

## Comments to the author:

Editorial Comments on Mineral surface area in deep weathering profiles reveals the interrelationship of iron oxidation and silicate weathering by Fisher et al.

Thank you for the submission of your revised manuscript. I have now read it, along with the reviews and your detailed response to the reviews. Your dataset is clearly interesting and I think you have an important finding that the SSA (effectively a proxy for secondary Fe oxide minerals) shows a continuous increase towards the surface, similar to an elemental profile, but this is decoupled vertically from non-redox sensitive mobile elemental depletion profiles. The inference being that acid base and redox reactions are happening at different depths. However, I have to agree with many of reviewer 1's comments overall and even in its revised format this is still a challenging read and would really benefit from being substantially improved before it can be considered for publication. Putting in some more effort now will help this work achieve its full potential and get maximum citations in the future.

1. The presentation of the data really would benefit from having a single consolidated figure that illustrates the key patterns. It seems to me that one of the key observations (perhaps the most important one) is that the very nice depth trend with SSA does not mirror depth trends in either mineralogy or elemental depletion. Or rather, if I have understood, the depth trend in chemical depletion is in a different place to SSA increase. Although the chemical data is not new, I think it is really very important to see the SSA plotted alongside the elemental depletion plots from previous work. This certainly echoes some of the comments from R1. Demonstrating this point clearly with a single figure seems essential before going on to consider what might be causing the differences. This is shown schematically on Fig. 2 but I would consider an overhaul of Fig 3, adding literature data to be important. The mineralogy data should presumably mirror the elemental data and that could also be plotted as a function of depth. We are told something about plagioclase showing a subtle trend on line 685, but it is never plotted.

Response: I have removed the schematic figure and and generated some new plots that show the elemental distributions with the mineralogy plots. I have plotted the SSA trends in different ways that I believe more clearly show the relationship between different data types. In devising the new plots I didn't find a logical way to put so many element distributions in the the same plot as SSA, but I think the display of data is greatly improved from this set of comments.

I struggled to understand how the change in slope in SSA was below the soil to rock boundary but above the chemical depletion boundary (line 602). This seems a little misleading if you are conceptualising this in terms of the definitions in the paragraph beginning on line 69, where the colluvium may be been transported by gravitational processes. In this case, there is a soil (or source material) but with discontinuities present. The SSA changes appear continuous across this soil to rock boundary.

Response: I'm not sure what was misleading in the original iteration, but I think the new figures, which separate data from Well 1 and Well 2 helps with clarity for the reader. I also think the new figure with drill core photos alongside SSA data for the top 4.5 m also helps the reader see where the SSA changes occur, and on this figure I have placed the soil C horizon. I hope it no longer feels misleading that the SSA changes do not coincide with the field identification of soil morphology.

2. Potential explanations are considered for the change in SSA at ~3m. One of the issues that worries me a bit about this data set and interpretation is that the SSA is made up of both Fe oxides minerals as well as phyllosilicates. Clearly oxidation should enable Fe-oxides to form from primary minerals. Mechanistically however, are no mobile elements released during the oxidation process?

Response: By adding the figures with element mass balance data for the full depth profiles, we show how Fe and Al distribute with weathering, in which both seem to undergo a small amount depletion (the depletion profiles are all scaled the same, so this turns into a relative relationship compared to the largest depletions, Ca and Na). A second figure in the discussion section shows how the iron concentration hovers very close to 10% while the Al concentration decreases across the top 4.5 meters. These data reveal that iron appears to largely be retained in the weathering profile, but aluminum seems to be partly released.

It is interesting that oxidation reactions have been conceptualised as being entirely distinct from one another. I wonder the extent to which that is correct. For example, why is there no dissolved O<sub>2</sub> in the water that is transporting the carbonic acid to the elemental depletion front? Presumably, if you were to plot Tau\_Fe, it would remain constant throughout the profile, but the Fe is being redistributed between oxides and “primary” phases?

Response: I'm not sure which oxidation reactions were presented as distinct from one another. Regardless, dissolved O<sub>2</sub> making it to the elemental depletion front is not something I observe in weathering or soils literature. Although many authors try to separate the biotic and abiotic components of weathering, I struggle to imagine a context in nature which O<sub>2</sub> is not an energy source for organisms. Studies of soil and weathering ubiquitously measure and model O<sub>2</sub> decreasing with depth. Even if O<sub>2</sub> persists in subsurface fluid, as it penetrates deeper into the earth the concentration of O<sub>2</sub> in the water decreases and the biological and mineralogical reactions that consume O<sub>2</sub> are faster than diffusive process can recharge O<sub>2</sub>. (e.g. Brantley et al 2013).

3. One potentially interesting thought could relate to the kinetics of

dissolution/crystallisation. Some recent work has suggested that Fe oxides can form/recrystallise very rapidly. I wonder how this might play into the story.

It's true that oxide minerals have fast kinetics in the environment (as leveraged in stormwater and other remediation efforts), but I'm not sure how this contributes to the manuscript's main ideas of CO<sub>2</sub>/O<sub>2</sub> and SSA. I also note that the Piedmont weathering profile has had 12,000 years to develop since the last glacial maximum, which suggests that oxide mineral kinetics are not a limitation in this system. I am concerned that adding a discussion on kinetics would be far more speculative than the discussion of oxygen as the limiting agent for weathering in this setting.

4. Are there no other literature soil profiles that have looked at Fe oxide chemical extractions? SSA data might not be available, but one might imagine similar trends might have been detected by alternative methods. This is discussed relatively briefly in the paragraph beginning on line 764, but the key thing for me is how does this compare to element depletion profiles of the mobile non-redox sensitive elements.

I added the following from two studies where I was able to find extracts of profiles:

“Other studies of soil profiles with extractable oxides removed reveal that the distribution of extractable oxides depends on soil type and morphology. Aburto et. al. 2017 studied glacial deposits in the Tahoe region in which the amount of extractable oxides increased with decreasing depth, but unlike our profiles, the extracted oxides decreased in the uppermost 30 cm. In transects in loess deposits in southern Illinois, Wilson et. al. 2013 saw extractable oxides in most profiles reach their maximum in the zone of clay accumulation (Bt horizon), with little to no decrease in extractable oxide at the 2 m depth extent of their study.”

5. Tree rooting and frost damage are interesting to consider as explanations for the ~3m depth. However, fundamentally the elemental depletion profile is much deeper, suggest water, acids and likely O<sub>2</sub> should also have been able to penetrate deeper.

The discussion of tree rooting depth has been removed from the manuscript. The point of that discussion was to address how we might facilitate an oxygen penetration deep enough to explain the extractable oxides to 3 m deep. The notion that oxygen would be likely to be deeper is not one that I see in the literature.

In summary, this is an interesting manuscript and dataset that is in the road to improvement to make a clear coherent and important study. Some additional work is required before publication.