We thank the Reviewer #2 for his/her constructive comments on our manuscript. As raised by the other reviewer, one of the major issue of the first version of our manuscript is the inappropriate use of the "nutrient" term for Ba. We will thus significantly rewrite some part of the manuscript to remove this confusion. Additionally, Reviewer #2 mentions some lack in the graphical report of uncertainties for some parameters we used in the manuscript. We will solve this issue by adding error bars when necessary (see the answers to the specific comments below).

The manuscript is well written and organized and a reader can follow the authors' argumentation. The English of the second version of the manuscript benefited greatly from proof reading after the access reviews. I encourage the authors to also check the supplement for English grammar and style. I recommend publication of the manuscript in *Biogeosciences* after moderate revisions. My comments, which I hope the authors will find useful and constructive, are listed below.

We are pleased that the reviewer found improvement in English grammar and style. During revision, we will further improve the English in the parts mentioned by the reviewer, and everywhere necessary.

Specific comments:

The main conclusion of the study is that a considerable amount of Ba dissolved from the bedrock and transported by the rivers is taken up and stored by biota. I was wondering how exactly plants and/or (micro)organisms utilize Ba. To my knowledge, Ba is not considered an important nutrient. The authors even state that Ba could be a limiting factor for biota growth (page 19, line 424). This has to be further discussed.

This comment has also been made by the Reviewer #1. We will remove any wrong use of the term nutrient (*i.e.* according to the definitions of Marschner (2011)). The statement made that Ba can be a limiting nutrient is inappropriate, and will be removed in the next version of the manuscript. Therefore, we will remove the term “micro nutrient” for Ba from the manuscript. To answer the reviewer's question, we note that although the biological role(s) of Ba are still far from being elucidated – apart from the apparent toxicity of high Ba amounts for plants (*e.g.* Lamb et al., 2013, and reference therein) – previous studies have shown that its chemical similarity with other rock-derived nutrients enables its uptake by plants. Indeed, Bullen and Bailey (2005) have demonstrated the significant biological uptake of Ba and attributed it to its similar ionic radius compared to other alkali-earth elements (Ca, Sr), as well as likely K. A more recent study has shown that Ba uptake scales with that of Ca even if the exact role of Ba in plants has not been identified (Myrvang et al., 2016). Finally, significant uptake and recycling of Ba by the vegetation have been shown by Bullen and Chadwick (2016) using the Ba isotope composition. Therefore, despite the poor constraints on the specific role of Ba in plants, existing knowledge argues in favor of a “nutrient-like behavior” for Ba. This is the term we plan to use in the revised version of the manuscript, and we will slightly extend the justifications in the introduction as to why Ba can be, to some extent, used as a tracer of other rock-derived nutrients. However, we also acknowledge that the behavior of Ba during uptake and recycling by plants is likely to differ to some extent from that of major rock-derived nutrients such as Ca, Mg, K or P (these different major nutrients already showing various behaviors in ecosystems). To reflect the potential decoupling between the cycling of Ba and that of other rock-derived nutrients, we will clearly state in the introduction that using Ba as a tracer of other rock-derived nutrients should be understood as a "working hypothesis" of the manuscript.

Did the authors propagate uncertainties of single parameters in their models? Most figures do not have error bars and it is thus difficult to assess whether apparent trends are real or not within uncertainty. Furthermore, I am missing estimations on the uncertainties of, for instance, the 20% underestimated CO_2 consumption, the main impact this study might have.

For some parameters such as F^{Ba}_{bio} (eq. 8) or $w^{Ba}_{isotopes}$ (eq. 12), uncertainties were already propagated (Tab. S2, Figs. 8b,c) using a Monte Carlo method. However, the reviewer is right when noting that some uncertainties were not graphically displayed in the first version of manuscript, such as w^{Ba}_{fluxes} and (Ba/Th)N (Figs 8). We will display all these uncertainties in the next version of the manuscript.

For the isotope mass balance in section 4.2 the authors assume congruent dissolution of the bedrock, i.e., no Ba isotope fractionation. Assuming this assumption is wrong, how large would be the impact of isotope fractionation during rock dissolution on the model output? Would it be negligible?

First, we would emphasize that most of the current batch and/or open flow-through model assume that the dissolution of rocks operates in a congruent manner (see Bouchez et al. 2013; Dellinger et al. 2015). This is confirmed by a wealth of experimental work, for example by Ziegler et al. 2005 (Si), or Wimpenny et al. (2010) (Mg and Li). Nonetheless, a way to take into account the uncertainty associated to potential incongruent dissolution of the bedrock is to consider the variability in Ba isotope composition in the average bulk rock undergoing weathering (this variability being due to different mineralogical compositions and different Ba isotope composition between these minerals). As our analysis already takes into account the variability of bedrock Ba isotope composition through the term $\delta^{138}Ba_{rock}$ of eq. B3 (equal to -0.02 ± 0.04 to 0.07 ± 0.02), we assume that incongruent dissolution – if any – is included within the uncertainties we present for the obtained parameters.

In section 4.4 the authors describe an apparent trend in biological Ba cycling with ecosystem dynamics (Fig. 7c,d). As the figure is now, I fail to see the trend. The only obvious is that the Madeira tributaries have lower GPP and TER values than the rest. However, there is a discrepancy between GPP data in Fig. 7a and 7c. Also, error bars are missing.

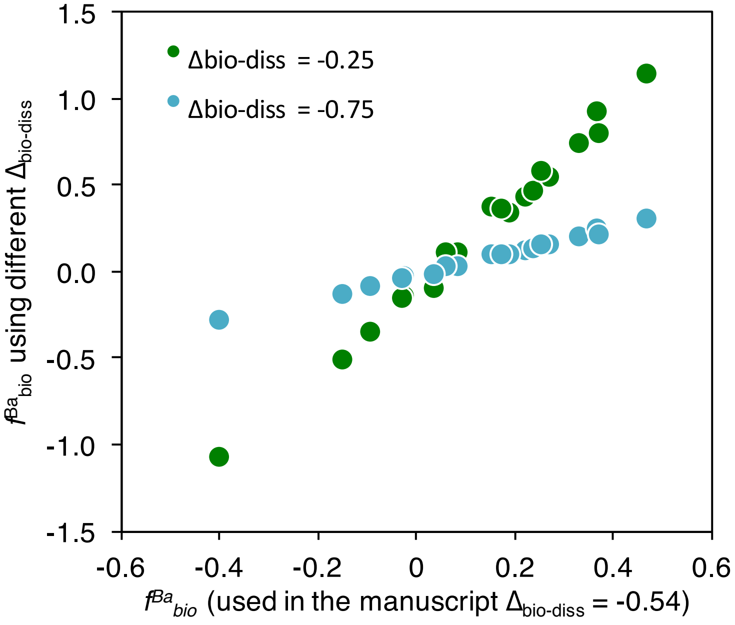
We acknowledge that the relations in Fig. 7c,d are not very clear. But given the scope of our manuscript, we expected that the readers would like to see these figures, and this, regardless whether or not GPP and TER show clear relations with F^{Ba}_{bio}. Nonetheless we note that, even if the relation is weak, we can clearly see the distinction between rivers group units *ie.* Andean tributaries, Main tributaries and “dilute” tributaries. We will rephrase the discussion around this figure in this way. We will add error bars on the model, as the reviewer suggests.

In section 4.7, I do not agree with the authors' interpretation that $R_{(sil+bio)/sil}$ increase with very low W/D, based on Fig. 12. The argumentation is apparently based on one data point. Also, this figure lacks error bars.

We will remove this argument based on one data point, and provide error bars.

Appendix B: The authors estimated the Ba isotope fractionation between dissolved Ba and Ba taken up by biota, admitting that it is poorly constrained. Yet, they state the fractionation with a fairly high precision of ±0.05 ‰. How reliable is the estimated fractionation?

We acknowledge that the uncertainties on this parameter was underestimated in our present analysis. Actually, the number of data on vegetation is very scarce but consistently show negative values (resulting in an estimate of the fractionation factor associated to biological uptake between -0.25 to -0.75). However, although its exact value is under-constrained, the fact that this parameter is negative is the main driver of our findings. To show this, we plot in the attached figure the computed f^{Ba}_{bio} of the manuscript against f^{Ba}_{bio} calculated using the two extreme values we can estimate from the literature for the isotopic fractionation for biological uptake ($\Delta_{bio\text{-}diss}$ of -0.25 to -0.75). We note that by doing so, each f^{Ba}_{bio} shows positive correlation, thus leaving the trends shown in our manuscript unchanged, and lending confidence in our interpretations. Nonetheless, during revision we will give to the $\Delta_{bio\text{-}diss}$ a more realistic uncertainty, and modify the resulting error bars on f^{Ba}_{bio} (and all derived parameters) in the relevant figures accordingly.



The authors made a great effort in computing and quantifying data and parameters. However, not all derivations of equations can be followed easily. For instance, I failed to understand how equations C5, D5 and D6 are derived given the provided information.

We will provide further details for the derivation of these equations.

Page 3, line 59: ^{130}Ba is a primordial nuclide and can be considered stable under geochemical aspects.

We will mention the fact that the very long decay of ^{130}Ba allows to consider it as a "stable" isotope.

Page 5, line 134: What are plutonic rocks in this case? What is their lithology?

We consider that these plutonic rocks are mostly granites (felsic igneous rocks; see Stallard and Edmond 1981).

Page 10, line 214: Please define * in the main text, not only in the figure caption and supplement.

We will add the definition of "*" in the main text.

Page 19, line 433: Why is the residence time of water longer along steeper slopes?

Although counter-intuitive, a longer water residence time below steeper slopes is the correct interpretation proposed Torres et al. (2015) based on the isotopes of the water molecule. Indeed, following these authors deep in soils in the plains might prevent water infiltration allowing for the rapid transfer of water to the streams during / after precipitation events. By contrast, in mountainous areas the presence of fractured and jointed (because of faulting due to tectonic activity) and presence of fractures at shallow depths below ground allows for the formation of large rock-hosted aquifers, which in turn results in longer water residence time.

Page 29, line 654: I could not find any data/figure supporting the argument that mainly K weathering flues are influenced by biological cycling. If they are to be found, e.g., in the supplement, please refer to it. Otherwise data have to be provided.

We acknowledge that the influence of biological uptake on the K cycling was somehow too implicit in the text. Indeed, the reason leading us to think that K deriving from rock weathering is strongly affected by biological cycling (Chaudhuri et al., 2007). In addition, when significant uptake by vegetation is found (as inferred by our Ba isotope mass balance), it appears that the addition of K “stored” in the vegetation represent in average around 40% of the K release from rock dissolution. We will clarify the text in that way.

Page 30, line 669: Please quantify this significant uncertainty!

We will add uncertainty on these data (and the figure as well).

Page 31, line 672: [...] to be source mainly from silicate rocks [...]

We will add "mainly" to the sentence.

Page 33, line 754: Charbonnier et al. (2018) is a review paper. When literature data are used, please cite the original publications (also later in that appendix).

We will add reference of the original publications and where it is necessary.

Technical comments:

Fig. 7: GPP data are different in panel a) and c)!

We fixed the mistake (see the comment on the lack of relation on this figure above).

Fig. S2: Are the error bars correct? They show approximately ±0.15 ‰ on $\delta^{138/134}$Ba. Long-term precision for BaBe27 and JB-2 is however given as ±0.08 ‰.

These error bar represents the uncertainties for each single measurement and not the S.D value or the confidence interval (CI95%).

Reference used:

Bouchez, J., Von Blanckenburg, F., & Schuessler, J. A. (2013). Modeling novel stable isotope ratios in the weathering zone. *American Journal of Science*, *313*(4), 267-308.

Bullen, T. D., & Bailey, S. W. (2005). Identifying calcium sources at an acid deposition-impacted spruce forest: a strontium isotope, alkaline earth element multi-tracer approach. *Biogeochemistry*, *74*(1), 63-99.

Bullen, T., & Chadwick, O. (2016). Ca, Sr and Ba stable isotopes reveal the fate of soil nutrients along a tropical climosequence in Hawaii. *Chemical Geology*, *422*, 25-45.

Chaudhuri, S., Clauer, N., & Semhi, K. (2007). Plant decay as a major control of river dissolved potassium: a first estimate. *Chemical Geology*, *243*(1-2), 178-190.

Myrvang, M. B., Hillersøy, M. H., Heim, M., Bleken, M. A., & Gjengedal, E. (2016). Uptake of macro nutrients, barium, and strontium by vegetation from mineral soils on carbonatite and pyroxenite bedrock at the Lillebukt Alkaline Complex on Stjernøy, Northern Norway. *Journal of Plant Nutrition and Soil Science*, *179*(6), 705-716.

Torres, M. A., West, A. J., & Clark, K. E. (2015). Geomorphic regime modulates hydrologic control of chemical weathering in the Andes–Amazon. *Geochimica et Cosmochimica Acta*, *166*, 105-128.