

Biological weathering and its consequences at different spatial levels – from nanoscale to global scale

The ms is intended as a review of current progress made in understanding biological processes contributing to weathering. It is based on a very extensive reading in the literature (I counted 215 references) on a very wide range of topics, including chemistry, physics and biology, combining empirical and modelling approaches, based on a large range of experimental techniques. The authors also make a very laudable attempt to scale up, both spatially (as indicated by the title) and temporally, when they link past and current weathering processes as an evolutionary and ecological force. The ms is also very well written. Despite all this initial praise, reading the ms did not fully satisfy me. In my view this is to a large extent due to the fact that it succeeds very well as a review, but succeeds to a lesser extent as a synthesis. Several empirical observations seem to contradict other observations, and one would like to read how much consensus has been reached on the biology of weathering. In that respect I found the final section (key questions and knowledge gaps) somewhat disappointingly short. Considering the lack of consensus on the importance or generality of several processes, a more cynical reader may easily be inclined to think that almost anything goes in biological weathering. In fact this kind of mild cynicism is almost encouraged by the authors: after having presented so many data the authors (p. 17, l. 33 – p. 18, l. 1; note that one of the authors of the paper referred to is also an author of this ms) state that "Smits & Wallander (2017) consider that there is *no clear evidence* [emphasis mine] that processes at the laboratory-scale play a significant role in soil-scale mineral dissolution rates". If so, what is the main message of this paper?

Let me try to back up my dissatisfaction with a couple of general observations. Before providing more detailed comments.

1. The point of departure for the study is that weathering is the only or main supplier of base cations and phosphorus to compensate for losses through harvesting and leaching. However, on p. 17, l. 21 the reader is informed about atmospheric deposition (the only mention of this input source) where we are informed that a study found that atmospheric deposition was four times as important as weathering; and that the weathering flux was less than 0.3% of calcium uptake. This statement then raises questions about its quantitative importance over ecological time scales and evolutionary time scales, an issue treated very implicitly at best.
2. Despite the generality of the title (biological weathering), the focus is almost exclusively on the role of mycorrhizal fungi plus associated mycorrhizosphere bacteria and the trees with which they associate (I like the focus on the plant as holobiont). Lichens, generally considered as major weathering agents in the first stages of primary succession, are mentioned only once (p. 12, l. 10-12). There the authors state that "the ubiquity and significance of lichens (...) as a model for understanding weathering (...) are well understood." However, the reader is not

informed about this understanding, nor is (s)he informed whether fungal weathering is similar or dissimilar from lichen weathering in any significant respect. There is also very limited attention for fungi other than mycorrhizal fungi, however from an evolutionary perspective this is a missed opportunity. A fungus often used in weathering studies is *Paxillus involutus*, a species derived from a clade of brown-rotting fungi characterized by oxalate production. It could be possible that the ability to produce and excrete oxalate in the environment evolved for different purposes and was even maintained in the ectomycorrhizal groups in this clade for different purposes.

3. There are many parts in the ms where the possible difference in weathering ability between arbuscular mycorrhizal fungi+plants and ectomycorrhizal fungi+plants are mentioned. Some of these are quite explicit in suggesting that the ectomycorrhizal symbiosis allows higher weathering rates than the arbuscular mycorrhizal symbiosis. However, we also learn that weathering evolved in the arbuscular mycorrhizal symbiosis (p. 11, l. 26) and that some studies did not find differences in weathering rates under ectomycorrhizal and arbuscular mycorrhizal vegetation (p. 21, l. 26-27). The reader of his paper will therefore remain in doubt what the current consensus view is (if there is consensus), what likely hypotheses exist to explain such different data and what kinds of research approaches exist to resolve that issue. (One option would be a common-garden experiment with sister clades of plants with the different mycorrhizal symbioses, in analogy of the approach by Koele et al. (New Phyt. 196: 845-852. 2012) when they tested for stoichiometric differences (leaf N:P ratio) between both guilds. I am sure there must be other ways to make progress as well.) Another group of mycorrhizal fungi + plants, which form the ericoid mycorrhizal symbiosis, is mentioned once (p. 7, l. 21) even though they have been suggested to be strong weathering agents as they can produce copious amounts of low-molecular-weight organic acids (Martino et al., Soil Biol. Biochem. 35: 133-141. 2003).
4. Addressing (and putatively answering) the question of the role of different mycorrhizal symbioses in weathering is, in my view, particularly relevant when it comes to understanding mechanisms. If weathering is driven by the production of LMWOA and siderophores, then it should be clear that the ectomycorrhizal symbiosis is much more important for weathering than the arbuscular mycorrhizal symbiosis (as AMF have not been reported to produce LMWOA, the AMF symbiosis has been reported to downregulate LMWOA production by plants (Ryan et al., Plant Cell Environ. 35: 2170-2180. 2012), and AMF do not produce siderophores as far as I know). If other mechanisms are more important (e.g., acidification driven by excess uptake of cations over anions and proton exudation to maintain charge balance; or dissolved CO₂ as a consequence of respiratory activity), the contribution by both guilds could be more important – with differences still related to the amount of extraradical hyphal biomass and / or respiratory activity.
5. The issue about the relative importance of weathering mechanisms has been debated since mycorrhizal researchers entered that field in the early 2000s. When

enthusiastic claims were made for a major role of mycorrhizal fungi (and I admit having been such an enthusiast as well), these ideas were criticised by Sverdrup, who essentially claimed that weathering was driven by CO₂ flux and that the contribution by ectomycorrhizal fungi was around 2%. While his claim has been challenged (Van Schöll et al., *Plant Soil* 303: 35-47. 2008), I think this review would have been a good place to synthesise current understanding. Sverdrup (cited in the ms – pls note that the journal has Volume 23, Issue 4; not Volume 4) has maintained his suggestion about the major importance of respiration / CO₂ production, rather than the production of organic acids) as the driver for weathering, stating: “the growth of trees represents quantitatively largest single biological process that can affect weathering, followed closely by decomposition of organic matter.” It is evident that the authors of this ms disagree with Sverdrup, however, without fully discussing this alternative view. I think this is a missed opportunity. The same applies to the origin of pores, with Sverdrup claiming that they are of abiotic origin (as cited in p. 3, l. 30). How would the authors of this ms evaluate our current knowledge and understanding? (Note that because of the extent of tunnelling the contribution to weathering might be limited, irrespective of the question on their origin.)

6. While I agree about the importance of upscaling, both spatially and temporally, I think that progress depends on the extent to which we can quantify rates. Unfortunately, the paper is quite frugal in giving numbers. This may give the impression that despite such many studies there has been little progress in quantifying processes. That conclusion seems also implied in p. 1, l. 31-32 (“opinion appears to be divided with respect to the quantitative significance [of interactions between microbes and minerals]”). If opinions are divided, please give equal hearing to arguments from both sides. But if a clearer picture has emerged in the view of the authors, please provide more quantitative detail. In order to have feedback mechanisms to work over both ecological and evolutionary times, we need such data.
7. The authors refer (p. 2, l. 28) to twelve testable hypotheses on the geobiology of weathering. It would help the reader to list those (rather than to invite them to look up the paper themselves) and to indicate to what extent their review helps addressing these hypotheses. For instance hypothesis 8 (elevated CO₂ will enhance weathering) seems to assume that weathering fluxes and its ultimate consequences of drawdown of CO₂ occur at very different time scales, which could put constraints on feedback mechanisms postulated in the ms. With respect to hypothesis 2, the importance of stoichiometry, I think that the studies done of mycorrhizal weathering provide much of the needed data. In none of their hypotheses they draw attention to different kinds of mycorrhizal symbiosis (but this could be a refinement of hypothesis 1), although it may not be coincidental that their figure 1 depicts an ectomycorrhizal conifer...

Some of these comments will make the manuscript longer, so I think it may help if I indicate cases where shortening of the ms is possible. I noted several digressions (also in the Abstract) that result in a less focused paper. Examples are: reference to acquisition of N and P by mechanisms other than weathering (p. 7, l. 18-34), hydraulic lift / redistribution (p. 9, l. 6-8), oxygenic and anoxygenic photosynthesis (p. 11, 14-24), autotrophic and heterotrophic respiration in forests (p. 13, l. 30 – p. 14, l. 6; unless the authors think that Sverdrup is, essentially, right...), differential carbon storage in ectomycorrhizal and arbuscular mycorrhizal forests (p. 14, l. 4-28; note that of the two biomes where both guilds occur larger C storage per unit N was shown for the temperate biome, not for the (sub-)tropical biome – so we should not take Averill's claim too seriously), nitrogen in the rhizosphere (p. 24, l. 1-2).

Page-by-page comments

p. 3, l. 9 Is the Finlay & Clemmensen paper on biogenic weathering? The title of the paper would suggest otherwise

p. 5, l. 14 Please provide a reference for the suggestion about the importance of horizontal gene transfer in such microbial consortia in EPS.

p. 7, l. 1 Here I disagree. In such habitats, in case of a low pH, plants with cluster roots (or proteoid roots; I think they are the same) or dauciform roots produce carboxylates that *desorb* phosphorus from mineral surfaces. But desorption is not weathering, dissolution of minerals. Weathering would happen in the case of high-pH with calcium phosphates; in low-pH soils P is far too scarce to form substantial amounts of Fe- and Al-phosphates that are weathered.

p. 9, l. 14 In the light of current criticisms of humic and fulvic acids as large molecules (Lehmann & Kleber claim these to be aggregates of essentially small molecules) this statement may need reconsideration in terms of underlying mechanisms.

p. 15, l. 21 When introducing the Blum et al. hypothesis, they should also refer to contradictory data by Dijkstra and Smits (now only referred to on p. 17, l. 18-22; however I interpret that paper as showing that Blum et al.'s conclusion is grossly overstated – but I would love to see the opinion of the authors of this ms).

p. 17, l. 19 Please provide a reference to that further study.

p. 19, l. 17 Note that exudation of carboxylates / organic anions can also have a major function in the desorption of iron-oxide bound soil organic matter and the subsequent acquisition of carbon (Keiluweit et al., *Nature Clim. Change* 5: 588-595. 2015) and nitrogen (Jilling et al., *Biogeochemistry* 139: 103-122. 2018) (and possibly phosphorus, as both inorganic and organic P are sorbed on such surfaces).