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Interactive comment on "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" *by* Eric McGivney et al.

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RE3 = referee 3 ; AU = author's response, (y) = comment no. y

RE3(1): However, the main question I have is whether the actual values reported are in any way meaningful. Throughout the paper the authors report changes that are relatively small (e.g. "At Aneboda, the pH across all horizons dropped by an average 0.13 (WTH) and 0.12 units (CH) compared to the NH scenario by the year 2080"). However when I look at the calibration figures and simulation figures (3-5) it is quite

clear to me that parameters such as pH and base cations are very poorly matched. For example, Figure 3b shows simulated Mg in soil solution and observed values for the various horizons – they don't match very well and I don't see any clear separation of the observed data by horizon.

AU(1): Thank you for this comment. You are correct that we should have provided a discussion of why the model predictions did not always agree with the observations, we regret that we didn't in the first version of the manuscript. For Aneboda the results from individual lysimeters differ considerably, which is important to note when addressing the discrepancies, and therefore we have included a new supplemental figure (S4) that shows the individual lysimeter results. We have now included text towards the end of section 3.1 that addresses these issues, as follows: "For the Gårdsjön and Kindla sites, the modelled results align well with the lysimeter data, with a few exceptions. Interestingly, the model underestimated Mg^{2+} concentrations at both sites. This could be caused by the use of A2M estimates, i.e., that the normalization model underestimated the presence of easily weathered Mg-containing minerals. In a recent study, Casetou-Gustafson et al. (2018) compared A2M with the mineralogy obtained by X-ray powder diffraction (XRPD) for two soils that were similar to the soils studied here. They found that trioctahedral mica and hydrobiotite were consistently underestimated by A2M, which is consistent with our modelling results as these Mg-containing minerals have relatively high weathering rates. Moreover at Kindla, SO_4^{2-} concentrations were underestimated. There may be several explanations, but one possibility is mineralization and oxidation of organically bound S (Löfgren et al., 2001, 2014). The delay in SO_4^{2-} decrease at Kindla was, however, predicted well in the B1 horizon of the model.

For the Aneboda site, the discrepancies between model and observations were more substantial. For example, while SO_4^{2-} and pH were grossly underestimated, Ca^{2+} and Mg^{2+} were overestimated. It is important to note that the lysimeter data plotted in Figures 1, 2 and 3 (previously 3, 4 and 5) are averages based on data from several lysimeters, and it has previously been observed that there are large variations in the results of individual lysimeters at the Aneboda site (Löfgren et al., 2010; Löfgren et al., 2011; Löfgren et al., 2014). As an example, for the B horizon the averaged results are based on 8 lysimeters. Three of these, nos. 7102, 7104 and 7105, had results that were clearly divergent from the others (Fig. S4, Supplement). Dissolved SO42-, Ca2+ and Mg2+ were all considerably higher, whereas the pH was lower. Possible reasons include a net mineralization and oxidation of organically bound sulphur in response to decreased S deposition (Löfgren et al., 2001, 2014), a process which was not taken into account in the model. It may also be observed that if the results from the three lysimeters were removed, there would be a clearly improved agreement between the model and the observations."

RE3(2): Likewise, there is no Al3+ chemistry shown nor any description of how well the model simulations match the observed chemistry for the various soil horizons.

AU(2): In this case, the reason is that there were no measured Al3+ lysimeter data available.

RE3(3): Hence, the authors have a model that performs as expected but I have no confidence in the actual numbers nor the timeframe of the reported changes. Without a detailed evaluation of the model performance that provides the reader with some confidence that the simulated changes are in any way meaningful I cannot recommend that this paper should be published.

AU(3): As mentioned in the response to comment no. 2, we have now provided additional discussion of the model performance. Moreover, we never intended to give the impression that we thought our model was perfect. We do believe, however, that the model gives an idea of the direction of the changes, and of the relative significance of acid rain and forest harvesting, as the underlying chemical processes are well known and integrated into the model (with the exception of N). However, we have now included text that underlines the uncertainty of the model, above all in the following text in section 3.2: "The absolute magnitude of the model-predicted changes is of course

uncertain, not least in the light of the mixed success of the model to predict the available lysimeter data, as discussed in the previous section. Nevertheless, the simulated results suggest that the acidification due to a harvesting event at 2020 would be less impactful, over the time range studied, than that of historic atmospheric acidification."

RE3(4): Section 2.6 describes model calibration but looking at the figures it seems to be a very poor match for the various horizons.

AU(4): See response to comment no. 1

RE3(5): Quality of Figures is poor.

AU(5): High quality images will be uploaded.

RE3(6): The authors show several spikes in soil solution chemistry but these cannot be validated.

AU(6): The spikes are reported also in previous studies and have been mentioned in the manuscript:

Line 19, page 7: "The anomalous blip in pH (and BC) at Aneboda occurring between 2008 and 2018 is the result of a large storm that downed many trees in 2005, followed by a bark beetle infestation, resulting in acidification and increased dissolved Ca2+ (Löfgren et al., 2014). " Line 31, page 4: "The dips in deposition at Gårdsjön in the early 1900s and Kindla around 1890 were due to historical harvesting events. The dips that occur in 2020 were due to the simulated harvest scenarios."

RE3(7): There is no real discussion – the results/discussion section is primarily a description of the simulations and how the model is parameterized.

AU(7): See response to comments no. 1 and 3

RE3(8): The reported weathering rate changes (see Figure 6 and 7) are so small as to be insignificant compared with other inputs/outputs (deposition/plant uptake) and of course there is no way of knowing whether this is actually happening (assumes

PROFILE is correct).

AU(8): Thank you for bringing this up, the changes are indeed very small in comparison to other inputs and outputs. We have considered this more closely, and you are quite right that the main message of this part of the manuscript should more logically be that the model-predicted weathering rates remained more or less the same, both during the historic acidification era and during the harvest scenarios. This has prompted us to rewrite certain sections in which we discuss these results, e.g., as follows: Abstract, last lines: "In general, the predicted changes in weathering rates were very small, which can be explained by the net effect of decreased pH and increased Al3+, which affected the weathering rate in opposite ways. Similarly, weathering rates after the harvesting scenarios in 2020 remained largely unchanged according to the model." Section 3.3: "During the historic acidification period, the annual weathering rates at several sites and horizons (Aneboda: E; Gårdsjön: E, B1, and B2; Kindla: B1 and B2) actually decreased (Fig. S5), although not by much. This is due to the brakes in the weathering function (i.e. from Al3+ and BC, Eqs. 2 and 3) and to the fact that total dissolved Al and BC were much higher during this period (Figs. 1, 2 and 3). In other words, the increased weathering rate expected from a decreased pH was offset by the increase in dissolved Al and BC, resulting in a very small net effect, which according to PROFILE was negative. However, the exact patterns varied from site to site, and from layer to layer. The low weathering rates in the E horizon at Gårdsjön were likely due to its mineralogy, i.e., minerals other than quartz, K-feldspar, and plagioclase were essentially absent from the E horizon, but they were present further down in the profile. Another factor leading to low weathering rates in the Gårdsjön E horizon was the relatively thin layer thickness.

Contrary to the decrease in weathering rates during the historic acidification of the 1970s, the simulated weathering rates after the harvesting scenarios at 2020 generally increased compared to NH by 2080 (Fig. 5) although again, the net change was very small. However, the dynamics were quite different across each layer and site. The B1

horizon at Aneboda saw the strongest increase in weathering rates after harvesting, increasing by 9% (CH) and 11% (WTH) by 2080 compared to the NH scenario. The other horizons at Aneboda (E and B2) had almost unchanged weathering rates (<2% increase by 2080 compared to a NH scenario). It is worth noting that the difference between harvesting events and NH at the Aneboda B1 and B2 horizons would likely continue to diverge beyond 2080. At Aneboda, by the year 2080, the sum of weathered BC for the CH and WTH scenarios were 32 (CH) and 46 (WTH) meq m-2 higher, respectively, than the BC weathered in the NH scenario (Fig. S6). To put this into perspective, this difference is equivalent to only 1.1 % and 1.7 %, respectively, of the atmospherically deposited BC in the WTH plots over the same period. Such a small change in the weathering rate cannot be experimentally verified, and is unlikely to be of any ecological significance."

Consequential changes were made also in the remaining part of section 3.3, when discussing the results of Gårdsjön and Kindla, and also in the Conclusions.

RE3(9): Ignoring N dynamics is an issue – trees take up N and it can change both because of deposition and/or harvesting.

AU(9) To what extent this is an issue depends largely on to what extent nitrification (and subsequent nitrate release) is occurring. We acknowledged this already in the first version of the manuscript, but to make it clearer we have also added more text discussing this. Here are some examples:

Section 2.6, immediately before Results and Discussion: "However, at this point it needs to be added that these conclusions may not be relevant in cases when nitrification following harvest is substantial, in which case the acidification effect could be considerably larger; this possibility was not considered in our simulations. Most Swedish forests are N-limited (Högberg et al., 2017), but increased nitrate concentrations are found in soil solution for some years after final felling. Nitrification is dependent on site productivity, which is between 4 and 8 m3 ha-1 yr-1 in the sites studied. According to the estimates of Futter et al. (2010), the total accumulated harvest effect should generally not exceed 220 and 500 meq NO3- m-2 for site productivities of 4 and 8 m3 ha-1 yr-1, respectively (Futter et al., 2010), indicating rather modest nitrification effects on the long-term acid-base status of the soils. As an example, this value represents between 5 and 15 % of the atmospherically deposited BC over a full rotation period, hence nitrification is a relatively minor proton source as compared to other processes in the forest soils under study."

Page 13, lines 15-18: "As mentioned previously, limitations in the model prevented us from addressing possible nitrification effects resulting from long-term N deposition, which may influence these results. A future task is to upgrade the HD-MINTEQ model to include N transformations, so that the effects arising from e.g. N deposition and nitrification can be more accurately assessed."

RE3(10): Conclusions – first sentence depends where in the world you are.

AU(10): This sentence has been removed, as its statement was debatable and in addition not necessary for the paper.

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