

## ***Interactive comment on “Decoupling silicate weathering from primary productivity – how ecosystems regulate nutrient uptake along a climate and vegetation gradient” by Ralf A. Oeser and Friedhelm von Blanckenburg***

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### **Reply to Referee 2**

**COMMENT:** the authors present total and bioavailable stocks of a number of nutrients and some other elements, in soil, saprolite, and vegetation along a steep climatic gradient in the coastal cordillera of Chile. From their data, they calculated a number of fluxes between the various ecosystem compartments and in and out of the whole

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system. Their main interpretations are that the weathering rates – which they term abiogenic – did not change substantially along the transect in spite of the marked climatic differences while the nutrient uptake rates increased with standing biomass following the rising rainfall rates. I think that the authors produced high-quality data from an interesting environmental gradient but I disagree with large parts of their data interpretation.

**REPLY** We are grateful for the reviewer to appreciate the production of high-quality data from a spectacular vegetation gradient. Critical Zone inorganic chemical data of both the weathering zone and the plants that grow on it are still not common, in particular of the rates and fluxes involved. Presenting these quantifications is one of the main objectives of this paper. Independent of our own interpretation our large data set shall allow other scientists to evaluate hypotheses of geo-bio links that was hitherto rarely possible for lack of data that include the rates of these processes that are very hard to measure.

Yet the reviewer disagrees with large parts of our interpretation, and in doing so implicitly disputes major concepts widely accepted in weathering geochemistry. In particular, the reviewer objects to our use of the term “abiogenic weathering”. We note that almost any textbook on weathering begins on the premise that rock weathering takes place through inorganic chemical reactions. In practice this was the case early in Earth’s history and is possible the case at our arid field site where there is barely any vegetation (but microbes), and is likely the case on Mars. .). However, we never state in the manuscript that abiogenic weathering is the only process. The objective of this paper is to evaluate whether biota accelerates, damps, or changes in any way the weathering chemistry that would have happened anyway by abiotic reactions. This is one of the grand questions regarding the long-term climate regulation of the Earth (Berner et al., 2003; Pagani et al., 2009). We will make this objective very explicit in a revised version to avoid this misunderstanding.

Furthermore, the reviewer highlights the different states of soil development between the sites (last part of review section 1) and thus contradicts anonymous reviewer 1 who

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suggests that all sites are within the same “pedogenic process domain”. It would be disconcerting having to embark in this discussion which seems to be about definitions, rather than about the quantification of biogeochemical fluxes and their interpretation. Of importance may be here the starting viewpoint: both the reviewers’ views seem to be based on the perspective of “soil development” on stable surfaces of different ages; implying continuous development through different states. But this is not the system we analyze here: our sites are continuously eroding and thus are not subject to different stages of one evolution. Material is continuously turned over. Such a system has no age, but rather a residence time. This is the contrast between two suggested models of soil development – steady-state and continuous evolution (Lin, 2010). In a revised version we will emphasize this important difference, and that here we deal within the steady state perspective.

In general, we realize that this MS requires better introduction of paradigms and assumptions that are the basis of our interpretation and how these differ from other common perspectives. Such introductory text will make this study more easily accessible to a multi-disciplinary readership. We thank the reviewer for making us aware of this deficit.

We reply point by point to the reviewer’s comments.

**COMMENT 1.1** It is hard to believe that weathering rates do not respond to the strong climatic gradient.

**REPLY** Indeed, this is really hard to believe, and it is a stunning result of this study. In particular the weathering rates between the barely vegetated semi-arid site and the fully vegetated humid site (where topographic relief and hence total denudation is similar) are almost the same, despite huge differences in climate. There is key information of biotas role in weathering here.

**COMMENT 1.2** From the explanation in l. 603-607, I read that the “most negative tau values from the shallowest mineral soil sample of each regolith profile were

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used” [to calculate weathering rates] and that these values are shown in red in Fig. A1. I do not understand this. The red symbols in Fig. A1 do neither refer to the shallowest horizon nor are they consistently the most negative value. Why did you choose the most negative tau value and not the thickness-weighted average tau value of the whole regolith profile?

**REPLY** We select the shallowest sample not affected by atmospheric input or plant recycling as it reflects integrated elemental release over each entire regolith profile in an eroding regolith column. This principle is discussed extensively for eroding soils in many publications (Granger and Riebe, 2014; Riebe et al., 2004; Dixon et al., 2009; Hewawasam et al., 2013; Wackett et al., 2018). Thus, we used the most negative tau value from the shallowest mineral soil sample (B Horizon) as the metric for the weathering-column integrated fractional mass loss.

**COMMENT 1.3** I anyway think that the tau values cannot be used to estimate weathering, because they lump together a complex mixture of many processes including e.g., leaching losses, deposition input, plant uptake, or soil-internal redistribution. The latter is particularly pronounced in Nahuelbuta, where Podzols occur which show strong depletion (E) horizons and strong accumulation (Bhs) horizons.

**REPLY** There are many different ways to define, study, and quantify weathering, and thus it is no surprise that the reviewer defines estimates of weathering differently than we do. However, we disagree with the suggestion that tau values cannot be used to estimate weathering rate (where we emphasize that rates are our main objective). Deriving weathering rates from CDF (fraction of total dissolved mass loss) or from tau (fraction of elemental dissolved mass loss) in conjunction with a denudation or soil production rate from cosmogenic nuclides is now common practice in many studies of Critical Zone Geochemistry (see also Brantley and Lebedeva, 2011; Ferrier et al., 2010; Uhlig and von Blanckenburg, 2019 in addition to the references named above), and the background paper on which this publication is based on (Oeser et al., 2018). Weathering rate is defined here as the net loss of solutes from regolith and is a figure

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that can be compared to e.g. river solute fluxes. Ultimately it is a metric used to calibrate global weathering fluxes. The reviewer instead seems to refer to “weathering” to denote the quantification of internal redistribution fluxes. This is an equally valid, albeit entirely different way of quantifying the state of weathering, but it is not what this study (and many other weathering studies) aims for.

**COMMENT 1.4** Instead, I think that an equation such as that proposed by Likens (2013), *Biogeochemistry of a forested ecosystem* (3rd ed). Springer, New York, USA should be used:  $TD_i + W_i = ST_i \pm \Delta B_i \pm \Delta OM_i \pm \Delta X_i \pm \Delta M_i$  where TD is total deposition, i a selected element, W is weathering release from primary minerals, Delta is change, B is storage in biomass, OM is long-term storage in soil organic matter, X is the exchangeable pool and M is the secondary mineral pool.

**REPLY** We acknowledge that the reviewer proposes a different suite of metrics to quantify the internal material fluxes in the Critical Zone (i.e. Eq. (2); Likens, 2013). Indeed, it would be very insightful to establish such budget. However, due to the setup of our study we are unable to determine all the variables required to solve this equation, which was never the aim of the study. We are aware that one of the successful studies in this regard (e.g. Wilcke et al., 2017) was based on many years of careful field monitoring of many ecosystem and water variables – something that is totally beyond the scope of this project. Some of these variables, like weathering release from primary minerals, are in any case very hard to determine in a field setting. We maintain that our method to determine budgets is equally valid, even though it targets different components of the system.

**COMMENT 1.5** I particularly think that the immobilization in soil organic matter and the formation of secondary minerals belong to the weathering rates.

**REPLY** We do not disagree that it would be beneficial to know these parameters for a full characterization of the weathering system. Besides the difficulties arising in establishing rates of these parameters we suggest that whether these parameters

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belong to the weathering rates is purely a matter of definition. For the question we ask – namely what are the net weathering release rates from regolith, they are not required.

**COMMENT 1.6** An advancing weathering from N to S is also reflected by the soil development ranging from Regosols (initial A-C soils) via Cambisols (A-B-C soils) to Podzols (A-E-Bhs-(Bw-)C soils), which goes along with increasing mineral formation and in the most advanced stage also translocation of organic matter and soil minerals. This is in line with the cited soil production rates.

**REPLY** It is not entirely clear to us what the suggested link between soil production rates and soil development would be telling us, besides descriptions of soil state variables that have been reported elsewhere (Bernhard et al., 2018). Maybe there is an underlying misunderstanding. As all our sites are continuously eroding (Oeser et al., 2018; Schaller et al., 2018; van Dongen et al., 2019) they do not reflect a continuous series of soil development in the sense of a “chronosequence”. To quantify the permanent material turnover, we report soil production rates in L140f and Table 1. They are lowest in the arid site and highest in the mediterranean site. In the semi-arid and the humid-temperate site, they are similar. In contrast to reviewer 1, reviewer 2 (this review) seems to attribute the soils to different process domains. We appreciate these contrasting views on the pedogenetic processes at the EarthShape sites. That the reviewers suggest to employ such opposing frameworks to argue against our determination of the degree of weathering makes designing a useful revision strategy difficult. We hope for further advice from the editor.

**COMMENT 2.1** I have also difficulties to understand what your “ecosystem nutrient uptake fluxes” are. Is this the current annual uptake or is it a mean of a certain period?

**REPLY** As reported in Table 2, Eq. (4) and Table 3, and explained in detail in appendix A the total nutrient uptake fluxes are reported in  $\text{mg m}^{-2}\text{yr}^{-1}$ . They are thus reported as annual uptake fluxes. However we also describe that NPP was derived from the

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LPJ GUESS model (Werner et al., 2018) and they thus reflect average Holocene values – a time scale relevant to regolith weathering.

**COMMENT 2.2** In l. 613, you mention net primary production (NPP) and one line later “GrowthRate”.

**REPLY** Thank you for pointing out this inconsistency which we will correct accordingly.

**COMMENT 2.3** Total nutrient uptake would, however, be related with gross primary production and growth rate sounds as if the standing nutrient stock is disregarded. Please clarify.

**REPLY** The derivation of these equations are thoroughly described in Uhlig and von Blanckenburg (2019) (as cited in L256). These authors state that  $U_{total}^X$  is calculated from NPP. This is a flux estimate, for which knowledge of the standing biomass stock is not required, since we do not evaluate short-term fluctuations in biomass.

**COMMENT 2.4** What you call “recycling rate” (i.e. uptake rate divided by weathering rate) seems to me to be more of an “accumulation rate”, because the vegetation accumulates part of the nutrients released by weathering and deposited from the atmosphere. When enough nutrients have accumulated to support a mature forest, the majority of these nutrients is recycled with losses smaller than the weathering release (or the deposition from the atmosphere on very poor soils) until the forest starts to break down and rejuvenate.

**REPLY** This statement is correct if we would be exploring an ecosystem featuring a growing stock of biomass, such that nutrient accumulate in this biomass. Of course, this may well be the case after e.g. deforestation, sustained drought, or wildfires. However, with the metrics we use, we evaluate, per definition, a much longer timescale. Our weathering release rates average over millennia, the model-derived NPP estimate over the Holocene, and even the residence time of the bioavailable pool is centuries or more (inevitably only the plant chemical concentrations we measure do not integrate over

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a longer time scale). Over these long timescales, elements can be safely assumed to be returned from biomass, taken up again, or, as the reviewer states, returned in a small fraction to the weathering flux. They thus do not reflect an accumulation rate.

**COMMENT 3.1** Because all sites are about 80 km away from the Pacific Ocean, they should be similarly affected by Sea Spray. I would even expect an increasing Sea Spray deposition with increasing rainfall. This can, however, possibly not be detected because of the simultaneously higher leaching rates at the wetter sites.

**REPLY** We do not understand this comment. We have determined atmospheric contribution relative to weathering input from Sr isotopes using a two-component mixing calculation using bedrock and seaspray as endmembers. Accordingly, up to 93 and 43% of Sr derived from seaspray are incorporated in the regolith profiles of Pan de Azúcar and Santa Gracia, respectively. In La Campana, this seaspray contribution is zero. In Nahuelbuta Sr isotopes do not discriminate such input. However, we regard it as unlikely as we see no elemental increase at the surface of the profiles that cannot be explained by bio-lifting. In particular Na as the most-abundant cation in seaspray is distributed uniformly with depth in the bio-available fraction at all sites except Pan de Azúcar (Table S4), which we interpret do show the absence of major net input of marine aerosol-derived elements. We discuss the impact of these results in the discussion and quantify them in Table 6. We further assume that even if there were an undetected component of that sea spray its input would immediately be flushed away at the wetter sites such that it may not be as relevant as the weathering input. In a revised version we will make these inferences more explicit.

**COMMENT 3.2** Nevertheless, the Sr isotope ratio in the plants can be explained as a mixture of the isotope ratio of the rock and Sea Spray at three of the four sites. The only exception is La Campana, where perhaps indeed the uptake from the subsoil dominates the Sr isotope ratio in the plant. Please also mind that plants can take up their nutrients with all surfaces and forests therefore retain nutrients in their canopy.

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Besides Sea Spray, I would also expect aeolian redistribution of soil material at the driest site. How can this be considered in your budgets?

**REPLY** We agree that in part Sr can be taken up by plants through leaves and cite the review by Burger and Lichtscheidl (2019) for this purpose. If the plants' radiogenic Sr composition would indeed be determined by a two-component mixing between Sr derived from rock and through atmospheric deposition, one would expect higher proportions of atmospheric derived Sr in leaves than in e.g. twig and stem samples. This would lead to gradients in  $^{87}\text{Sr}/^{86}\text{Sr}$  between the different plant compartments of a single plant. However,  $^{87}\text{Sr}/^{86}\text{Sr}$  in the different plant organs are (mostly) identical within uncertainties (see associated data set; Oeser and von Blanckenburg, 2020). Thus, we rule out direct input of atmospheric Sr into leaves. We will clarify this point. Concerning internal aeolian redistribution there are, unfortunately, precious little means to directly quantifying this. In absence of such means a common view is: if material leaves the system it would be included in the cosmogenic nuclide-derived denudation rate. If material is internally redistributed without gains or losses it would not affect the mass balance.

**COMMENT 4** The question "Are nutrient sources setting plant stoichiometry?" can be clearly answered with "no" based on well-established textbook knowledge. As an example, I cite an introductory sentence from the textbook "Regulation of Plant Nutrient Uptake" by G.N. Mitra (2015, Springer): "To cope with wide variations in mineral concentrations in soil, plants have evolved mechanisms so that net intake of a nutrient depends on the plant's need for this element rather than its concentration in the rooting medium." I could have cited any other textbook on plant nutrition. The author's use of plant nutritional terms is in large parts wrong. While Fe is an essential micronutrient (not only beneficial), Al can only be considered as beneficial for some plants at low concentrations in soil solution. For most plants it can instead be toxic. All essential elements are needed at the same time so that there is not a "most plant-essential nutrient". However, the plant requires different amounts of each

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nutrient. In plant tissue, e.g., the mean molar ratio of the K:P concentrations is 4.2 and the mass ratio is 5.3 (Marschner, 1995, Nutrition of Higher Plants, Academic Press). Nevertheless, P is more frequently growth limiting than K. In most cases, forests are limited by one nutrient or co-limited by two and sometimes even three nutrients. You cite Stock et al. (2019) who stated that your study ecosystems are N-limited, which is in line with the global mapping of nutrient limitation by Du et al. (2020, Nat. Geosci. 13, 221-225). If this is true, no other element than N will be growth limiting. Finally, nutrient concentrations in plants usually vary by less than one order of magnitude. If you now allow for a range of two orders of magnitude (as done in Fig. 6), you will for sure get an overlap between the soil and plant stoichiometry, which is, however, meaningless. Although the organic layer will more closely reflect the plant stoichiometry, its composition is still different from that of the plants, because some nutrients are to a large degree retranslocated prior to leaf/needle abscission (e.g., P) and others are quickly leached from the organic layer (e.g., K). Overall, I think that this part of the discussion must be completely revised or omitted. It would likely make sense to seek the support of a plant nutrition specialist.

**REPLY** We see the point that we may have over-interpreted Fig. 6 with respect to nutrient limitation and plant stoichiometry. As this section is not essential for our analysis and we can remove this without loss in any to the papers' conclusions. Thus, we will follow this advice and remove this part of the discussion, with the exception of the use of Fig.6 as these correlations are evidence that the plant-available pool is indeed the mineral nutrient source.

**COMMENT 5.1** I don't think that your view of "abiogenic" weathering is correct. The weathering in the deep subsoil (which you call saprolith) is strongly influenced by the acids produced by biological activity. The CO<sub>2</sub> concentration in soil air is frequently one order of magnitude higher than in the free atmosphere resulting in the formation of carbonic acid which is leached to the weathering front together with organic acids driving the chemical weathering. Moreover, it is highly unlikely that your saprolith is

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free of biological activity (i.e. plant roots, fungi, bacteria, soil animals), which would result in a direct biological acidification by the release of protons during cationic nutrient uptake, the release of organic acids, and the production of CO<sub>2</sub> by respiration. If there are roots, as suggested for La Campana, there would even be mechanical weathering by the plants.

**REPLY** We are puzzled that this comment is even made. The term “abiogenic weathering” appears only once in the manuscript (in the abstract). In the remainder of the paper we explicitly summarize the potential influence of plants on weathering (L503ff). Our list of mechanisms includes the respiratory release of CO<sub>2</sub> (point 4). We never state that the saprolite in the EarthShape sites is free of biological activity. Rather, we discuss a variety of mechanisms on how plants might accelerate weathering, including the forces introduced by plant roots and the effects of mycorrhizal fungi and soil microbiota.

However, as stated above, common weathering geochemistry is treating the weathering zone as a conceptual abiogenic endmember model, onto which biological processes are superimposed either conceptually or from observation. Deciphering whether weathering is more intense, or its fluxes are higher in the presence of plants has never been explicitly resolved. This is the objective of this study. This is a major question in weathering Geochemistry (e.g. Berner et al., 2003; Calmels et al., 2014; D’Antonio et al., 2019; Kump et al., 2000; Porder, 2019; Quirk et al., 2012).

**COMMENT 5.2** The source of acids originating from CO<sub>2</sub> dissolved in rainfall is much smaller than the biological CO<sub>2</sub> sources and it is already buffered in the vegetation canopy and the topsoil and does not reach the weathering front. I would therefore even turn around your conclusion, stating that in vegetated areas, there is likely hardly any abiogenic weathering. The latter might be restricted to not vegetated areas and mainly driven by insolation, salt and ice blasting.

**REPLY** This is pretty much EXACTLY what we say (Line 547 point 4). A view is suggested here that we never made. The title of this section is: “Is weathering modulated

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by biota?”. The question whether all weathering is “abiogenic” is an artificial one not posed in this paper. Rather, the question is whether weathering RATES depend on the production of biomass, above- and belowground. We refer to a recent hypothesis paper exposing this question explicitly (Brantley et al., 2011).

**COMMENT 6** The cation-exchange/carbonic acid proton buffer system has a small capacity and a high buffer rate. It is therefore quickly passed. More important are the carbonate/carbonic acid (pH 7-8), the Al oxide/strong acid (pH 3-5), and in the E horizon of the Podzol at Nahuelbuta the Fe oxide/strong acid (pH 2-3) buffer systems. All soils acidify in the course of their development because of a number of proton sources of which the carbonic acid formed by the much higher CO<sub>2</sub> concentration in the soil air than in the free atmosphere is the largest one. This acidification results in acid soils with a pH < 5 in the dissolution of Al oxides and the subsequent replacement of exchangeable base metals by Al<sup>3+</sup>. I don’t understand how the cation-exchange capacity can exceed the bioavailable cation pool (determined as the sum of water-extractable and NH<sub>4</sub>OAc- extractable fractions – if I understood the description of the methods correctly, where it is not described that the two fractions are summed up), because in one of the standard methods to determine the cation-exchange capacity NH<sub>4</sub>OAc is used as extractant (and anyway any salt added in excess will exchange all exchangeable cations from the soil). The apparent difference can possibly be explained by the spatial heterogeneity but not by any mechanistic background. Exchangeable cations are entirely considered plant-available. The cation-exchange process is very fast so that the composition of the ion mixture on the cation exchanger surfaces is always in equilibrium with the soil solution.

**REPLY** We not understand the point the reviewer is making. We will address this issue by removing the section on cation exchange capacity. The main point is that the bioavailable pool is smaller at the humid site. This can be easily explained by the lower pH and the higher runoff.

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**COMMENT 7** The finding that the more developed the soils are, the larger is the part of the nutrients needed to satisfy the plant demand that is directly cycled between the vegetation and the organic layers/mineral topsoils and thus decoupled from weathering is known since the early 1980s going back to the work of e.g., Jordan (1982, *Ecology* 63, 647- 654).

**REPLY** The cited paper by Jordan (1982) does not show recycling. The paper reports on an imbalance in atmospheric and dissolved export fluxes (interpreted to show net forest growth) but not nutrient recycling. We wonder how recycling rates were determined in the 1980's, as soil profile estimates of weathering fluxes (like from cosmogenic nuclides) did not exist, and high-quality data on plant stoichiometry was rare. We would be grateful for recommendations of other literature than Jordan (1982). In any case, filling these gaps by suggesting means to quantify nutrient recycling is one of the major objective of this paper.

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