after whole-tree harvesting. Despite the difference between the methods, the results clearly indicate that the effects of whole-tree harvesting in spruce forests in northern Sweden are substantially smaller than for spruce forests in southern and central Sweden.

All pine sites where comparisons could be made in this study were situated in northern or central Sweden (Fig. 2; Table 1). For all three sites, the PROFILE weathering rates were substantially higher than the harvest losses, both for the stem-only (150-240%) and whole-tree harvesting scenario (90-170%) (Table 5, Fig. 7). Weathering rates calculated with the Depletion method were of similar size as the harvest losses at whole-tree harvesting, except for Kloten where the weathering rates were 50% lower. Thus, the conclusions about pine forests are similar to those for spruce forests in northern Sweden, that long-term losses are less of a concern, although the variation in weathering rates makes it difficult to say whether the weathering rates are higher or lower than the harvest losses in those forests.

In the above assessment, the extent to which whole-tree harvesting itself affected the weathering rates was not explicitly considered. As an increased forest harvest intensity leads to slightly more acidic conditions, it could be hypothesised that increased intensity leads to an increased proton-promoted dissolution of minerals, thereby providing a feedback mechanism in which increased weathering could partially alleviate the effect on soil acidity and base cation status. However, according to recent HD-MINTEQ modelling in which PROFILE was used to simulate weathering, the weathering rate was largely unaffected by soil solution pH and by the harvesting method used (McGivney et al. (in review). This was explained as being the net result of the opposing effects of pH and dissolved Al on the weathering rate. While a decreased pH itself leads to an increased weathering rate, it also leads to increased levels of dissolved Al, which is a potent weathering ‘brake’, offsetting the pH effect. Another source of uncertainties is the potential effect of whole-tree harvesting on mycorrhiza activity, which is further discussed in section 3.4, and in Finlay et al. (in review).

In the assessments of base cation sustainability, it is important to not only to focus on uncertainties in the actual soil weathering rates. Other important topics are how much of the weathered material the tree roots can reach, the size of the base cation deposition, the uncertainties in the assessment of base cations through harvesting, and how the base cation losses are distributed between soil, biomass and runoff water. The use of a constant and static rooting depth introduces uncertainties in the sustainability assessments, since root depths varies both spatially and temporally, depending on variations in site coditions (Hodge, 2013; Rosengren and Stjernquist, 2004). The base cation deposition in Sweden is assessed to be of similar size as the base cation weathering (Akselsson et al., 2007), but a national survey of total base cation deposition, including dry deposition, is not available, so uncertainties of base cation deposition are large. The uncertainties in the assessments of base cation losses at harvesting can be divided in uncertainties in the amount of biomass extracted and the concentration of BC in biomass (Akselsson, 2005). A sensitivity analysis for Ca showed that the lack of site specific nutrient concentration data was the main source of uncertainties in calculations of harvest losses of Ca, whereas the estimations of biomass available for extraction, and the amount of branches left on the ground, contributed less to the uncertainties (Zetterberg et al., 2014). Finally, the effect of base cation losses in soil is reduced by the fact that the rates of tree growth and leaching decline after whole-tree harvesting, mitigating some of the impacts of harvest on soil base cation status (Zetterberg et al., 2013; Egnell, 2016).

3.4 ~~5~~ Potential for biological weathering

~~Plants play a fundamental role in soil formation, since root activity and decomposing plant material increase weathering rates by producing acidifying substances (H+, organic acids) and ligands that form complexes with metals in the minerals. In addition, uptake of ions released from weathering reduces the likelihood of saturated conditions that retard weathering rates. Many of these effects are mediated by mycorrhizal fungi. Biological weathering often takes place in conjunction with physical and chemical processes but there is still disagreement over the extent of its quantitative contribution to overall weathering (Finlay and Clemmensen, 2017; Leake and Read, 2017; Smits and Wallander, 2017). Below, a description of how biological weathering is presently represented in the PROFILE/ForSAFE models is given, followed by a discussion about biological weathering, the role of mycorrhizal fungi and potential shortcomings in the PROFILE/ForSAFE models. A more thorough description of the state of knowledge and a more comprehensive discussion can be found in the article by Finlay et al. (in review, this issue).~~

~~Chemical elements are released from minerals to a dissolved form following four pathways in the PROFILE/ForSAFE models: the first dependent on soil solution H+ concentrations, the second on water availability, the third on the partial CO_2 pressure, and the fourth on the concentrations of dissolved organic carbon (DOC) (Sverdrup and Warfvinge, 1993). The four weathering pathways are chemical (the dismantling of mineral matrices by charged or dissolving particles to produce free~~

~~elements), but their drivers are strongly dependent on biological activity in the soil. Soil solution H^+ is determined by the charge balance resulting from uptake, ion exchange, mineralisation of organic matter (solid and dissolved), and hydrological transport, all of which are affected by biological activity. Water availability is directly controlled by water uptake. The partial pressure of dissolved CO_2 stems from decomposition and hydrological transport. Finally, DOC is directly and indirectly produced by plants.~~

~~The transition state theory governing the weathering kinetics in PROFILE/ForSAFE dictates that the net weathering rates should decline towards zero near equilibrium. This is represented in the model by retardation factors that increase in strength with the concentrations of the weathering products (Erlandsson et al., 2016). These concentrations are in turn dependent on biological activity, such as uptake reducing nutrient base cation concentrations or the mobilisation of aluminium through biological acidification.~~

~~Although the weathering process is strongly affected by biological processes in the current generation of the PROFILE/ForSAFE family of models, the models still fail to capture the biological feedback mechanisms in their entirety. Carbon allocation for example is still rudimentary in the model, and more elasticity in carbon allocation is needed to capture the empirical observations. Exudation, another example, seems to be a more active process in response to nutrient status than is currently assumed in the models.~~

~~The possible roles of fungi in biological weathering in boreal forests were summarised by Finlay et al. in 2009, and have been the subject of many subsequent studies. Prior to the QWARTS project the widespread occurrence of tubular pores, 3-10 µm in diameter, was demonstrated in weatherable minerals in podzol surface soils and shallow granitic rock under European coniferous forests (van Breemen et al., 2000; Jongmans et al., 1997; Landeweert et al., 2001). Fungal hyphae were found occupying some of these pores and it was speculated that they could be formed by the weathering action of hyphae (and possibly associated bacteria), releasing organic acids and siderophores. The aetiology of pore formation has been questioned, however, with some authors claiming that the observed pores are of abiotic origin (Sverdrup, 2009), and their quantitative contribution to total weathering rates has been calculated to be negligible (Smits et al., 2005). This means either that fungal weathering is negligible, or that tunnel formation reflects only a small proportion of the total weathering effect of the fungi. The endolithic biosignatures of rock-inhabiting microorganisms can be distinguished from purely abiotic microtunnels (McLoughlin et al., 2010), and the biomechanical mechanisms used by fungi to penetrate rock have received increasing attention. Fungal-mineral attachment, biomechanical forcing, and altered interlayer spacing associated with depletion of potassium from biotite by a mycorrhizal fungus have been demonstrated (Bonneville et al., 2009). Extensive mineral surfaces are accessible for microbial colonisation, and atomic force microscopy has been used to demonstrate nanoscale alteration of surface topography and attachment and deposition of organic biolayers by fungal hyphae (McMaster, 2012; Gazze et al., 2013; Saccone et al., 2012). These nanoscale mineral-fungal interactions undoubtedly occur, but their quantitative significance has yet to be revealed.~~

~~There is an extensive literature on the role of fungi as biotic agents of geochemical change (Gadd, 2013a, b; 2017). We know that ectomycorrhizal fungi can allocate carbon selectively to different minerals (Rosling et al., 2004; Smits et al., 2012). The~~

~~latter (laboratory) study demonstrated that, when P was limiting, 17 times more plant-derived C was allocated to ectomycorrhizal fungal mycelium of Paxillus involutus colonising apatite than to mycelium colonising quartz, as well as that fungal colonisation of the substrate increased the release of P by a factor of almost three. Grain-scale 'biosensing' (differential colonisation) of different minerals by the same fungus has also been demonstrated by Leake et al. (2008) and oxalate secretion by this fungus also appears to be mineral-specific (Schmalenberger et al., 2015). These, and other, similar laboratory experiments suggest that plant-fungal-mineral interactions are tightly coupled, and that distinct, local weathering environments exist.~~

~~However, there is disagreement over the extent to which the observed laboratory-scale processes contribute to soil-scale mineral dissolution rates and field processes. One view is that the coevolution of fungi and plants has enabled them to exert increasing influence as biogeochemical engineers. The ubiquity and significance of lichens as pioneer organisms in the early stages of mineral soil formation, and as a model for understanding weathering in a wider context, are well understood (Banfield et al., 1999). It is argued that, during evolution, successive increases in the size of plant hosts and the extent of substrate colonisation by their fungal symbionts (Taylor et al., 2009; Leake and Read, 2017) have enabled them to have larger effects as biogeochemical engineers, affecting the cycling of nutrients and C at an ecosystem and global level. These ideas are based on observations of alteration of silicate surfaces in the proximity of roots and associated mycorrhizal fungi of different trees exposed to atmospheres with different levels of CO_2 (Quirk et al., 2012; 2014).~~

~~It is accepted that ectomycorrhizal fungi access and degrade organic nitrogen sources (Lindahl and Tunlid, 2015), and soil carbon storage has been shown to be greater in ecosystems dominated by ectomycorrhizal plants than in systems dominated by other types of mycorrhiza (Averill et al., 2014). Transfer of increasing amounts of photosynthetically derived carbon to ectomycorrhizal fungi and improved colonisation of mineral substrates during evolution of plants (Quirk et al., 2012; 2014) is consistent with the idea that weathering of silicate minerals and sequestration of C into ocean carbonates has led to drawdown of global CO_2 levels during the past 100 M years (Taylor et al., 2011). Enhanced weathering of minerals applied to different ecosystems has now been suggested as a global CO_2 reduction technology (Taylor et al., 2016; Beerling et al., 2018).~~

~~However, Smits and Wallander (2017) pointed out that there is no clear evidence that processes observed at the laboratory scale play a significant role in soil-scale mineral dissolution rates. Furthermore, many of the theories about evolutionary development of weathering have been elaborated in the absence of detailed molecular identification of the microorganisms involved. Detailed studies of the liquid chemistry of local weathering sites at the micrometre scale, together with up-scaling to soil-scale dissolution rates, are advocated, and the authors suggest that future research should focus on whole-ecosystem dynamics, including the behaviour of soil organic matter, and that early-stage primary succession ecosystems on low reactive surfaces, such as fresh granites, should be included. Smits and Wallander (2017) also recommend the use of stable isotopes by choosing minerals and soils with distinct isotope ratios.~~

~~Most studies of ectomycorrhizal influence on weathering rates have been done over short periods in laboratory settings, and there is no clear evidence that processes observed at laboratory-scale play a significant role in soil-scale mineral dissolution~~

~~rates. In an attempt to span a longer time-scale for biological weathering studies, Smits and Wallander (2017) used a vegetation gradient from bare soil, via sparse grass to Norway spruce forest in a natural, lead-contaminated area in Norway. This gradient had probably been present since the last glaciation, and made it possible to study long-term effects of vegetation on apatite weathering in moraine material deposited at the end of the last glaciation. The presence of vegetation had a strong stimulatory effect on apatite weathering, mainly because of the acidifying effect of plant growth, which was probably mediated through activity of the associated ectomycorrhizal fungi.~~

~~This effect of plant growth on weathering of apatite, through changes in soil pH, is probably captured sufficiently in weathering models like PROFILE/ForSAFE under situations when P is not in short supply and nitrogen is limiting tree growth, which is the general case in boreal forests. However, under conditions of low P availability, intensive colonisation of apatite particles by EMF has been seen both in laboratory experiments (Rosling et al., 2004; Smits et al., 2012) and in the field (Bahr et al., 2015, Rosenstock et al., 2016, Almeida et al., in press). Weathering of apatite may be enhanced under these conditions through biomechanical mechanisms and accumulation of weathering agents in localised microenvironments that are colonised by EMF but separated from the soil solution as explained below.~~

~~The nutrient status of the forest is probably of great importance when estimating the role of biota in mineral weathering. Belowground carbon allocation is usually increased under N and P limitation, but reduced under K and Mg limitation (Ericsson 1995). For this reason, weathering of K- and Mg-containing minerals may not be enhanced under K and Mg limitation, since root and mycorrhizal activity is expected to decline under these conditions. Support for this view was found by Rosenstock et al., (2016), who studied EMF colonisation of biotite and hornblende under varying K and Mg conditions in Norway spruce forests in the Czech Republic.~~

~~Laboratory studies of the capacity of different fungi to mobilise P and base cations from granite particles (conducted within QWARTS) (Fahad et al., 2016) suggest that some ectomycorrhizal fungi can mobilise and accumulate significantly higher concentrations of Mg, K and P than non-mycorrhizal fungi. The mycorrhizal fungi fractionate Mg, discriminating against heavier isotopes, and we found a highly significant inverse relationship between δ^{26}Mg tissue signatures and mycelial concentration of Mg. This provides a theoretical framework for testing hypotheses about fungal weathering of minerals in future experiments.~~

~~If active mobilisation and uptake of lighter ^{24}Mg isotopes results in relative enrichment of heavy Mg isotopes left in soil solution and soil, this should be evident in areas of active weathering. Mesocosm experiments conducted within the QWARTS project (Mahmood et al., unpublished), employing a gradient of increasing organic matter depletion to simulate progressively more intense biomass harvesting, revealed significant and successive enrichment of ^{26}Mg signatures in the soil solution in the B horizon, associated with increased availability of organic matter and resultant increases in plant and fungal biomass. No such enrichment was found in other horizons or in systems without plants (and therefore without mycorrhizal fungi). This suggests that significant biological weathering of Mg takes place in the B horizon, driven by higher plant biomass that enables improved carbon allocation to the fungal mycelium and also constitutes a larger sink for uptake of mobilised base cations.~~

~~Although the experiments provide strong support for the idea of biologically driven mobilisation of Mg from B horizon mineral soil, the process was not sufficient to maintain tree growth in systems severely depleted of organic matter. Mycorrhizal fungi play a central role in mobilising N and P from organic substrates (Lindahl and Tunlid, 2015) and when these are depleted, N and P limit tree growth resulting in reduced C supply to the mycorrhizal mycelium and reduced capacity for mobilisation of base cations from the mineral horizons. Although mobilisation of Mg from the B horizon was sufficient to support increased biomass production in systems supplied with extra organic material, it was not sufficient to sustain plant growth when organic material was most depleted and insufficient N was available. The results of these experiments are therefore consistent with the predictions of modelling that, under intensive forestry with removal of organic residues, base cation supply will not be sustainable in the long term.~~

Biological weathering often takes place in conjunction with physical and chemical processes but there is still disagreement over the extent of its quantitative contribution to overall weathering (Finlay and Clemmensen, 2017; Leake and Read, 2017; Smits and Wallander, 2017). Insufficient representation of biological weathering in weathering models such as PROFILE, and its effects of weathering rate uncertainties, has been frequently discussed (Finlay et al., 2009). Below, a description of how biological weathering is presently represented in the PROFILE/ForSAFE models is given, followed by a discussion about potential shortcomings in the light of the latest research about biological weathering. A more thorough description of the state of knowledge and a more comprehensive discussion can be found in the article by Finlay et al. (in review).

Although the four weathering pathways, upon which PROFILE is built (the reaction with H^+, CO_2 and DOC) are chemical (the dismantling of mineral matrices by charged or dissolving particles to produce free elements), their drivers are strongly dependent on biological activity in the soil. Soil solution H^+ is determined by the charge balance resulting from uptake, ion exchange, mineralisation of organic matter (solid and dissolved), and hydrological transport, all of which are affected by biological activity. Water availability is directly controlled by water uptake. The partial pressure of dissolved CO_2 stems from root and root symbiont respiration, decomposition and hydrological transport. Finally, DOC is directly and indirectly produced by plants (Sverdrup and Warfvinge, 1993). The transition state theory, governing the weathering kinetics in PROFILE/ForSAFE, dictates that the net weathering rates should decline towards zero near equilibrium. This is represented in the model by retardation factors that increase in strength with the concentrations of the weathering products (Erlandsson et al., 2016). These concentrations are in turn dependent on biological activity, such as uptake reducing nutrient base cation concentrations or the mobilisation of aluminium through biological acidification.

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Although the weathering process is strongly affected by biological processes in the current generation of the PROFILE/ForSAFE family of models, the models still fail to capture the biological feedback mechanisms in their entirety. The possible roles of fungi, especially ectomycorrhizal fungi, in biological weathering in boreal forests were summarised by Finlay et al. (2009), and have been the subject of many subsequent studies (Finlay et al., in review). These fungi can acidify their surrounding environment and release organic acids and siderophores, which may enhance weathering. They can also exert biomechanical forcing and alter interlayer spacing associated with depletion of potassium from biotite (Bonneville et al

2009). Furthermore, recent work with atomic force microscopy has demonstrated nanoscale alteration of surface topography of minerals and attachment and deposition of organic biolayers by fungal hyphae. Many fungal hyphae produce extracellular polysaccharides (EPS) at their hyphal tips, providing an interface that ensures intimate contact between the hyphae and mineral substrates. The contact area between hyphae and mineral surfaces is increased by EPS haloes (Gazze et al., 2013), and many fungal exudation products such as organic acids and siderophores may be released into polysaccharide matrices (Flemming et al., 2016) in close proximity to mineral surfaces. Here, they are effectively isolated from the bulk soil solution and may be protected from microbial decomposition by antibiotic compounds also produced by the fungi. This is in contrast to the assumption in the models that soil solution is homogeneous at any given depth, not discerning bulk solution from the said EPS haloes. This may increase the effective concentrations of organic weathering agents at sites of active weathering, and structure the bacterial communities associated with particular mycorrhizal fungi (Marupakula et al., 2016).

The potential mechanisms for biological enhancement of mineral weathering and the current debate about the importance of these processes for overall weathering are discussed in detail by Finlay et al. (in review). The biological activity of symbiotic ectomycorrhizal fungi and the evolution of their interactions with their tree hosts have led to systems that are highly adapted to efficient recycling of plant nutrients from organic matter, as well as release of base cations from mineral substrates through weathering. Ectomycorrhizal mobilization of N and P through decomposition of organic residues is dependent on carbon supplied from tree hosts. Mycorrhizal weathering of minerals is also dependent on carbon supply from trees and on-going experiments (Finlay et al., in review) suggest that depletion of organic substrates (containing N) will restrict tree growth and therefore also reduce carbon supply to ectomycorrhizal fungi colonizing mineral substrates, with concomitant, negative effects on base cation release from biological weathering. Existing models are therefore probably sufficient to give guidelines about sustainable forestry, including the prediction that, under intensive forestry with removal of organic residues, base cation supply will not be sustainable in the long term. However, the biological feedbacks during transition from one state to another may not be fully covered by the models. For instance, a forest exposed to N deposition may pass from N limitation to limitation by another nutrient, which may have consequences for belowground carbon allocation. Ectomycorrhizal fungi are dependent on carbon supplied from their host plants and can be expected to exert a stronger effect on mineral weathering if they have access to more carbon, which may influence mobilization of nutrients that are limiting. A possible way to include the biological effects on mineral weathering would be to better describe belowground carbon allocation in the models. Enhanced weathering rates of apatite has been seen when host trees suffer from P shortage, which is known to enhance belowground carbon allocation (Smits et al, 2012). More elasticity in carbon allocation in the models is needed to capture these empirical observations. Furthermore, carbon allocation will also regulate exudation from roots and associated, which is another process involved in mineral weathering that is not covered in the models.

3.5~~6~~ Implications of improved model descriptions of base cation exchange and aluminium complexation

~~**Implication of higher resolution of chemical reactions in weathering modelling**~~

Aluminium (Al) and base cation concentrations are the primary weathering brakes in unsaturated soil (Warfvinge and Sverdrup, 1992). Higher concentrations of these elements have a negative effect on the dissolution rates of the minerals containing the elements (Sverdrup et al., in review~~, this issue)~~). It is therefore imperative to correctly simulate the concentrations of Al and base cations in the soil solution.

Different soil chemical models simulate the dynamics of inorganic Al and base cations in different ways. These can be classified into two categories: 1. Simpler ion-exchange equations (e.g. Gaines-Thomas or Gapon) that conceptualise sorption and desorption of Al3^{+}, H^{+} and base cations as a series of ion-exchange reactions, and 2. More advanced ~~'state-of-the-art'~~ organic complexation models such as WHAM, NICA-Donnan or SHM (Tipping, 2002; Kinniburgh et al., 1999; Gustafsson, 2001) that treat organic matter as the main cation sorbent, where proton dissociation over a wide pH range drives complexation and exchange of Al and base cations.

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In general, the use of organic complexation models to simulate base cation and Al dynamics is strongly supported by empirical evidence (e.g. Tipping, 2002) ~~.~~ but f~~F~~or a long time, the simpler ion-exchange equations~~former type of model approach~~ ha~~s~~ves been more widely used in popular biogeochemical models such as MAGIC~~C~~, PROfFILE and ForSAFE. However, ~~already~~ in 1996, the CHUM model was introduced, which incorporates a version of WHAM (Tipping, 1996), and today SMARTml and HD-MINTEQ provide additional examples of (bio)geochemical codes that employ organic complexation models (Bonten et al., 2011; Löfgren et al., 2017).

Gustafsson et al. (2018~~, this issue~~) investigated the implications of using the two model approaches on the dynamics of Al, base cations and acidity. Overall, the two model approaches provided the same type of response to changes in input chemistry, implying that in many cases, there may be a rather limited benefit from using organic complexation models when calculating weathering rates. However, ~~However, the exchange models tested stress the importance of including H^{+} in the exchange dynamics to be able to account for the effects of rapid changes in proton concentrations, such as in the case of sea spray events (Gustafsson et al., 2018 (this issue)).~~

~~A~~although~~, for the most part,~~ these results suggest that the current model setup in e.g. ForSAFE may be sufficient in many cases, certain differences remain between the two categories of models. The Gaines-Thomas and Gapon exchange equations produce a relatively stronger buffering of soil solution pH over a relatively narrow pH range. Together with the general oversimplification of the cation binding process this also causes the ion-exchange equations to overestimate the historical levels of exchangeable base cations (Gustafsson et al., 2018~~ (this issue)~~). Consequently, it may be necessary to include organic complexation under such conditions as prolonged or substantial changes in acidic input, such as in the case of sea spray events. ~~.~~Not explicitly simulating organic complexation may require additional coefficients that account for temporal changes in cation selectivity to correctly predict pH, base cations and Al, thereby entailing more uncertainty. Excluding organic complexation can bring into question the ability of biogeochemical models to predict the effect of large changes in acidic input on weathering rates (Gustafsson et al., 2018).

~~7 Weathering in a sustainability perspective~~

~~The effect on policy applications of the variability in weathering rates among different methods depends on the context in which they are to be used. A few decades ago, weathering rates were discussed in the context of acid rain and critical loads of acid deposition. Then the weathering rates were mainly compared with deposition, which at that time (1970-1980) was much greater than estimated weathering. Now, when the focus has shifted to sustainable forest management, the interesting comparison is between base cation weathering rates and base cation losses through harvesting. The effect of uncertainties in this context is dependent on site properties and the size of other base cation flows.~~

~~Budget calculations, with weathering and deposition as inputs and harvest losses and losses through leaching as outputs, are often used in sustainability assessments (Akselsson et al., 2007; Hultberg et al., 2004). Simplified calculations, based only on weathering and harvest losses, are also often used (Olsson et al., 1993; Klaminder et al., 2011; Stendahl et al., 2013). Weathering rates that are substantially higher than the harvest losses indicate less risk for depletion of base cations in soils than if weathering rates are lower than harvest losses.~~

~~Harvest losses were estimated and compared with weathering rates for the sites in Table 1 where weathering has been calculated for the root zone (defined here as <0.7 m) and for which data on site index could be found, in spruce forests (Fig. 5) and pine forest (Fig. 6). The calculations of harvest losses were based on the stand index of the forest and generalised densities and nutrient base cation concentrations in different tree parts. Two types of harvesting were considered, conventional stem-only harvesting and whole-tree harvesting, where, in addition to the bole (stem), tops and branches are removed for biofuel. It was assumed that, in whole-tree harvesting, all stems and 60% of the branches were harvested, and that 75% of the needles were removed with the harvested branches. The methodology is more thoroughly described in Akselsson et al. (2007).~~

~~Although the difference in weathering rates between methods is large on several sites, a number of general conclusions can be drawn. For the stem-only harvesting scenario, the harvest losses were generally at the same level or lower than PROFILE weathering rates. The depletion method gave generally higher weathering rates than harvest losses at stem-only harvesting in the northern sites, but lower in the southern sites.~~

~~In most of the spruce forests in southern and central Sweden (the sites to the right in Fig. 5), the harvest losses in the whole-tree harvesting scenario were substantially higher than the weathering rates, regardless of the method used to calculate weathering rates. Exceptions were Asa, where the weathering rates from the mass balance calculations gave a very high weathering rate, higher than the harvest losses, and Gårdsjön, where the weathering rate for most of the methods was similar to the harvest losses. On some of the sites in these areas the variation in weathering rates between methods was large, for~~

~~example in Bodafors and Skånes Värsjö, but both methods still led to the same conclusions regarding the sustainability of harvests.~~

~~On the four spruce sites in northern Sweden (to the left in Fig. 5), PROFILE gave higher weathering rates than harvest losses after whole-tree harvesting, whereas it was the other way around for the depletion method. The mass balance method in Flakaliden gave very high weathering rates, similar to those at Asa. However, according to the discussion above, these mass balance calculations do not give reliable estimates of weathering rates, since they most likely also include release from other base cation pools (Casetou-Gustafson et al., in review b (this issue)).~~~~The differences between the methods made it more difficult to draw conclusions, as for the spruce sites in northern Sweden. However, the results indicate that the effects of whole-tree harvesting in pine forests in central and northern Sweden are substantially smaller than for spruce forests in southern and central Sweden.~~

~~All pine sites where comparisons could be made in this study are situated in northern or central Sweden. For all three sites, the PROFILE weathering rates were substantially higher than the harvest losses, both for the stem-only and whole-tree harvesting scenario. Weathering rates calculated with the depletion method were similar to the harvest losses at whole-tree harvesting, except for Kloten where the weathering rates were lower (Fig. 6).~~ ~~The differences between the methods made it more difficult to draw conclusions, as for the spruce sites in northern Sweden. However, the results indicate that the effects of whole-tree harvesting in pine forests in central and northern Sweden are substantially smaller than for spruce forests in southern and central Sweden.~~

~~In the above assessment the extent to which whole-tree harvesting itself affected the weathering rates was not explicitly considered. As an increased forest harvest intensity leads to slightly more acidic conditions, it could be hypothesised that increased intensity leads to an increased proton-promoted dissolution of minerals, thereby providing a feedback mechanism in which increased weathering could partially alleviate the effect on soil acidity and base cation status. However, according to recent HD-MINTEQ modelling in which PROFILE was used to simulate weathering, the weathering rate was largely unaffected by soil solution pH and by the harvesting method used (McGivney et al., in review (this issue)). This was explained as being the net result of the opposing effects of pH and dissolved Al on the weathering rate. While a decreased pH itself leads to an increased weathering rate, it also leads to increased levels of dissolved Al, which is a potent weathering 'brake', offsetting the pH effect.~~

~~Despite the uncertainties, the results clearly indicate that whole-tree harvesting is not sustainable in the long term in spruce forests in southern and central Sweden, since the weathering rates are much lower than the base cation losses at harvest. In spruce forests in northern Sweden and in pine forests, long-term losses are less of a concern, although the uncertainties in weathering rates make it difficult to say whether the weathering rates are higher or lower than the harvest losses in those forests.~~

~~When discussing base cation sustainability, it is important to not only focus on uncertainties in the actual soil weathering rates. Other important topics are how much of the weathered material the tree roots can reach, the size of the base cation deposition, and how the base cation losses are distributed between soil, biomass and runoff water. The base cation deposition~~

~~in Sweden is assessed to be of similar size as the base cation weathering (Akselsson et al., 2007), but a national survey of total base cation deposition, including dry deposition, is not available, so uncertainties of base cation deposition are large. The rate of tree growth and leaching can decline after whole-tree harvesting, mitigating some of the impacts of harvest on soil base cation status (Zetterberg et al., 2013; Egnell, 2016). Currently there is even less research on weathering rates in relation to root depth, base cation deposition and alterations in base cation flows as pools change, than on weathering.~~

3.6~~8~~ ~~Future research~~Prospects for method development

Since quantifying uncertainties for weathering rates is difficult, the use of multiple methods is often proposed as a way of increasing the robustness of weathering rate estimates. However, the number of available methods is low, and all are burdened with different types of limitations and uncertainties. In the review of weathering studies in this paper, only six locations could be found where at least three methods had been implemented and where the criterion of the same depth was fulfilled. Thus, the recommendation in Futter et al. (2012), that at least three independent methods should be used to quantify weathering rates on a site for sustainability assessments, is unrealistic.

Still, comparisons between weathering rates from different approaches for the same sites, and continuous development of the different approaches, will contribute to more robust sustainability assessments. In the next sections, main uncertainties and development potential related to process descriptions and input data for PROFILE/ForSAFE are discussed, followed by uncertainties and potential development areas for the Depletion method/Total analysis regression approach and the Budget approach.

~~By far the most widely used, and most evaluated, method for estimating weathering rates for soils in Sweden is the PROFILE model. In this paper, as well as in Kronnäs et al. (in press, this issue), weathering rates from the dynamic model ForSAFE, which contains the same kinetic equations as PROFILE, have been compared with PROFILE results, leading to the conclusion that the two models produce similar weathering rates on average, as long as the hydrology input to PROFILE is similar to the modelled hydrology in ForSAFE (Kronnäs et al., in press, this issue). However, ForSAFE offers the opportunity to dynamically model weathering rates, and their variations within and between years, which is essential for sustainability assessment in a future with a changing climate and management intensity. A number of different research areas where further research is needed have been identified in the QWARTS programme, regarding development of those two models and reducing uncertainties in input data, as well as further evaluations and comparisons with other weathering estimates.~~

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~~8.1~~3.6.1 ~~Model development~~PROFILE/ForSAFE – Process descriptions~~: Biological weathering~~

By far the most widely used, and most evaluated, method for estimating weathering rates for soils in Sweden is the PROFILE model. The successful testing of weathering rate modelling with ForSAFE (Kronnäs et al., 2019) in QWARTS, will be the starting point for more studies on weathering dynamics, using ForSAFE. To better represent ectomycorrhizal fungi processes in the PROFILE/ForSAFE models, and thus reduce uncertainties in modelled weathering rates, three main improvements need to be made: (1) the EPS microenvironments, described in section 3.4, need to be determined in field and considered in models, (2) methods to distinguish between roots and mycorrhizal hyphae need to be developed, to be able to better represent the process of nutrient uptake and translocation towards the plant root and (3) more elasticity in carbon allocation in the models is needed, to be able to better describe the carbon availability for fungi, and to represent the regulation of exudation from roots and associated hyphae.

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The uncertainties in the simplified description of base cation exchange and aluminium complexation were generally small, according to studies in QWARTS (Gustafsson et al., in review). ~~Many fungal hyphae produce extracellular polysaccharides (EPS) at their hyphal tips, providing an interface that ensures intimate contact between the hyphae and mineral substrates. The contact area between hyphae and mineral surfaces is increased by EPS haloes (Gazze et al., 2013), and many fungal exudation products such as organic acids and siderophores may be released into polysaccharide matrices (Flemming et al., 2016) in close proximity to mineral surfaces. Here, they are effectively isolated from the bulk soil solution and may be protected from microbial decomposition by antibiotic compounds also produced by the fungi. This may increase the effective concentrations of organic weathering agents at sites of active weathering, and structure the bacterial communities associated with particular mycorrhizal fungi (Marupakula et al., 2016).~~

~~Bacteria associated with ectomycorrhizal fungi may have a significant influence on mobilisation of different nutrients (Calvaruso et al., 2013). Although weathering at the mineral surface is determined by chemical reactions (chemical weathering), the conditions and agents involved in these reactions are often derived from biological activity and may be defined as biological weathering.~~

~~Existing models that explicitly simulate mineral weathering use soil solution chemistry to determine the weathering rates (Erlandsson et al., 2016). It remains challenging to consider the EPS microenvironments described above in models, as there is a lack of knowledge of the processes determining the former. This difficulty starts in the empirical description of the difference between bulk soil solution and micropore EPS chemistry. Extraction methods using high speed centrifugation may remove EPS from micropores and hyphal interfaces but the resulting bulk concentrations of weathering agents will not reflect those at active sites of weathering. The implication of excluding the said difference between EPS micropore chemistry and soil solution chemistry on weathering rates remains unclear.~~

~~Active uptake of weathering products by fungal hyphae, followed by translocation towards the plant root, will prevent their accumulation at sites of weathering. Mineral elements mobilised by fungal hyphae may remain within the fungal mycelium for different lengths of time before becoming available for plant uptake, and this may represent an important pool of base cations to be included in models. Currently, the active uptake process in the PROFILE/ForSAFE models does not distinguish between roots and mycorrhizae, treating both as a lumped uptake organ. Since both minerals and organic residues contain ectomycorrhizal fungal cycle nutrients, it is imperative that better methods are developed to distinguish between these two sources.~~

~~The stable isotope fractionation patterns of ectomycorrhizal fungi, shown by Fahad et al. (2016) to involve discrimination against heavier isotopes of Mg, provide a useful tool for use in future studies. They can be applied in field situations but further information about isotope fractionation patterns in organic and inorganic substrates is needed, since it is important to distinguish between the de novo supply of elements supplied via weathering and re-circulation of elements via decomposition of organic residues by both mycorrhizal and saprotrophic fungi~~

~~8.2 Model development: Higher resolution chemical reactions~~

~~The comparisons performed in QWARTS indicated that adding a higher resolution description of aluminium complexation and cation exchange reactions to ForSAFE generally led to small effects on long-term chemical dynamics of Al and base cations, which induces small effects on modelled weathering rates. The strongest effect was seen when replacing the ion-exchange equations to describe base cation dynamics with an organic complexation model such as SHM, whereas the replacement of the gibbsite model with more sophisticated model descriptions for Al mattered less (Gustafsson et al., 2018 (this issue)). The effects were rather small, except when large pH fluctuations occur in the data, caused by large changes in acid input. It was concluded that other factors such as uncertainties in deposition and uptake values, as well as the calibration procedure, are likely to be of larger importance for the model performance (Gustafsson et al., 2018 (this issue)).~~ However, we note that a modification would be desirable concerning (1) long-term simulations over hundreds of years when large changes occur in the chemical drivers, and (2) sites experiencing frequent or strong sea salt episodes causing large changes in the chemical composition of the influent water.

~~Due to the significant reduction in execution speed caused by the introduction of organic complexation models, this modification will currently be less prioritised for inclusion in PROFILE/ForSAFE. However, we note that such a modification would be desirable concerning (i) long-term simulations over hundreds of years when large changes occur in the chemical drivers, and (ii) sites experiencing frequent or strong sea salt episodes causing large changes in the chemical composition of the influent water. HD-MINTEQ will be developed further as a scenario tool with a relatively long time steps (weekly).~~

~~8.3 Model development: Implementing weathering brakes~~

~~According to the transition state theory, mineral dissolution rates in PROFILE/ForSAFE are retarded by elevated soil solution concentrations of weathering products, as the equilibrium between the solid and aqueous phases is approached. In the unsaturated zone, weathering retardation is mainly caused by elevated concentrations of base cations and aluminium, called weathering brakes.~~ Based on the~~is~~ assumption that weathering retardation is mainly caused by elevated concentrations of base cations and aluminium, PROFILE/ForSAFE produces reasonable weathering rates in the unsaturated rooting zone (Sverdrup and Warfvinge, 1993; Erlandsson et al., 2016). However, moving into the saturated zone, the strength of the usual weathering brakes fails to slow down the mineral dissolution, which leads to grossly overestimated rates ~~estimates~~ of weathering ~~rates~~ (Stendahl et al., 2013; Erlandsson et al.~~,~~ (in review~~ (this issue)~~). In this environment, soil solution silicate concentrations play a central role in hindering mineral dissolution (Sverdrup et al., ~~, in review (t~~in review~~his issue)~~). For this reason, the kinetics of silicate release from mineral dissolution has been added to the traditional elements, as well as the dynamics of silicate concentrations in the soil solution. Erlandsson et al. (in review~~, this issue~~) tested a prototype of this addition, and the results proved promising in keeping weathering rates within observation levels in the saturated zone, but this is yet to be implanted and tested in PROFILE/ForSAFE.

~~8.4 Model development: Weathering below the root zone – for surface water quality assessments~~

~~Water residence times in the hillslope, and the proportion of old water generating stream flow, need to be more accurately characterised, since this fraction influences delivery of weathering products from within the catchment to the stream (Bishop et al., 2004). This older water has higher concentrations of weathering products. It is not sufficient to predict the rate of weathering within a catchment; the spatial distribution of that weathering in relation to catchment flow pathways and water residence times must also be quantified (Erlandsson et al., in review (this issue)). The possibility that the older water will never even reach the headwater streams most sensitive to acidification, but will appear further downstream in a larger catchment as groundwater subsidy (Ameli et al., 2018), needs to be examined.~~

Lateral flow has recently been included in ForSAFE, and a new version, ForSAFE-2D, has been developed (Zanchi et al., 2016). The model has been evaluated on the basis of hydrological flows and chloride concentrations and transport, with good results. Evaluating the modelled base cation concentrations in surface water highlighted the need for adjusting the weathering brakes (see discussion above about silicate brakes), and also a need to revisit the decomposition process descriptions, thereby validating them for the saturated zone. Further development of ForSAFE-2D has the potential to provide a mechanistic tool for assessing weathering rates also for surface water applications. The importance of correctly defining the flow pathways and residence times for the delivery of weathering products to the surface waters, and the potential value of concentration-discharge relationships for calibrating biogeochemical models was explored by Ameli et al. (2017).

3.6.2 PROFILE/ForSAFE – Input data

Although continuously improved process descriptions are desirable to get more robust weathering estimates, improvements related to input data are more urgent. ~~8.5 Reducing uncertainties in model input data~~

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Mineralogy, specific surface area and soil moisture are of key importance in weathering modelling, but are often burdened with high uncertainties. To reduce input data uncertainties, a focus should be placed on those three parameters. ~~In the A2M model, m~~Mineralogy inputs to PROFILE/ForSAFE are~~is~~ often estimated from total chemistry with the A2M model (Posch and Kurz, 2007)~~based on total chemistry~~, since direct mineralogy measurements are not available on most sites. To accurately estimate a probable mineralogy, not only are good soil chemistry measurements ~~are~~ required, but also information about which minerals can be expected in the soil. In Sweden, ~~three~~ four different geographical mineralogy regions have been used since the 1990s to assign qualitative mineralogy to a site (Warfvinge and Sverdrup, 1995). Casetou-Gustafson et al. (~~in review a, this issue~~2019) compared weathering rates calculated based on three sets of mineralogies: one based on direct measurements of quantitative mineralogy, one based on normative modelling with A2M using direct measurements of qualitative mineralogy, and one based on normative modelling with A2M using data from the regions mentioned above. ~~The results gave weathering rates for both normative methods that were close to the weathering rates based on directly measured mineralogy.~~ It could not be concluded that the A2M runs based on direct measurements of qualitative mineralogy~~the normative mineralogy based on the regions~~ gave better~~worse~~ results. Although these results strengthen the credibility for the normative mineralogy regions, Casetou-Gustafson et al. (~~in review a, this issue~~2019) recommend continued work to reduce uncertainties related to mineralogy, mainly by revisiting and, if appropriate, updating mineral rate coefficients. More comparisons of weathering rates from normative mineralogies based on generalised and site-specific quantitative mineralogy are needed, to adequately assess whether the regional divisions need to be revised and refined in order to further reduce the uncertainties in the mineralogy estimates.

~~A2M~~A2M gives as output a multidimensional space of solutions, all of which have the same probability. Often, the centre point of the space is used for weathering calculations. However, the span can be quite broad, which leads to uncertainties in the calculated weathering rates (Casetou-Gustafson et al., ~~in review a (this issue))~~2019). Future research focusing on constraints that could help to narrow the space of possible solutions that A2M creates, e.g. based on the grain size distribution, could reduce those uncertainties.

Minerals are assumed to be evenly distributed among grain sizes in PROFILE and ForSAFE. The effect of this assumption has not been fully analysed. The most obvious example showing that minerals are not evenly distributed among grain sizes is clay minerals, which are found in the clay fraction. The extremely high surface area of clays leads to very high base cation weathering rates when the clay fraction is high, although the content of base cations is low. Due to this, Phelan et al. (2014) introduced a correction factor. A thorough analysis of all grain size fractions can help to further refine these methods.

The surface area of soils is often calculated with regressions based on old BET measurements ~~-~~(Warfvinge and Sverdrup, 1995). The regressions reveal that the uncertainties are large. ~~The uncertainties could most likely be reduced through~~ ~~r~~Revisions of the regressions, based on a larger data material, could reduce the uncertainties~~, using modern technology~~. The soil moisture is one of the most important factors ~~in weathering modelling~~ that introduces large uncertainties in the results, both in PROFILE where it is an input (Rapp and Bishop, 2003), ~~-~~and in ForSAFE where it is modelled based on hydrological parameters (~~-~~Kronnäs et al., ~~(in press, this issue~~2019). ~~applied both PROFILE and ForSAFE on two sites, and used a rough assessment of soil moisture as input data for PROFILE, whereas soil moisture was modelled by ForSAFE based on soil properties and precipitation. In both those cases, the modelled soil moisture was relatively close to the rough assessment of soil moisture, but the difference was bigger for one of the sites, which could also be seen in the difference in modelled weathering rates.~~ Improved input data quality for soil moisture would substantially reduce uncertainties in

PROFILE and, even more importantly, soil moisture modelled by ForSAFE needs to be evaluated, and the sensitivity to soil input data needs to be examined.

~~8~~3.6.3~~.6 Comparison between modelled weathering and other estimates of weathering~~The Depletion method and the Total analysis regression approach

Next to the PROFILE model, the ~~depletion~~ Depletion method is the most used method in Sweden, often in combination with the ~~total~~ Total analysis regression ~~method~~approach. ~~This method is relatively easy to perform on new sites, although detailed data of the soil profile is needed. As for PROFILE, the method requires soil sampling and total elemental analysis of the soil. In an undisturbed soil profile, if it can be assumed that most of the soil was developed after the last glaciation as well as that zirconium does not weather, the depletion method should give an accurate measure of the average weathering since the last glaciation.~~

To further evaluate the accuracy of results from the ~~depletion~~ Depletion method, as a proxy for the weathering rates of today, the reliability of the assumptions needs to be further evaluated, and the relationship between the average weathering rate since the last glaciation and today's weathering rate needs to be assessed. The latter can be done by performing ForSAFE modelling on a site where the ~~depletion~~ Depletion method has been applied. A similar exercise has been done with the SAFE model (Warfvinge et al., 1995), but the inclusion of tree growth and decomposition in ForSAFE can be expected to improve the results. Furthermore, standardised methods for setting the weathering depth based on the elemental content curve and for the analysis of fulfilment of the requirements in the soil profile, would enable objective and comparable estimates~~a manual for the depletion method needs to be developed~~, including requirements that must be fulfilled for soil profiles to be regarded as undisturbed. The Total analysis regression approach will give more robust results if more Depletion method estimates are available for the regressions. ~~Standardised methods for setting the weathering depth based on the elemental content curve in the soil profile would enable objective and comparable estimates.~~

3.6.4 The Budget approach

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~~The budget method requires more measurements than the depletion method.~~ Different applications of the ~~budget~~ Budget approach~~method~~ handle the distinction between sources of base cations in the soil in different ways, affecting the uncertainties in the estimated weathering rates~~, as explained above~~. T~~Also, t~~he uncertainties in base cation deposition add to the overall uncertainties~~contains large uncertainties~~. ~~In the compilation of weathering rates in this paper, the most extreme outliers came from the budget method, which can be explained by the fact that other sources than weathering are included (Rosenstock et al., in review (this issue)).~~ For a fair comparison between weathering rates from the ~~budget~~ Budget approach~~method~~ and from other methods, ways to distinguish between different sources and sinks need to be further developed.

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An advantage of the B~~b~~udget ~~method~~ approach using the Sr isotope ratio is that it can distinguish between weathering and release from the exchangeable pool. As for all budget approaches, deposition and leaching measurements are required as inputs. The few comparisons made in this study show promising results, and we therefore encourage estimates on more sites to enable evaluation of the ~~b~~Budget ~~method~~approach based on the Sr isotope ratio.

~~It has been applied on three sites in the research performed in the 1990s. The results were on the same level as those from PROFILE, and also from the depletion method and the total analysis regressions for the sites where those methods were used. It requires deposition and leaching measurements (as in all budget calculations), which may be the reason why it has not been used more. The few comparisons made in this study show promising results, and we therefore encourage estimates on more sites to enable evaluation of the budget method based on the Sr isotope ratio.~~

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In the MAGIC model, the release from the exchangeable pool is thoroughly modelled, but for other sources and sinks of base cations, the same problems apply as for other budget approaches~~the budget method~~, e.g.~~i.e.~~ uncertainties in base cation deposition. These uncertainties exacerbate the uncertainties in weathering rates that derive from the mass balances in MAGIC. Nevertheless, MAGIC theoretically provides a good basis for conducting independent weathering rate assessments. On sites with relatively small input data uncertainties, our recommendation is to carry out such comparisons.

~~Futter et al. (2012) recommended that at least three independent methods be used to quantify weathering rates on a site for sustainability assessments. They also emphasised the importance of similar assumptions in such comparisons, most importantly calculating weathering rates to the same soil depth. In the review of weathering studies in this paper, we found only five locations where at least three methods had been implemented and where the criterion of the same depth is fulfilled. In four of those sites, budget calculation was one of the methods, and on three of those sites, the budget calculations gave unreliably high weathering rates. The explanation is that weathering cannot be distinguished from base cation exchange and release from other sources. Consequently, the recommendation to always use three independent methods seems unrealistic in practice, and the reliability of the different methods needs to be considered in the comparison.~~

~~9~~ 4 Conclusions

Uncertainties in weathering rates have often been presented as an obstacle in the assessment of sustainable forestry. The comparison between approaches in this paper, on a regional level as well as on a site level, suggests that both weathering rate gradients and approximate weathering rate levels can be captured with available methods. Although the variation in weathering ~~rates~~ estimates ~~between methods~~ was large on single sites, most of the sites could be grouped into broader classes representing very low, low and intermediate weathering rates, which can be used for general, but not specific, weathering rate assessments at site level. The more and better input data that is available, and the more methods that are applied and compared for a single site, the more robust overall assessments can be done at site level, provided that~~ing~~ the conceptional differences, boundary conditions and assumptions between methods are kept in mind.

Based on the results from this study, we argue that modelled weathering rates can be used for sustainability assessments, as long as the uncertainties, i.e. the intervals on single sites presented in this paper, are recognised. The ability to draw conclusions about sustainable forestry at site level depends not only on uncertainties in weathering rates, but also on other site properties, related~~ing~~ to forest properties and other base cation flows, such as base cation deposition, and the associated uncertainties. Irrespective of the uncertainties related to the sustainability assessments, a robust conclusion was that weathering rates in spruce forests in southern and central Sweden generally were substantially lower than the harvest losses at whole-tree harvesting, indicating that whole-tree harvesting without nutrient compensation is not sustainable in these areas. ~~However, t~~There is less risk of negative effect for spruce forests in northern Sweden, as well as pine forests in central and northern Sweden.

The research performed in the five years of the QWARTS programme supports the continued use of the PROFILE/ForSAFE models. ForSAFE is the only method that gives time-resolved results, ~~so is~~i.e. the only method that can be used to study dynamic effects of changing climate and changing management methods. Although there is still scope for improving process understanding and incorporation of that understanding into PROFILE and ForSAFE, e.g. regarding ~~weathering brakes and~~ biological weathering and weathering brakes, the most important way to reduce uncertainties in modelled weathering rates is to reduce input data uncertainties, mainly regarding soil texture and associated hydrological parameters. However, it is also important to continue ~~evaluating and comparing~~to compare with ~~other approaches~~results from the Depletion method and the Budget approach.~~.~~

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Data availability

Weathering data presented in this synthesis paper is compiled from other studies, which are published in other papers and referred to in the paper.

Author contributions

C. Akselsson planned and led the work, performed most of the calculations and wrote most parts of the paper. K. Bishop and S. Belyazid were highly involved in the planning and writing of the paper from start. S. Belyazid particularly contributed to the parts about modelling, including the chapters about biological weathering and the implications of higher resolution chemical reactions. J. Stendahl mainly contributed to parts about the ~~depletion~~ Depletion method and the ~~total~~ Total analysis regression approach, including the recalculation of weathering rates on a national scale using those methods. R. Finlay was the main author of the chapters about biological weathering, which also H. Wallander and S. Belyazid substantially contributed to. B. Olsson contributed to the methods descriptions, results and discussions concerning the ~~budget~~ Budget approach~~method~~. J-P. Gustafsson wrote about the implications of higher resolution chemical reactions together with S.

Belyazid and contributed to other parts in the paper where the chemistry in the weathering models was discussed. M. Erlandssons main contributions concerned the modelling parts and the parts about weathering brakes.

Competing interests

The authors declare that they have no conflict of interest.

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References

~~Aagaard, P. and Helgesson, H. C. Thermodynamic and kinetic constraints on reaction rates among minerals and aqueous solutions. 1. Theoretical considerations, American Journal of Science, 282, 237-285, doi:10.2475/ajs.282.3.237, 1982.~~

Aber, J. D. and Federer, C. A.: A generalized, lumped-parameter model of photosynthesis, evapotranspiration and net primary production in temperate and boreal forest ecosystems, Oecologia, 92, 463-474, doi:10.1007/BF00317837, 1992.

~~Aherne, J., Posch, M., Forsius, M., Lehtonen, A., and Härkönen, K.: Impacts of forest biomass removal on soil nutrient status under climate change: a catchment-based modelling study for Finland, Biogeochemistry, 107, 471-88, doi:10.1007/s10533-010-9569-4, 2012.~~

Akselsson, C., 2005. Regional Nutrient Budgets in Forest Soils in a Policy Perspective, PhD thesis, Department of Chemical Engineering, Lund University.

Akselsson, C., Holmqvist, J., Alveteg, M., Kurz, D., and Sverdrup, H.: Scaling and mapping regional calculations of soil chemical weathering rates in Sweden, Water, Air, & Soil Pollution: Focus, 4, 671-681, doi:10.1007/978-94-007-0952-2_46, 2004.

Akselsson, C., Hultberg, H., Karlsson, P. E., Pihl Karlsson, G., and Hellsten, S.: Acidification trends in south Swedish forest soils 1986-2008: slow recovery and high sensitivity to sea salt episodes, Science of the Total Environment, 444, 271–287, doi:10.1016/j.scitotenv.2012.11.106, 2013.

Akselsson, C., Olsson, J., Belyazid, S., and Capell R.: Can increased weathering rates due to future warming compensate for base cation losses following whole-tree harvesting in spruce forests?, Biogeochemistry, 128, 89-105, doi:10.1007/s10533-016-0196-6, 2016.

~~Akselsson, C., Westling, O., Alveteg, M., Thelin, G., Fransson, A-M., and Hellsten, S.: The influence of N load and harvest intensity on the risk of P limitation in Swedish forest soils, Science of the Total Environment 404, 284-289, doi:10.1016/j.scitotenv.2007.11.017, 2008.~~

Akselsson, C., Westling, O., Sverdrup, H., Holmqvist, J., Thelin, G., Uggla, E., and Malm, G.: Impact of harvest intensity on long-term base cation budgets in Swedish forest soils, Water, Air, & Soil Pollution: Focus, 7, 201-210, doi:10.1007/978-1-4020-5885-1_22, 2007.

~~Almeida, J.P., Rosenstock, N.P., Forsmark, B., Bergh, J., and Wallander, H.: Ectomycorrhizal community composition and function in a spruce forest transitioning between nitrogen and phosphorus limitation, Fungal Ecology, In press.~~

Alveteg, M.: Projecting regional patterns of future soil chemistry status in Swedish forests using SAFE, Water, Air, and Soil Pollution: Focus, 4, 49-59, doi:10.1007/978-94-007-0952-2_4, 2004.

Alveteg, M., Sverdrup, H., and Warfvinge, P.: Regional assessment of the temporal trends in soil acidification in southern Sweden, using the SAFE model, Water, Air, and Soil Pollution, 85, 2509-2514, doi:10.1007/BF01186211, 1995.

Ameli, A. A., Beven, K., Erlandsson, M., Creed, I. F., McDonnell, J. J., and Bishop, K.: Primary weathering rates, water transit times, and concentration-discharge relations: A theoretical analysis for the critical zone, Water Resources Research 53(1), 942-960, doi:10.1002/2016WR019448, 2017.

~~Ameli, A. A., Gabrielli, C., Morgenstern, U., and McDonnell, J. J.: Groundwater subsidy from headwaters to their parent water watershed: A combined field-modeling approach, Water Resources Research, 54(7), 5110-5125, doi: 10.1029/2017WR022356, 2018.~~

~~Andersson, M., Carlsson, M., Ladenberger, A., Morris, G., Sadeghi, M., and Uhlbäck J.: Geochemical Atlas of Sweden, Geological Survey of Sweden, 2014.~~

~~Averill, C., Turner, B. L., and Finzi, A. C.: Mycorrhiza-mediated competition between plants and decomposers drives soil carbon storage, Nature, 505, 543-545, doi:10.1038/nature12901, 2014.~~

~~Bahr, A., Ellström, M., Bergh, J., and Wallander, H.: Nitrogen leaching and ectomycorrhizal nitrogen retention capacity in a Norway spruce forest fertilized with nitrogen and phosphorus, Plant and Soil, 390, 323-335, doi:10.1007/s11104-015-2408-6, 2015.~~

Banfield, J. F., Barker, W. W., Welch, S. A., Taunton, A.: Biological impact on mineral dissolution: application of the lichen model to understanding mineral weathering in the rhizosphere, Proceedings of the National Academy of Sciences of the USA, 96, 3404-3411, doi:10.1073/pnas.96.7.3404, 1999.

~~Beerling, D. J., Leake, J. R., Long, S. P., Scholes, J. D., Ton, J., Nelson, P. N., Bird, M., Kantzas, E., Taylor, L. L., Sarkar, B., Kelland, M., DeLucia, E., Kantola, I., Muller, C., Rau, G. H., and Hansen, J.: Farming with crops and rocks to address global climate, food and soil security, Nature Plants, 4, 138-147, doi: 10.1038/s41477-018-0108-y, 2018.~~

Belyazid, S. and Akselsson, C.: Weathering dynamics in a changing climate – Scaling up the weathering rates in Swedish forests with the dynamic ecosystem model ForSAFE, ~~To be submitted to Biogeosciences~~ In review (this issue).

Belyazid, S., Westling, O., and Sverdrup, H.: Modelling changes in forest soil chemistry at 16 Swedish coniferous forest sites following deposition reduction, Environmental Pollution, 144, 596–609, doi:10.1016/j.envpol.2006.01.018, 2006.

Bishop, K., Seibert, J., Köhler, S., and Laudon, H.: Resolving the Double Paradox of rapidly mobilized old water with highly variable responses in runoff chemistry, Hydrological Processes, 18, 185-189, 10.1002/hyp.5209, 2004.

Bonneville, S., Smits, M. M., Brown, A., Harrington, J., Leake, J. R., Brydson, R., Benning, L. G.: Plant-driven fungal weathering: Early stages of mineral alteration at the nanometer scale, Geology, 37, 615-618, doi:10.1130/G25699A.1, 2009.

Bonten, L.T.C., Groenenberg, J.E., Meesenburg, H., and de Vries, W.: Using advanced surface complexation models for modelling soil chemistry under forests: Solling forest, Germany. Environmental Pollution, 159, 2831-2839, doi:10.1016/j.envpol.2011.05.002, 2011.

Börjesson, P., Hansson, J., and Berndes, G.; Future demand for forest-based biomass for energy purposes in Sweden, Forest Ecology and Management, 383, 17-26, doi:10.1016/j.foreco.2016.09.018, 2017.

Brantley, S.L., Kubicki, J, and White, A.F. (eds.): Kinetics of Water-Rock Interaction, New York, Springer, 858 pages, doi: 10.1007/978-0-387-73563-4, 2008.

Brimhall, G. H., Dietrich, W. E.: Constitutive mass balance relations between chemical composition, volume, density, porosity, and strain in metasomatic hydrochemical systems: Results on weathering and pedogenesis, Geochimica et Cosmochimica Acta, 51(3), 567-587, doi:10.1016/0016-7037(87)90070-6, 1987.

Brimhall, G. H., Lewis, C. J., Ford, C., Bratt, J., Taylor, G., and Warin, O.: Quantitative geochemical approach to pedogenesis: importance of parent material reduction, volumetric expansion, and eolian influx in lateritization, Geoderma, 51(1-4), 51-91, doi:10.1016/0016-7061(91)90066-3, 1991.

~~Calvaruso, C., Turpault, M. P., Frey-Klett, P., Uroz, S., Pierret, M. C., Tosheva, Z., and Kies, A.: Increase of apatite dissolution rate by Scots pine roots associated or not with Burkholderia glathei PML1(12)Rp in open-system flow microcosms, Geochimica et Cosmochimica Acta, 106, 287-306, doi:10.1016/j.gca.2012.12.014, 2013.~~

Casetou-Gustafson, S., Akselsson, C., Grip, H., Hillier, S., Linder, S., Olsson, B.A., Simonsson, M., and Stendahl, J.: Current and historical weathering rates of base cations at two forest sites in N and S Sweden: a comparison of three methods, ~~To be submitted to Biogeosciences~~in review (this issue).

Casetou-Gustafson, S., Akselsson, C., Hillier, S., and Olsson, B.: The importance of mineral determinations to PROFILE base cation weathering rates: A case study, ~~In review in~~ Biogeosciences, 16, 1903-1920, doi.org/10.5194/bg-16-1903-2019, 2019. ~~(this issue).~~

Casetou-Gustafson, S., Hillier, S., Akselsson, C., Simonsson, M., Stendahl, J., and Olsson, B.: Comparison of measured (XRPD) and modeled (A2M) soil mineralogies: A study of some Swedish forest soils in the context of weathering rate predictions, Geoderma, 310, 77-88, doi:10.1016/j.geoderma.2017.09.004, 2018.

Cosby, B. J., Ferrier, R. C., Jenkins, A., Wright, R. F.: Modelling the effects of acid deposition: refinements, adjustments and inclusion of nitrogen dynamics in the MAGIC model, Hydrology and Earth System Sciences, 5, 499-517, doi:10.5194/hess-5-499-2001, 2001.

de Vries, W.: Soil response to acid deposition at different regional scales. Field and laboratory data, critical loads and model predictions, Cip-Gegevens Koninklijke Bibliotheek, Den Haag, 1994.

Egnell, G.: Effects of slash and stump harvesting after final felling on stand and site productivity in Scots pine and Norway spruce, Forest Ecology and Management, 371, 42-49, doi:10.1016/j.foreco.2016.03.006, 2016.

~~Ericson, T.: Growth and shoot:root ratio of seedlings in relation to nutrient availability, Plant and Soil, 168-169, 205-214, doi:10.1007/BF00029330, 1995.~~

Erlandsson, M., Oelkers, E. H., Bishop, K., Sverdrup, H., Belyazid, S., Ledesma, J. L. J., and Köhler, S.J.: Spatial and temporal variations of base cation release from chemical weathering on a hillslope scale, Chemical Geology, 441, 1-13, doi:10.1016/j.chemgeo.2016.08.008, 2016.

Erlandsson, M., Sverdrup, H. U., Bishop, K. H., Belyazid, S., Ameli, A. A., Köhler, S. J.: Catchment export of base cations: Improved mineral dissolution kinetics influence the role of water transit time. ~~To be submitted to SOIL~~In review (this issue).

~~Fahad, Z. A., Bolou-Bi, E. B., Köhler, S. J., Finlay, R. D., and Mahmood, S.: Fractionation and assimilation of Mg isotopes by fungi is species dependent, Environmental Microbiology Reports, 8, 956-965, doi:10.1111/1758-2229.12459, 2016.~~

Finlay, R. D., and Clemmensen, K. E. Immobilization of carbon in mycorrhizal mycelial biomass and secretions, in: Mycorrhizal Mediation of Soil Fertility, Structure, and Carbon Storage, edited by Johnson, N. C., Gehring, C., and Jansa, J.: Elsevier, Amsterdam, 413-440, 2017.

Finlay, R. D., Mahmood, S., Rosenstock, N., Bolou-Bi, E. B., Köhler, S. J., Fahad, Z., Rosling, A., Wallander, H., Belyazid, S., Bishop, K. and Lian, B.: Biological weathering and its consequences at different spatial levels – from nanoscale to global scale, Biogeosciences (this issue), 2019.

~~Finlay, R. D., Mahmood, S., Rosenstock, N., Fahad, Z., Wallander, H., Belyazid, S., and Lian, B.: Biological weathering and its consequences at different spatial levels – from nanoscale to global scale, To be submitted to Biogeosciences (this issue).~~

Finlay, R. D., Wallander, H., Smits, M., Holmstrom, S., Hees, P. v., Lian, B., Rosling, A., and Finlay, R. D.: The role of fungi in biogenic weathering in boreal forest soils, Fungal Biology Reviews, 4, 101-106, doi:10.1016/j.fbr.2010.03.002, 2009.

Flemming, H. C., Wingender, J., Szewzyk, U., Steinberg, P., Rice, S. A., and Kjelleberg, S.: Biofilms: an emergent form of bacterial life, Nature Reviews Microbiology, 14, 563-575, doi:10.1038/nrmicro.2016.94, 2016.

Fridman, J., Holm, S., Nilsson, M., Nilsson, P., Hedström Ringvall, A., Ståhl, G.: Adapting National Forest Inventories to changing requirements – the case of the Swedish National Forest Inventory at the turn of the 20th century, Silva Fennica, 48 (3), article id 1095, 1-29, doi: 10.14214/sf.1095, 2014.

Fumoto, T., Shindo, J., Oura, N., and Sverdrup, H.: Adapting the Profile Model to Calculate the Critical Loads for East Asian Soils by Including Volcanic Glass Weathering and Alternative Aluminum Solubility System, Water, Air & Soil Pollution, 130 (1-4), 1247-1252, doi:10.1023/A:101398121, 2001.

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Futter, M., Klaminder, J., Lucas, R., Laudon, H., and Köhler, S.: Uncertainty in silicate mineral weathering rate estimates: source partitioning and policy implications, Environmental Research Letters, 7, 1-8, doi:10.1088/1748-9326/7/2/024025, 2012.

~~Gadd, G. M.: Microbial roles in mineral transformations and metal cycling in the Earth's critical zone, Molecular Environmental Soil Science, 115-165, doi:10.1007/978-94-007-4177-5_6, 2013a.~~

~~Gadd, G. M.: Geomycology: Fungi as Agents of Biogeochemical Change, Biology and Environment Proceedings of the Royal Irish Academy, 113b(2), 139-153, doi:10.3318/BIOE.2013.16, 2013b.~~

~~Gadd, G. M.: Fungi, rocks, and minerals, Elements, 13, 171-176, doi:10.2113/gselements.13.3.171, 2017.~~

Gardelin, M. and Warfvinge, P.: Databas, in: Modelling groundwater response to acidification, edited by: Sandén, P. and Warfvinge, P., Report from the Swedish integrated groundwater acidification project, SMHI, Reports Hydrology, 13-29, 1992.

Gazze, S. A., Saccone, L., Smits, M. M., Duran, A. L., Leake, J. R., Banwart, S. A., Ragnarsdottir, K. V., and McMaster, T. J.: Nanoscale observations of extracellular polymeric substances deposition on phyllosilicates by an ectomycorrhizal fungus, Geomicrobiology Journal, 30, 721-730, doi:10.1080/01490451.2013.766285, 2013.

~~Godderis, Y., Francois, L. M., Probst, A., Schott, J., Moncoulon, D., Labat, D., Viville, D.: Modelling weathering processes at the catchment scale: The WITCH numerical model, Geochimica et Cosmochimica Acta 70(5), 1128-1147, doi:10.1016/j.gca.2005.11.018, 2006.~~

Godderis, Y., Francois, L. M., Probst, A., Schott, J., Moncoulon, D., Labat, D., Viville, D.: Modelling weathering processes at the catchment scale: The WITCH numerical model, Geochimica et Cosmochimica Acta 70(5), 1128-1147, doi:10.1016/j.gca.2005.11.018, 2006.

Graf Pannatier, E., ~~Graf Pannatier, E.,~~ Thimonier, A., Schmitt, M., Walthert, L., and Waldner, P.: A decade of monitoring at Swiss long-term forest ecosystem research (LWF) sites: can we observe trends in atmospheric acid deposition and in soil solution acidity?, Environmental Monitoring and Assessment, 174. 3-30, doi:10.1007/s10661-010-1754-3, 2011.

Gustafsson, J. P.: Modeling the acid-base properties and metal complexation of humic substances with the Stockholm Humic Model, Journal of Colloid and Interface Science, 244, 102-112, doi:10.1006/jcis.2001.7871, 2001.

Gustafsson, J.P., Belyazid, S., McGivney, E., and Löfgren, S.: Aluminium and base cation chemistry in dynamic acidification models – need for a reappraisal?, ~~Soil~~SOIL, 4, 237-250, doi:10.5194/soil-4-237-2018, 2018.

~~Hägglund, B.: A new Swedish National Forest Survey, Swedish University of Agricultural Sciences, Report 37, Uppsala, 1985.~~Hedin, L, Granat, L., Likens, G., Buishand, A., Galloway, J., Butler, T., and Rodhe, H.: Steep declines in atmospheric base cations in regions of Europe and North America. Nature, 367, 351–354, doi:10.1038/367351a0, 1994.

Hodge, A.: Plant Uptake, in: Reference Module in Earth Systems and Environmental Sciences, edited by: Elias, S.A., doi: 10.1016/B978-0-12-409548-9.05232-5, 2013.

Hodson, M.: Experimental evidence for mobility of Zr and other trace elements in soils, Geochimica et Cosmochimica Acta, 66(5), 819-828, doi:10.1016/S0016-7037(01)00803-1, 2002.

Holmqvist, J., Ögaard, A. F., Öborn, I., Edwards, A. C., Mattsson, L., and Sverdrup, H.: Application of the PROFILE model to estimate potassium release from mineral weathering in Northern European agricultural soils, European Journal of Agronomy, 20(1-2), 149-163, doi:10.1016/S1161-0301(03)00064-9, 2003.

~~Hultberg, H. and Ferm, M.: Temporal changes and fluxes of sulphur and calcium in wet and dry deposition, internal circulation as well as in run-off and soil in a forest at Gårdsjön, Sweden, Biogeochemistry, 68, 355-63, doi:10.1023/B:BIOG.0000031037.40067.55, 2004.~~

Irving, P. M.: Acidic deposition: state of science and technology: summary report of the US National Acid Precipitation Assessment Program, National Acid Precipitation Assessment Program, Office of the Director, 1991.

~~Iwald, J., Löfgren, S., Stendahl, J., and Karltun, E.: Acidifying effect of removal of tree stumps and logging residues as compared to atmospheric deposition. Forest Ecology and Management, 290, 49-58, doi:10.1016/j.foreco.2012.06.022, 2013.~~

Jacks, G. and Åberg, G. Calcium budgets for catchments as interpreted by strontium isotopes, in: Surface Water Acidification Programme, edited by: Mason, B. J., The Royal Society, The Royal Swedish Academy of Sciences, The Norwegian Academy of Science and Letters, London, Stockholm, Oslo, 120-131, doi:10.2166/nh.1989.0007, 1987.

~~Jongmans, A. G., van Breemen, N., Lundstrom, U., van Hees, P. A. W., Finlay, R. D., Srinivasan, M., Unestam, T., Giesler, R., Melkerud, P. A., and Olsson, M.: Rock-eating fungi, Nature, 389, 682-683, doi:10.1038/39493, 1997.~~

Jönsson, C., Warfvinge, P., and Sverdrup, H.: Uncertainty in predicting weathering rate and environmental stress factors with the PROFILE model, Water, Air and Soil Pollution, 81, 1-23, doi:10.1007/BF00477253, 1995.

Kinniburgh, D. G., van Riemsdijk, W. H., Koopal, L. K., Borkovec, M., Benedetti, M. F., and Avena, M. J.: Ion binding to natural organic matter: stoichiometry and thermodynamic consistency, Colloid Surface A, 151, 147-166, doi:10.1016/S0927-7757(98)00637-2, 1999.

Klaminder, J., Lucas, R. W., Futter. M. N., Bishop, K. H., Köhler, S. J., Egnell, G., and Laudon, H.: Silicate mineral weathering rate estimates: are they precise enough to be useful when predicting the recovery of nutrient pools after harvesting?, Forest Ecology and Management, 261, 1-9, doi:10.1016/j.foreco.2010.09.040, 2011.

Koptsik, G., Teveldal, S., Aamlid, D., and Venn, K.: Calculations of weathering rate and soil solution chemistry for forest soils in the Norwegian-Russian border area with the PROFILE model, Applied Geochemistry, 14(2), 173-185, doi:10.1016/S0883-2927(98)00048-1, 1999.

Köhler, S., Zetterberg, T., Futter, M., Fölster, J., and Löfgren, S.: Assessment of Uncertainty in Long-Term Mass Balances for Acidification Assessments: A MAGIC Model Exercise, AMBIO, 40, 891-905, doi:10.1007/s13280-011-0208-7, 2011.

Kronnäs, V., Akselsson, C., and Belyazid, S.: Dynamic modelling of weathering rates – Is there any benefit over steady-state modelling?, ~~In press,~~ SOIL, 5, 33-47, doi.org/10.5194/soil-5-33-2019, 2019. ~~(this issue).~~

~~Landeweert, R., Hofflund, E., Finlay, R. D., and van Breemen, N.: Linking plants to rocks: Ectomycorrhizal fungi mobilize nutrients from minerals, Trends in Ecology and Evolution, 16, 248-254, doi:10.1016/S0169-5347(01)02122-X, 2001.~~

Langan, S., Sverdrup, H., Coull, M.: The calculation of base cation release from the chemical weathering of Scottish soils using the PROFILE model, Water, Air and Soil Pollution, 85(4), 2497-2502, doi:10.1007/BF01186209, 1995.

~~Langner, J., Persson, C., Robertson, L., and Ullerstig, A.: Air pollution assessment study using the MATCH Modelling System. Application to sulphur and nitrogen compounds over Sweden 1994, Swedish Meteorological and Hydrological Institute, Report no. 69, Norrköping, Sweden, 1996.~~

~~Leake, J. R., Duran, A. L., Hardy, K. E., Johnson, I., Beerling, D. J., Banwart, S. A., and Smits, M. M.: Biological weathering in soil: the role of symbiotic root-associated fungi biosensing minerals and directing photosynthate-energy into grain-scale mineral weathering, Mineralogical Magazine, 72, 85-89, doi:10.1180/minmag.2008.072.1.85, 2008.~~

Leake, J. R. and Read, D. J.: Mycorrhizal symbioses and pedogenesis throughout Earth's history, in: Mycorrhizal Mediation of Soil Fertility, Structure, and Carbon Storage, edited by: Johnson, N. C., Gehring, C., and Jansa, J., Elsevier, Amsterdam, 9-33, 2017.

Lidskog, R. and Sundqvist, G.: The Role of Science in Environmental Regimes: The Case of LRTAP, European Journal of International Relations, 8(1), 77-101, doi:10.1177/1354066102008001003, 2002.

~~Lindahl, B. D. and Tunlid, A.: Ectomycorrhizal fungi - potential organic matter decomposers, yet not saprotrophs, New Phytologist, 205, 1443-1447, doi:10.1111/nph.13201, 2015.~~

Lindström, G. and Gardelin, M.: Modelling groundwater response to acidification, in: Modelling groundwater response to acidification, edited by: Sandén, P. and Warfvinge, P., Report from the Swedish integrated groundwater acidification project, SMHI, Reports Hydrology, 33-36, 1992.

Lundström, U.: Laboratory and lysimeter studies of chemical weathering, in: The surface water acidification programme, edited by Mason, B. J., Cambridge University Press, Cambridge, UK, 267-274, 1990.

Löfgren, S., Ågren, A., Gustafsson, J. P., Olsson, B. A., and Zetterberg, T.: Impact of whole-tree harvest on soil and stream water acidity in southern Sweden based on HD-MINTEQ simulations and pH-sensitivity, Forest Ecology and Management, 383, 49-60, doi:10.1016/j.foreco.2016.07.018, 2017.

Maher, K., Steefel, C. I., White, A. F., and Stonestrom, D. A.: The role of reaction affinity and secondary minerals in regulating chemical weathering rates at the Santa Cruz Soil Chronosequence, California, Geochimica et Cosmochimica Acta, 73(10), 2804-2831, doi:10.1016/j.gca.2009.01.030, 2009.

Marshall, C. E. and Haseman, J. F.: The quantitative evaluation of soil formation and development by heavy mineral studies: a Grundy silt loam profile, Soil Science Society of America Proceedings, 7, 448-453, 1942.

Marupakula, S., Mahmood, S., and Finlay, R. D.: Analysis of single root tip microbiomes suggests that distinctive bacterial communities are selected by Pinus sylvestris roots colonized by different ectomycorrhizal fungi, Environmental Microbiology, 18, 1470-1483, doi:10.1111/1462-2920.13102, 2016.

Mason B.J. (Ed.): The Surface Waters Acidification Programme. Cambridge University Press, Cambridge, UK, 1990.

Maxe, L. (Ed.): Effects of acidification on groundwater in Sweden. Hydrological and hydrochemical processes, Report from the Swedish integrated groundwater acidification project, Swedish Environmental Protection Agency, Report 4388, 1995.

McGivney, E., Gustafsson, J.P., Belyazid, S., Zetterberg, T., Löfgren, S.: Assessing the impact of acid rain and forest harvest intensity with the HD-MINTEQ model – Soil chemistry of three Swedish conifer sites from 1880 to 2080, In review in SOIL (this issue).

~~Mcloughlin, N., Staudigel, H., Furnes, H., Eickmann, B., and Ivarsson, M.: Mechanisms of microtunneling in rock substrates: distinguishing endolithic biosignatures from abiotic microtunnels, Geobiology, 8, 245-255, doi:10.1111/j.1472-4669.2010.00243.x, 2010.~~

~~McMaster, T. J.: Atomic Force Microscopy of the fungi-mineral interface: applications in mineral dissolution, weathering and biogeochemistry, Current Opinion in Biotechnology, 23, 562-569, doi:10.1016/j.copbio.2012.05.006, 2012.~~

Morén, A-S. and Perttu, K.: Regional temperature and radiation indices and their adjustment to horizontal and inclined forest land, Studia Forestalia Suecica, No. 194, Swedish University of Agricultural Sciences, 1994.

Nilsson, J. and Grennfelt, P.: Critical Loads for Sulphur and Nitrogen. Report from a workshop held at Skokloster, Sweden, 19-24 March, 1988. Miljörapport 1988:15, 1988.

Odén, S.: Nederbördens och luftens försurning – dess orsak, förlopp och verkan i olika miljöer. Bulletin nr. 1, Statens Naturvetenskapliga Forskningsråd, Stockholm, Sweden, 1968.

Olsson, M. and Melkerud, P-A.: Determination of weathering rates based on geochemical properties of the soil, in: Proceedings of the Conference on Environmental Geochemistry in Northern Europe, edited by: Pulkinene, E., Symposius series no. 34, Geological Survey of Finland, 45-61, 1990.

Olsson, M., Rosén, K., and Melkerud, P.-A.: Regional modelling of base cation losses from Swedish forest soils due to whole-tree harvesting, Applied Geochemistry, 2, 189-194, doi:10.1016/S0883-2927(09)80035-8, 1993.

Paces, T.: Weathering rates of gneiss and depletion of exchangeable cations in soils under environmental acidification, Journal of the Geological Society, 143, 673-677, doi:10.1144/gsjgs.143.4.0673, 1986.

~~Persson, C., Langner, J., and Robertson, L.: Air Pollution Assessment Studies for Sweden Based on the Match Model and Air Pollution Measurements, in: Air Pollution Modeling and Its Application XI, edited by: Gryning, S-E. and Schiermeier, F.A., NATO · Challenges of Modern Society, vol 21. Springer, Boston, MA, 127-134, doi:10.1007/978-1-4615-5841-5_15, 1996.~~

Phelan, J., Belyazid, S., Kurz, D., Guthrie, S., Cajka, J., Sverdrup, H., and Waite, R.: Estimation of Soil Base Cation Weathering Rates with the PROFILE Model to Determine Critical Loads of Acidity for Forested Ecosystems in Pennsylvania, USA: Pilot Application of a Potential National Methodology, Water Air and Soil Pollution, 225(9), 2109-2128, doi:10.1007/s11270-014-2109-4, 2014.

Pihl Karlsson, G., Akselsson, C., Hellsten, S., Karlsson, P. E.: Reduced European emissions of S and N - effects on air concentrations, deposition and soil water chemistry in Swedish forests, Environmental Pollution, 159, 3571-3582, doi:10.1016/j.envpol.2011.08.007, 2011.

Posch, M. and Kurz, D.: A2M – A program to compute all possible mineral modes from geochemical analyses, Computers & Geosciences, 33, 563–572, 2007.

~~Quirk, J., Beerling, D. J., Banwart, S. A., and Leake, J. R.: Evolution of trees and mycorrhizal fungi intensifies silicate mineral weathering, Biology Letters, 8, 1006-1011, doi:10.1098/rsbl.2012.0503, 2012.~~

~~Quirk, J., Leake, J. R., Banwart, S. A., Taylor, L. L., and Beerling, D. J.: Weathering by tree-root-associating fungi diminishes under simulated Cenozoic atmospheric CO$_2$ decline, Biogeosciences, 11, 321-331, doi:10.5194/bg-11-321-2014, 2014.~~

Rapp, L. and Bishop, K.: Modeling Surface Water Critical Loads with PROFILE: Possibilities and Challenges, Journal of Environmental Quality, 32(6), 2290-2300, doi:10.2134/jeq2003.2290, 2003.

Reuss, J. O. and Johnson, D. W.: Acid deposition and the acidification of soils and waters, Ecological Studies 59, Springer Verlag, New York, US, doi:10.1007/978-1-4419-8536-1, 1986.

Rosengren, U. and Stjernquist, I.: Gå på djupet! Om rotdjup och rotproduktion i olika skogstyper, SUFOR – Sustainable forestry in Southern Sweden, Alnarp, ISBN: 9157666172 9789157666178, 2004.

~~Rosenstock, N., Berner, C., Kram, P., Smits, M., and Wallander, H.: The role of phosphorus, magnesium, and potassium availability in soil fungal exploration of mineral nutrient sources in Norway spruce forests, New Phytologist, 211, 542-553, doi:10.1111/nph.13928, 2016.~~

Rosenstock, N., Löfgren, S., and van der Heijden, G.: Over what time scales do mineral weathering rates become important for ecosystem function?, ~~To be submitted to Biogeosciences~~In review (this issue).

~~Rosling, A., Lindahl, B. D., and Finlay, R. D.: Carbon allocation to ectomycorrhizal roots and mycelium colonising different mineral substrates, New Phytologist,, 162, 795-802, doi:10.1111/j.1469-8137.2004.01080.x, 2004.~~

~~Saccone, L., Gazze, S. A., Duran, A. L., Leake, J. R., Banwart, S. A., Ragnarsdottir, K. V., Smits, M. M., and McMaster, T. J.: High resolution characterization of ectomycorrhizal fungal-mineral interactions in axenic microcosm experiments, Biogeochemistry, 111, 411-425, doi:10.1007/s10533-011-9667-y, 2012.~~

~~Schmalenberger, A., Duran, A. L., Bray, A. W., Bridge, J., Bonneville, S., Benning, L. G., Romero-Gonzalez, M. E., Leake, J. R., and Banwart, S. A.: Oxalate secretion by ectomycorrhizal Paxillus involutus is mineral-specific and controls calcium weathering from minerals, Sci. Rep, 5, ARTN 12187, 2015.~~

~~Schöpp, W., Posch, M., Mylona, S., Johansson, M.: Long-term development of acid deposition (1880-2030) in sensitive freshwater areas in Europe, Hydrology and Earth System Sciences, 7(4), 436-446, doi:10.5194/hess-7-436-2003, 2003.~~

~~Simonsson, M., Bergholm, J., Olsson, B., von Brömssen, C., Öborn, I.: Estimating weathering rates using base cation budgets in a Norway spruce stand on podzolised soil: Analysis of fluxes and uncertainties, Forest Ecology and Management 340, 135-152, doi:10.1016/j.foreco.2014.12.024, 2015.~~

~~Simpson, D., Benedictow, A., Berge, H., Bergström, R., Emberson, L. D., Fagerli, H., Flechard, C. R., Hayman, G. D., Gauss, M., Jonson, J. E., Jenkin, M. E., Nyiri, A., Richter, C., Semeena, V. S., Tsyro, S, Tuovinen, J-P., Valdebenito, A., Wind, P.: The EMEP MSC-W chemical transport model – technical description, Atmospheric Chemistry and Physics, 12, 7825-7865, doi:10.5194/acp-12-7825-2012, 2012.~~

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Smits, M. M., Bonneville, S., Benning, L. G., Banwart, S. A., and Leake, J. R.: Plant-driven weathering of apatite - the role of an ectomycorrhizal fungus, Geobiology, 10, 445-456, doi:10.1111/j.1472-4669.2012.00331.x, 2012.

~~Smits, M. M., Hoffland, E., Jongmans, A. G., and van Breemen, N.: Contribution of mineral tunneling to total feldspar weathering, Geoderma, 125, 59-69, doi:10.1016/j.geoderma.2004.06.005, 2005.~~

Smits, M. M. and Wallander, H.: Role of mycorrhizal symbiosis in mineral weathering and nutrient mining from soil parent material, in: Mycorrhizal Mediation of Soil Fertility, Structure, and Carbon Storage, edited by: Johnson, N. C., Gehring, C., and Jansa, J, Elsevier, Amsterdam, 35-46, 2017.

Starr, M., Lindroos, A.-J., Tarvainen, T. and Tanskanen, H.: Weathering rates in the Hietajärvi Integrated Monitoring catchment, Boreal Environmental Research, 3, 275–285, ISSN 1239-6095, 1998.

Stendahl, J., Akselsson, C., Melkerud, P-A., and Belyazid, S.: Pedon-scale silicate weathering: comparison of the PROFILE model and the depletion method at 16 forest sites in Sweden, Geoderma, 211-212, 65-74, doi:10.1016/j.geoderma.2013.07.005, 2013.

Stendahl, J., Lundin, L., and Nilsson, T.: The stone and boulder content of Swedish forest soils. Catena, 77 (3), 285-291, doi.org/10.1016/j.catena.2009.02.011, 2009.

~~Sverdrup, H.: Chemical weathering of soil minerals and the role of biological processes, Fungal Biology Reviews, 4, 94-100, doi:10.1016/j.fbr.2009.12.001, 2009.~~

Sverdrup, H. and de Vries, W.: Calculating critical loads for acidity with the simple mass balance method, Water, Air, and Soil Pollution, 72, 143-162, doi:10.1007/BF01257121, 1994.

~~Sverdrup, H., Hagen-Thorn, A., Holmqvist, J., Wallman, P., Warfvinge, P., Walse, C., and Alveteg, M.: Biogeochemical processes and mechanisms, in: Developing principles for sustainable forestry. Results from a research program in southern Sweden managing forest ecosystems, edited by: Sverdrup, H. and Stjernquist, I., Kluwer Academic Publishers, Amsterdam, 91-196, 2002.~~

Sverdrup, H. and Warfvinge, P.: On the geochemistry of chemical weathering, in: Chemical weathering under field conditions, edited by: Rosén, K., Reports from a Nordic seminar, 27-28 September 1990, Wik, Uppsala, Reports in Forest Ecology and Forest Soils, Report 63, Swedish University of Agricultural Sciences, 1991.

Sverdrup, H. and Warfvinge, P.: Calculating field weathering rates using a mechanistic geochemical model (PROFILE), Applied Geochemistry, 8, 273-283, doi:10.1016/0883-2927(93)90042-F, 1993.

Sverdrup, H., Warfvinge, P., and Wickman, T.: Estimating the weathering rate at Gårdsjön using different methods, in: Experimental Reversal of Acid Rain Effects, edited by: Hultberg, H. and Skeffington, R., John Wiley Sons Ltd, 1998.

Sverdrup, H., Oelkers, E. H., Belyazid, S., and Erlandsson, M.: Modelling weathering with the PROFILE and ForSAFE models using updated advanced weathering kinetics, ~~To be submitted to Biogeosciences (this issue).~~In review (this issue).

~~Taylor, L. L., Banwart, S. A., Leake, J. R., and Beerling, D. J.: Modelling the evolutionary rise of ectomycorrhiza on sub-surface weathering environments and the geochemical carbon cycle, Amer. J. Sci., 311, 369-403, 2011.~~

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~~Taylor, L.L., Banwart, S.A., Leake, J.R., Beerling, D.J. Modelling the evolutionary rise of ectomycorrhiza on sub-surface weathering environments and the geochemical carbon cycle. American Journal of Science, 311, 369-403, doi:10.2475/05.2011.01, 2011.~~

Taylor, A. and Blum, J. D.: Relation between soil age and silicate weathering rates determined from the chemical evolution of a glacial chronosequence, Geology, 23, 979-982, doi:10.1130/0091-7613(1995)023<0979:RBSAAS>2.3.CO;2, 1995.

~~Taylor, L. L., Leake, J. R., Quirk, J., Hardy, K., Banwart, S. A., and Beerling, D. J.: Biological weathering and the long-term carbon cycle: integrating mycorrhizal evolution and function into the current paradigm, Geobiology, 7, 171-191, doi:10.1111/j.1472-4669.2009.00194.x, 2009.~~

~~Taylor, L. L., Quirk, J., Thorley, R. M. S., Kharecha, P. A., Hansen, J., Ridgwell, A., Lomas, M. R., Banwart, S. A., and Beerling, D. J.: Enhanced weathering strategies for stabilizing climate and averting ocean acidification, Nature Climate Change, 6, 402-406, doi:10.1038/nclimate2882, 2016.~~

Tipping, E.: CHUM – a hydrochemical model for upland catchments, Journal of Hydrology, 174, 305-330, doi:10.1016/0022-1694(95)02760-2, 1996.

Tipping, E.: Cation Binding by Humic Substances, Cambridge University Press, Cambridge, UK, 2002.

Umweltsbundesamt: Manual on methodologies and criteria for mapping critical levels/loads and geographical areas where they are exceeded, UNECE Convention On Long-range Transboundary Air Pollution, Texte 71/96, Berlin, 1996.

UNECE: Protocol to the 1979 Convention on Long-Range Transboundary Air Pollution on Further Reductions of Sulphur Emissions, Article 1:8, 1994.

~~van Breemen, N., Finlay, R., Lundstrom, U., Jongmans, A. G., Giesler, R., and Olsson, M.: Mycorrhizal weathering: A true case of mineral plant nutrition?, Biogeochemistry, 49, 53-67, doi:10.1023/A:100625623, 2000.~~

van der Heijden, G., Bel, J., Craig, C. A., Midwood, A. J., Mareschal, L., Ranger, J., Dambrine, E., and Legout, A.: Measuring Plant-Available Mg, Ca, and K Pools in the Soil - An Isotopic Dilution Assay, ACS Earth and Space Chemistry, 2(4), 292-313, doi:10.1021/acsearthspacechem.7b00150, 2018.

Wallman, P., Belyazid, S., Svensson, M., Sverdrup, H.: DECOMP - a semi-mechanistic model of litter decomposition, Environmental Modelling and Software, 21(1), 33-44, doi:10.1016/j.envsoft.2004.09.026, 2004.

Wallman, P., Svenssson, M., Sverdrup, H., and Belyazid, S.: ForSAFE - an integrated process-oriented forest model for long-term sustainability assessments, Forest Ecology and Management, 207, 19-36, doi:10.1016/j.foreco.2004.10.016, 2005.

Walse, C., Berg, B., and Sverdrup, H.: Review and synthesis of experimental data on organic matter decomposition with respect to the effects of temperature, moisture, and acidity, Environmental Review, 6, 25-40, doi:10.1139/a98-001, 1998.

Warfvinge, P. and Sverdrup, H.: Calculating critical loads of acid deposition with PROFILE - A steady-state soil chemistry model, Water, Air, Soil Pollution, 63, 119-143, doi:10.1007/BF00475626, 1992.

Warfvinge, P., Sverdrup, H., Alveteg, M., and Rietz, F.: Modelling geochemistry and lake pH since glaciation at lake Gårdsjön, Water, Air and Soil Pollution, 85, 713-718, doi:10.1007/BF00476913, 1995.

Warfvinge, P. and Sverdrup, S.: Critical load of acidity to Swedish forest soils. Methods, data and results, Lund University, Department of Chemical Engineering II, Lund, Sweden, 1995.

Wickman, T. and Jacks, G.: Strontium isotopes as tools in weathering research, in: Chemical weathering under field conditions, edited by: Rosén, K., Reports from a Nordic seminar, 27-28 September 1990, Wik, Uppsala, Reports in Forest Ecology and Forest Soils, Report 63, Swedish University of Agricultural Sciences, 1991.

Zanchi, G., Belyazid, S., Akselsson, C., Yu, L., Bishop, K., Köhler, S., and Grip, H.: A Hydrological Concept including Lateral Water Flow Compatible with the Biogeochemical Model ForSAFE, Hydrology, 3(11), 1-19, doi:10.3390/hydrology3010011, 2016.

Zetterberg, T., Köhler, S. and Löfgren, S., T.: Sensitivity analyses of MAGIC modelled predictions of future impacts of whole-tree harvest on soil calcium supply and stream acid neutralizing capacity, Science of the total Environment, 494-495, 187-201, doi.org/10.1016/j.scitotenv.2014.06.114, 2014.

Zetterberg, T., Olsson, B., Löfgren, S., von Brömssen, C., and Brandtberg P-O.: The effect of harvest intensity on long-term calcium dynamics in soil and soil solution at three coniferous sites in Sweden, Forest Ecology and Management, 302, 280-294, doi:10.1016/j.foreco.2013.03.030, 2013.~~.~~

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Table 1: Sites included in the study, where at least two well-documented approaches for estimating wethering rates have been applied to the same depth.

Site	Depth	References
Gårdsjön A	0.5 m_b	Stendahl et al., 2013
Gårdsjön B (F1)	0.67 $\mathrm{m}_{a,c}$	Sverdrup et al., 1998; Köhler et al., 2011
Gårdsjön C	0.47 m_a	Sverdrup et al., 1998
Svartberget A	0.5 m_b	Stendahl et al., 2013
Svartberget B	0.8 m_a	Sverdrup et al., 1991; Sverdrup and Warfvinge,1993; Lundström, 1990
Vindeln	0.5 m_b	Stendahl et al., 2013
Risfallet A	0.5 m_b	Stendahl et al., 2013
Risfallet B	1.0 m_b	Sverdrup et al., 1991; Sverdrup and Warfvinge, 1993; Jönsson et al., 1995; Maxe, 1995
Fårahall	1.0 m_b	Sverdrup et al., 1991; Sverdrup and Warfvinge, 1993; Jönsson et al., 1995; Maxe, 1995
Stubbetorp	1.0 m_a	Maxe, 1995; Gardelin and Warfvinge, 1992
Flakaliden	0.5 m_b	Casetou-Gustafson et al., 2019
Asa	0.5 m_b	Casetou-Gustafson et al., 2019
Bodafors	0.5 m_b	Stendahl et al., 2013
Hjärtasjö	0.5 m_b	Stendahl et al., 2013
Hässlen	0.5 m_b	Stendahl et al., 2013
Kloten	0.5 m_b	Stendahl et al., 2013
Kullarna	0.5 m_b	Stendahl et al., 2013
Lammhult	0.5 m_b	Stendahl et al., 2013
Skånes Värsjö	0.5 m_b	Stendahl et al., 2013
Stöde	0.5 m_b	Stendahl et al., 2013
Söderåsen	0.5 m_b	Stendahl et al., 2013
Västra Torup	0.5 m_b	Kronnäs et al., 2019
Hissmossa	0.5 m_b	Kronnäs et al., 2019

$_a$ Including O-layer

$_b$ Not including O-layer

$_c$ For MAGIC the weathering rate was calculated to 0.6 m, including O layer

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Table 2: Mineral content in soil (50 cm depth) used as input for modelling weathering rates for the different sites: Qz (qwartz), Or (orthoclase), Pl (plagioclase), Am (amphibole), Ep (epidote). Bi (biotite), Ap (Apatite), Mu (Muscovite), Ch (chlorite), Il (illite), Ve (vermiculite), Hy (hydrobiotite). Minerals occurring in very small amounts, with minor effect of weathering rates, are not included in the table. Input data was not found for Gårdsjön C. The mineralogy has in some cases been slightly simplified, to make it fit in one table. For detailed mineralogy, see original references (Table 1).

Site	Qz	Or	Pl	Am	Ep	Bi	Ap	Mu	Ch	Il	Ve	Hy
Gårdsjön A	36.5	4.7	26.4	1.0	3.6	0.0	0.2	8.4	1.4	12.2	3.5	0.0
Gårdsjön B	56.2	19	16	1.5	1.0	0.5	0.3	0.0	0.4	0.0	5	0.0
Svartberget A	42.4	15.1	29.1	1.2	2.9	1.6	0.2	0.0	0.8	4.5	1.9	0.0
Svartberget B	60.3	7.6	16	7.7	2	0.0	0.4	0.0	2	0.0	4	0.0
Vindeln	39.1	9.8	24.3	1.0	2.3	1.2	0.3	0.0	0.7	17.1	5.4	0.0
Risfallet A	45.4	13.5	26.2	0.9	2.6	0.0	0.2	3.8	2.1	3.4	3.2	0.0
Risfallet B	44.9	25.0	26.0	3.5	0.0	0.0	0.1	0.0	0.5	0.0	0.0	0.0
Fårahall	30.7	29.0	28.0	4.0	0.0	3.0	0.3	0.0	0.0	0.0	5.0	0.0
Stubbetorp	48.5	30.0	15.0	2.4	0.0	0.2	0.2	0.0	1.1	0.0	0.0	0.0
Flakaliden[a]	41.5	14.4	25.9	3.5	1.8	2.1	0.0	3.8[b]	1.4	0.0	0.4	1.3
Asa[a]	43.2	15.3	26.7	2.3	3.2	0.2	0.0	3.0[b]	1.3	0.0	1.0	0.5
Bodafors	41.0	13.2	25.8	1.2	3.8	0.0	0.5	3.6	2.5	1.6	5.5	0.0
Hjärtasjö	49.3	3.0	20.4	0.8	1.9	0.0	0.2	9.4	1.9	9.1	4.1	0.0
Hässlen	40.2	15.1	21.8	1.0	2.5	0.0	0.2	6.3	2.1	6.6	3.6	0.0
Kloten	51.5	13.6	21.8	0.5	3.0	0.0	0.2	3.4	1.0	3.0	1.3	0.0
Kullarna	39.1	15.3	25.6	1.0	2.9	0.0	0.2	5.5	1.8	4.8	2.5	0.0
Lammhult	37.9	14.6	29.0	1.5	4.2	0.0	0.4	2.4	2.0	1.2	4.9	0.0
Skånes	38.8	16.5	29.7	0.7	2.4	0.0	0.3	4.3	1.0	2.2	1.4	0.0
Stöde	40.5	4.7	23.4	1.9	3.7	0.0	0.4	9.9	3.6	9.3	2.5	0.0
Söderåsen	48.9	10.2	21.3	0.5	1.4	0.0	0.3	9.9	0.7	4.9	0.9	0.0
Västra Torup	44.0	17.0	22.6	0.9	2.3	0.0	0.3	3.3	1.3	1.9	1.8	0.0
Hissmossa	37.0	18.0	23.7	0.7	2.0	0.0	0.2	5.5	0.9	3.7	1.3	0.0

[a] Mineralogy has been calculated with multiple models, but only the XRPD results are presented here.

[b] Muscovite and illite could not be separated with the XRPD method.

Table 3~~1~~: Weathering rates (meq m^{-2} ~~y~~y^{-1}) and statistics at the sites ~~where at least two approaches for estimating weathering rate have been applied to the same depth~~described in Table 1~~.~~

Site	PROFILE	Depletion	Budget	Sr	Tot. anal. regr.	MAGIC	ForSAFE	Median	Min-Max	Max % diff$_a$
Gårdsjön A	52	41	-	-	-	-	-	47	41-52	±12
Gårdsjön B	57	53	54	-	44-53 (49)	62	-	54	49-62	+15
Gårdsjön C	37	-	36	39	38-42 (40)	-	-	38	36-40	±5
Svartberget A	38	17	-	-	-	-	-	28	17-38	±38
Svartberget B	42	31	85	35	-	-	-	39	31-85	+121
Vindeln	30	13	-	-	-	-	-	22	13-30	±40
Risfallet A	68	29	-	-	-	-	-	49	29-68	±40
Risfallet B	29	-	-	25	-	-	-	27	25-29	±7
Fårahall	60	60	-	-	-	-	-	60	60-60	±0
Stubbetorp	67	-	-	-	35-51(43)	30-40	-	43	35-67	+56
Flakaliden	43	~~22~~34	61~~9~~	-	-	-	-	43	34-61	+42
Asa	37	11	131~~7~~	-	-	-	-	37	11-131	+254
Bodafors	41	22	-	-	-	-	-	32	22-41	±30
Hjärtasjö	29	20	-	-	-	-	-	25	20-29	±18
Hässlen	52	18	-	-	-	-	-	35	18-52	±49
Kloten	42	11	-	-	-	-	-	27	11-42	±58
Kullarna	36	16	-	-	-	-	-	26	16-36	±38
Lammhult	35	33	-	-	-	-	-	34	33-35	±3
Skånes Värsjö	37	8	-	-	-	-	-	23	8-37	±64
Stöde	41	18	-	-	-	-	-	30	18-41	±39
Söderåsen	36	19	-	-	-	-	-	28	19-36	±31
Västra Torup	58	-	-	-	-	-	63	61	58-63	±4
Hissmossa	25	-	-	-	-	-	21	23	21-25	±9

~~$_a$ Including O-layer~~

~~$_b$~~$_a$ ~~Not including O-layer~~Maximum difference from median (plus indicates that the maximum difference is higher, minus indicates that it is lower).

~~$_c$ For MAGIC the weathering rate was calculated to 0.6 m, including O layer~~

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Table 4: Classification of the sites with soil depth of approximately 0.5 m, in four classes, baased on intervals used in the critical load work of CCE (Coordination Centre of Effects) within the UNECE Convention on Long-range Transboundary Air Pollution (LRTAP Convention) (de Vries, 1994; Umweltsbundesamt, 1996).: very low, low and intermediate weathering rates and a group with non-conclusive results, i.e. that did not fit into any of the other groups. Sites were placed in one of the three weathering groups if the median fell within the main interval given, and if the maximum and minimum values fell within the extended interval (±5). A and C refer to different profiles, with different soil depths.

	Very low weathering rates	**Low weathering rates**	**Intermediate weathering rates**	**Non-conclusive results**
Interval	**10-37.5[a] (±5) meq m^{-2} y^{-1}**	**37.5-60[b] (±5) meq m^{-2} y^{-1}**	**≥60[c] meq m^{-2} y^{-1}**	
Sites	Vindeln	Gårdsjön A (0.5 m)	Fårahall	Hässlen
	Hjärtasjö	Gårdsjön C (0.47 m)		Risfallet A
	Söderåsen	Västra Torup		Asa
	Stöde	Flakaliden		
	Kullarna			
	Svartberget A			
	Bodafors			
	Lammhult			
	Skånes Värsjö			
	Kloten			
	Hissmossa			

[a] Corresponding to the lowest span in Fig. 3, 'acidic/intermediate parent material, coarse-textured'.

[b] Corresponding to the lower part of the two spans 'acidic/intermediate parent material, medium-textured' and "basic parent material, all grain sizes" in Fig. 3.

[c] Corresponding to the upper part of the span 'acidic/intermediate parent material, medium-textured' and the lower-intermediate part of the span 'basic parent material, all grain sizes' in Fig. 3.

Table 5: Weathering rates estimated with different approaches and harvest losses at stam and whole-tree harvesting (meq m^{-2} y^{-1}).

Site	Tree species	Harvest losses		Weathering			
		CH	WTH	PROFILE	Depletion	Budget	Tot. anal. reg.
Svartberget A	Spruce	13	21	38	17		
Flakaliden	Spruce	12	19	43	34	61	
Stöde	Spruce	19	32	41	18		
Kullarna	Spruce	18	30	36	16		
Hjärtasjö	Spruce	33	55	29	20		
Gårdsjön A	Spruce	33	55	52	41		
Gårdsjön B	Spruce	31	52	57	53	54	48.5
Bodafors	Spruce	37	62	41	22		
Lammhult	Spruce	34	56	35	33		
Asa	Spruce	42	70	37	11	131	
Skånes Värsjö	Spruce	41	67	37	8		
Vindeln	Pine	10	13	30	13		
Risfallet	Pine	20	25	68	29		
Kloten	Pine	17	22	42	11		

For

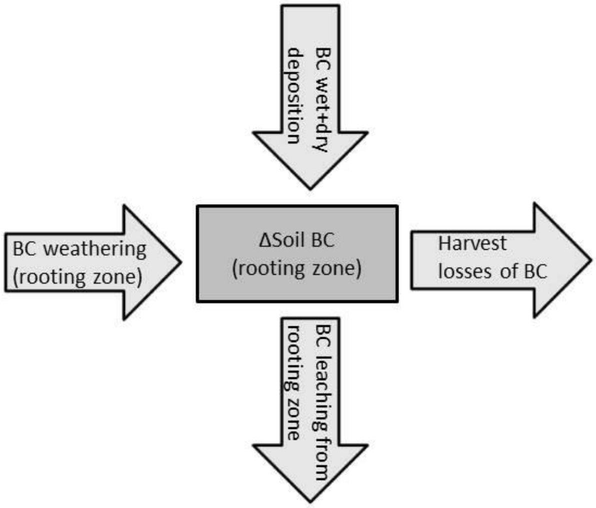
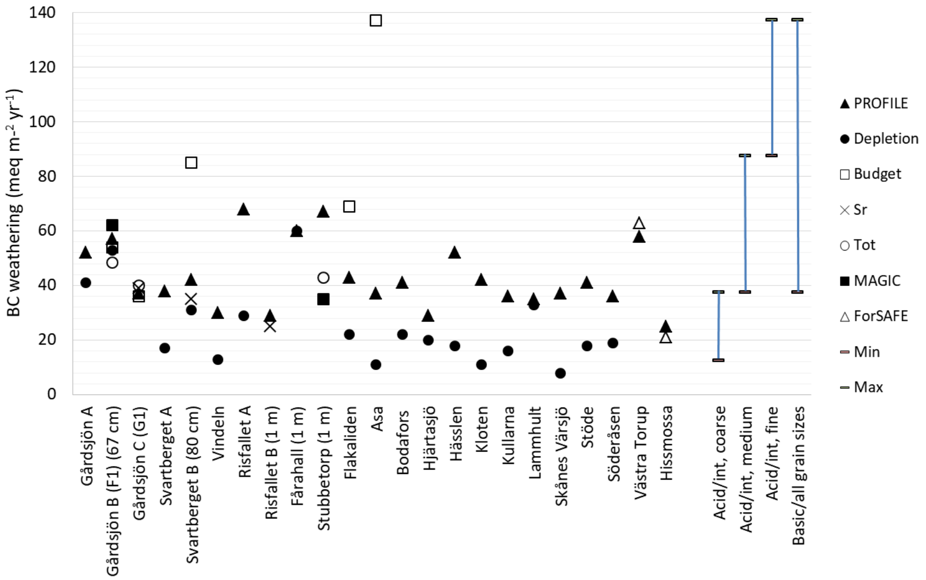


Figure 1: Base cation (BC) mass balance for the rooting zone of a well-drained forest soil. ΔSoil BC is the sum of the net change in BC in soil solution, the net change in soil exchangeable BC and the net change in soil organic material BC.

Figure ~~1~~2: Sites where weathering rates have been calculated for the same soil depth with at least two different approaches. See also Table ~~1~~ 1~~and Fig. 2~~.


BC weathering (meq m^{-2} yr^{-1})
140
120
100
80
60
40
20
0
Gårdsjön A
Gårdsjön B (F1) (67 cm)
Gårdsjön C (G1)
Svartberget A
Svartberget B (80 cm)
Vindeln
Risfallet A
Risfallet B (1 m)
Fårahall (1 m)
Stubbetorp (1 m)
Flakaliden
Asa
Bodafors
Hjärtasjö
Hässlen
Kloten
Kullarna
Lammhult
Skånes Värsjö
Stöde
Söderåsen
Västra Torup
Hissmossa
Acid/int, coarse
Acid/int, medium
Acid/int, fine
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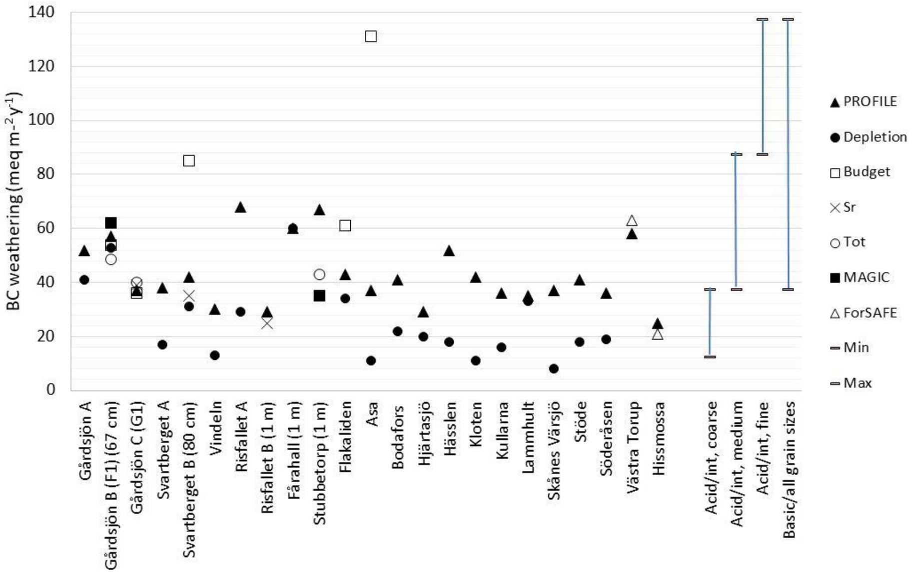

Figure ~~2~~3: Base cation weathering rates (sum of Ca, Mg, Na and K) for sites where different methods have been applied for the same depth on the same site (Fig. ~~1~~2). The soil depth is around 0.50 m (with or without organic layer, see Table 1), except for a few cases where greater depths are given. The four spans to the right are intervals that were commonly used in the critical load work of CCE (Coordination Centre of Effects) within the UNECE Convention on Long-range Transboundary Air Pollution (LRTAP Convention) (de Vries, 1994; Umweltsbundesamt, 1996). The intervals correspond to weathering rates for different parent material classes: acidic, intermediate and mafic, and different texture classes: coarse, medium (including the mix between medium and course material) and fine (including the mix between fine and medium material).

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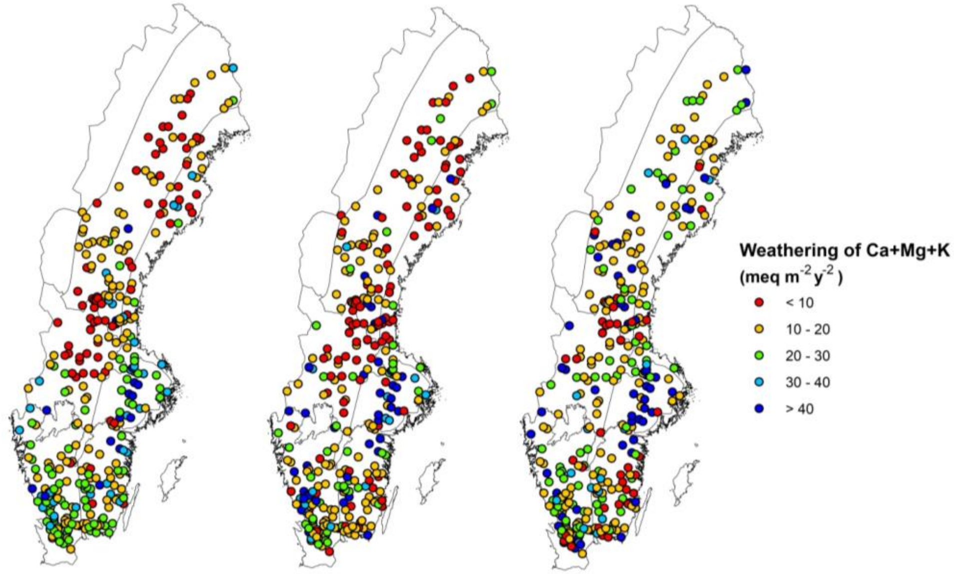


Figure ~~3~~4: Weathering rates calculated with the D~~d~~epletion method/Total analysis regression approach (left), modelled with PROFILE (centre) and with ForSAFE (right), in seven climate regions in Sweden, delimited by black lines.

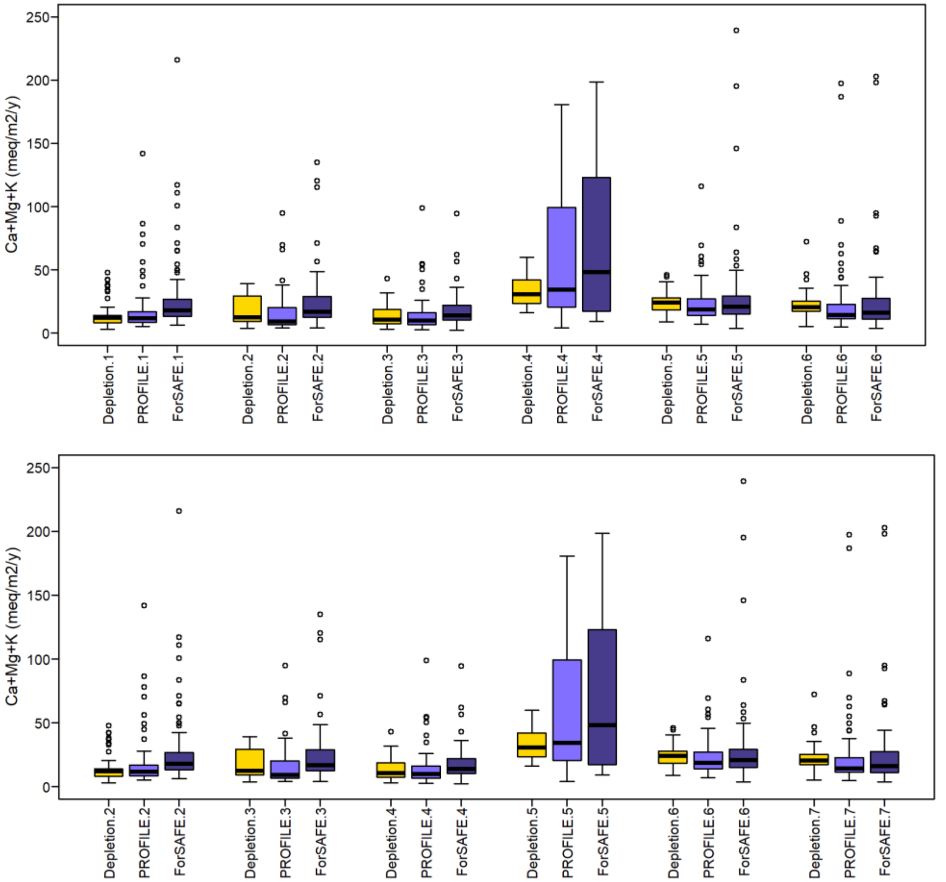
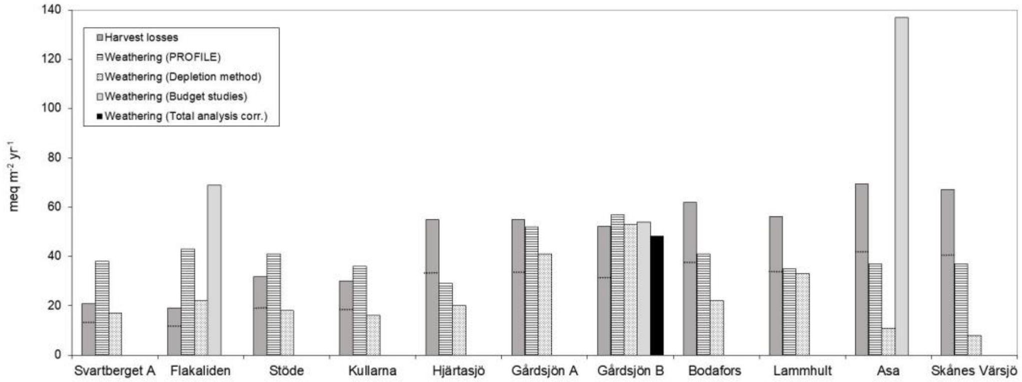


Figure ~~4~~5: Box plots for the three methods and for the climate regions: (~~1~~2) Inner part of northern Sweden, (3~~2~~) Coastal part of northern Sweden, (4~~3~~) Western part of central Sweden, (5~~4~~) Eastern part of central Sweden, (6~~5~~) Southwestern Sweden, and (7~~6~~) Southeastern Sweden. The Northwestern mountain region (1) was excluded since it only contained one site.



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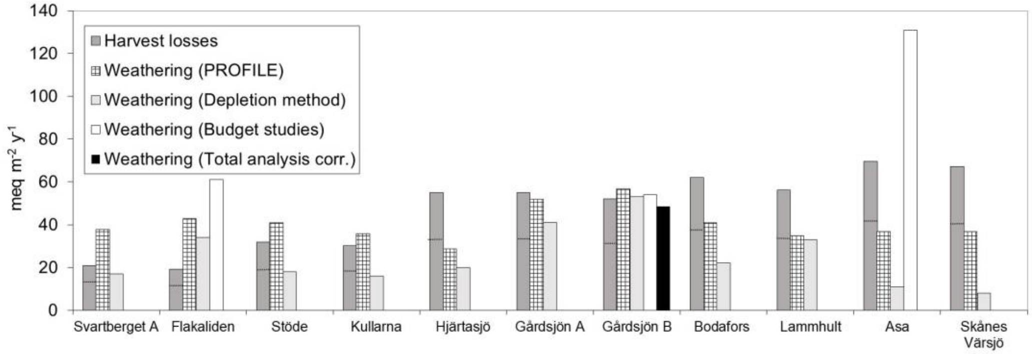
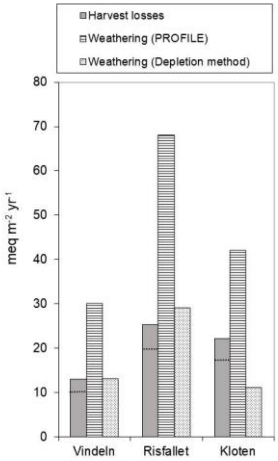


Figure 6~~5~~: Weathering rates of base cations calculated with different methods on spruce sites, compared with harvest losses of base cations at whole-tree harvesting (100 % of the stems and 60% of the branches harvested, 75% of the needles on the branches removed). The horizontal dashed lines in the harvesting bars show the levels for stem-only harvesting. The sites are ordered from north to south.



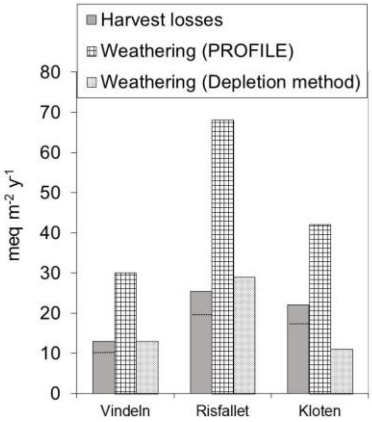


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Figure ~~6~~7: Weathering rates of base cations calculated with different methods on pine sites, compared with harvest losses of base cations at whole-tree harvesting (100% of the stems and 60% of the branches harvested, 75% of the needles on the branches removed). The horizontal dashed lines in the harvesting bars show the levels for stem-only harvesting. The sites are ordered from north to south.