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## *Interactive comment on* "Biogeochemical response of alpine lakes to recent changes in dust deposition" by A. P. Ballantyne et al.

## Anonymous Referee #3

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## General comments:

1) The paper aims to address the biogeochemical response of alpine lakes to recent changes in dust deposition. The topic is interesting and within the scope of BG. However, the conclusions achieved are not based on a proper consideration of the processes involved, and the methods, assumptions and description of the calculations are not sufficiently complete and clearly outlined to allow a proper evaluation of the results. There are three major issues in that respect: (i) spatial and temporal variability is not properly evaluated (or sufficiently explained) in the experimental design, this compromises all quantitative issues; (ii) the discussion of nutrient limitation based on sediment C:N ratios and total P in bulk sediment are based on assumptions that are not valid, sediment C:N:P stoichiometry do not reflect the actual stoichiometric constraints

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of the algae during growth in the water column or on benthic substrates; (iii) the title and conclusions go far beyond the actual content of the study, the two tarn studied are scarcely representative of most common alpine lakes, the authors should indicate and consider that they are dealing with a very particular subset of alpine lakes, those extremely shallow and small located in non-vegetated watersheds. Bellow I develop these issues in detail and suggest possible actions, which require an in-depth revision of the manuscript.

Detailed comments:

2) Title: It goes far beyond the particular case the paper describes. In that respect it is misleading. I suggest being more specific. The paper do not discuss the general response of alpine lakes to dust deposition, it describes two very particular cases in a very particular region of the planet with a lot of circumstantial evidence. There are three aspects that should be considered in particular when planning a new title: 1) geographical location; 2) size and depth of the lakes (the case described covers an extreme case within the alpine lake variability; and 3) soil development.

3) Introduction: The authors use a wealth of citations related to stoichiometric aspects. However, their case bases on sediment organic matter. There is not a straightforward connection between the literature cited and the present study in that respect. Sediments undergo an important diagenetic process. The characteristics of this process are as relevant as the initial composition of the organic matter accumulating. The whole issue of diagenesis is omitted in this paper from introduction to discussion. I think it is a mistake. Particularly, because diagenesis imply also a feed-back to the underlying water and, more importantly in this case, with the top sediment, which in a lake <1m deep is formed by a biofilm of both algae and heterotrophic microorganisms.

4) Study sites: Site description is poor. Please provide geographical coordinates. Which is the maximum depth of the lakes? Where the coring site was located within the lake? Are there studies that have evaluated the spatial heterogeneity in sedimentation

rates in these lakes? In such a shallow lake differences in a few meters can be large depending on the initial topography of the substrate and how the basin has been filling during the ontogeny of the lake. If snowfall is high, water level oscillations must be relevant, either vertically or horizontally, how can they affect the sedimentation rate in your particular sampling point? Is there temporal or permanent outflow? Are they seepage lakes? How many inflows are they? Do water flow during thawing form channels on the sediment surface? These and other details are vital to evaluate whether changes in sedimentation rates at the coring point can be taken as representative for the whole lake.

5) Sample collection: Bedrock types? What do you mean exactly? In table 1, Sr standard deviation is similar to average, this means bedrock types can highly differ in composition also for other elements. Does this variability correspond to these different bedrock types? Was the relative coverage of each type evaluated to eventually weighing the comparison with sediments and dust accordingly?

6) Sample collection: Here you mention that outer weathering rind of all bedrock samples was removed before analysis, later you mention that there were no significant differences between weathered and non-weathered parts. Could you clarify this issue? It is quite unusual a complete congruent chemical weathering of the rocks? You should show that this is the case. If not, you should consider discussing potential effects of climate fluctuations on weathering rates and characteristics, perhaps to eventually conclude that they must be irrelevant for some of the issues addressed.

7) Sample collection: How many dust deposition events did you finally measured? How spatial heterogeneity in snow accumulation and dust concentration was evaluated? How much can be the error in the final flux estimation? Has been the interannual variability considered in other studies? If the number of dust events is low, interannual variability can be huge.

8) Dating: I do not think that sedimentation and accumulation rates based on Pb210

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and C14 can be easily compared. You basically address to different time windows, particularly with such a small number of C14 dates. Therefore, any conclusion based on this comparison is weak. I think that at most, you can use C14 dates to estimate an average sedimentation rate for the Holocene as a reference for current sedimentation rates estimated with Pb210. However, you cannot demonstrate recent changes because they do not occur within you PB210 date period. So, in fact, you don't have a proper temporal framework to address that issue.

9) Dating: I am also quite concerned that these lakes may dry out episodically. The C14 is too poor for evaluating this point during Holocene. Therefore you should be cautious about this point or provide indirect evidence that this has not been the case. However, concerning the period dated with Pb210, we would be able to evaluate this point if greater detail of the age model is provided. I think that specific plots for Pb210 dating should be included, with the measurements and the age and sedimentation model.

10) Biogeochemical analyses: First paragraph in this section should clarify whether the elemental composition correspond to the total sediment or some specific fraction. In particular, it is unclear which P fraction is considered. This is a key point when comparing with C and N.

11) Biogeochemical analyses: What does a subset of samples mean? Please indicate here or in the table how many samples were analyzed and to which levels they correspond in the case of sediments.

12) Certain harmonization in the way to indicate the products used is necessary (e.g. H2O2 and hydrogen peroxide, are both used in this section).

13) Statistical and geospatial analyses: The way of constructing the age-depth model appears quite sophisticated in this section, but looking at figure 2, we can see that it ends up in a linear interpolation between C14 dates and linear fitting through the Pb210 dates. The combination produces a shoulder just at the intersection of the two dating procedures. In my opinion this two dating systems cannot be amalgamated in a single

age-depth model. Temporal resolution is so different that they address fundamentally different time windows, and therefore variability cannot be compared, since the one with less resolution (C14) produces smoother results (higher averaging).

14) Results: In this section there is some mixing of results and discussion. I will list specific points below, but I think the manuscript requires a general re-writing clearly distinguishing between facts (results) and more or less justified speculation (discussion).

15) Changes in the flux of dust to alpine catchments: 194 line: Changes in the mass flux to sediments in the recent past: As I mentioned it is not correct to compare sedimentation rates based on Pb210 dating and C14 dating in this case. Coarse temporal resolution averages over longer periods, missing the details of short term fluctuations. From the data reported, some more correct sentence should be: "during the last 150 years sedimentation rates have been higher than the average rate throughout the last 3 millennia".

16) 200 line: Increase in density: The increase in density does not occur regularly in Porphyry as we could expect from a compression process in homogenous material. What does it happen around 2200 years BP? Is there any interpolation artifact?

17) 203-211 lines: The change in accumulation rates coincides with the change in dating method. I do not think you can talk about a change in accumulation rate with this data.

18) 212-213 lines: I agree that sediment isotopic signal is closer to dust. However, I will like to see more details on the variability between samples for each compartment. How many samples did you analyze? Have you statistically tested the differences? Can you discard differential weathering due for instance to dryer climate which could enrich 87Sr/86Sr ratio? Table 1 indicates huge differences in Sr concentration between samples, is there any isotopic pattern related to this differences? Please provide more information and discussion about this issues.

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19) Biogeochemical response to changes in dust loading: 226-241: lines. Causal links are here introduced without supporting evidence. The results section has to be more descriptive of the observed patterns and the interpretation in a dynamical context has to be placed in the discussion section. All in all, concerning nutrient limitation and stoichiometric issues, there is a very weak aspect. The C:N of organic matter in the sediments do not only depends on the conditions where this organic matter was produced but also about the decomposition and diagenetic process that has suffered. In addition, you are mixing data on C and N in organic matter with total P in the sediments (from the methods section I think I have to assume that). The P in the sediments has very different biogeochemical pathways than organic C and N, including potential dissolution, mobility and diffusion to the water column. Therefore, discussion about the limiting nutrient has to go beyond simple comparison of C, N and P ratios.

20) 242-259 lines. The main difference in organic dC13 within a lake is related to the pelagic and benthic environments. This is not considered at all here, and in a so shallow lake production on top of the same sediments should be highly relevant. The high fractionation in benthic systems is largely due to diffusive boundary layer effects. Thinner boundary layers, in more windy conditions for instance, will let to less discrimination. Relative increase in pelagic production will also produce the same result. Therefore, concluding that the changes in dC13 reflect higher productivity is highly speculative, alternative hypothesis have to be considered and discussed. In addition to this production aspects, potential effects of diagenesis should also be considered.

21) 260-274: I think that elemental composition should go in a different section than isotopic C and N issues.

22) 281-282. Please note that the Fe, Cd, P and Sr different behavior in Porphyry and Senator Beck is also reflected in the PCA. These elements change the direction of their loading in the first axes. I suggest commenting first on figure 7 and later about fig. 6.

23) 286-308. In this paragraph you use updated names for diatom taxonomy, whereas

in fig.8 you use traditional names, please harmonize. Fig. 8 reports relative abundance for a few species, whereas in the main text you talk about abundance. Did you evaluate the diatom flux? Is there an increase in total diatom production or it is just an species substitution?

24) 4.0 Discussion. 4.1 Changes in dust fluxes and their geochemical composition. 319-339. Comparison with soils is interesting. However, I wonder to what extent the estimation can be conditioned by the limited sediment sampling points (one per lake). Let's assume that the dust contribution is very high anyway in these two shallow tarns. The last statement in the paragraph is really misleading: "alpine lakes are excellent recorders of changes in dust deposition and that there is very little geochemical influence from bedrock or soils in these watersheds". These two tarns are far from being representative of all or even average alpine lakes. Therefore, I think, it we'll be more faithful to state that "small alpine lakes on non-vegetated watersheds and crystalline bedrocks can be excellent recorders of changes in dust deposition".

25) 340-345. Could you explain how do you discard potential Sr isotope fractionation due to differential rock weathering? I think that your conclusions are correct, but I will see some comment about this alternative hypothesis.

26) 347-350. The dust deposition increase is based in one point per lake evaluation. Provided the lakes are extremely shallow I wonder about the spatial heterogeneity in the deposition process and how it depends on the water budget, for instance. Will changes in snow or rainfall modify the sediment focusing patterns?

27) 4.2. Implications for biochemistry. 352-375. I think it is totally inappropriate to discuss nutrient limitation based on C:N ratio in sediment organic matter and P from bulk sediment. First, C:N depends not only on autochthonous organic matter but also allochthonous inputs, which in this case they may com from far away with dust. On the other hand, the C:N ratio is further modified in the decomposition process, increasing with time. In addition to this objections, considering stoichiometry of P based on total

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P of bulk sediments is absolutely inappropriate. We do not know how much of this P has been really bioavailable, and in addition it may suffer a complex cycling depending on the redox conditions and elemental composition. All this stoichiometric discussion should be removed from the paper. And potential dust effect discussed in a more biogeochemical process based way.

28) 363. As mentioned before, I do not think that d13C change probe that productivity increases.

29) 396-397. The latter sentence seems not to be completed.

30) 402-420. This paragraph is terribly speculative. The authors have not evaluated spatial heterogeneity in sedimentation. Sediment focusing is not the only factor, just think on how many processes can affect sediment distribution in a tarn less than 1 m: changes in water level, redistribution of sediments by water flow during melting, wind action, etc.

31) 409. Authors use throughout the term "Holocene" to refer to their long term record. However, their data only arrive to about 3000 years ago. This is less than 1/3 of the Holocene. I suggest using "the last millennia" instead.

32) 421. "Knowledge of modern fluxes, weathering fluxes..." could you provide more information on how they were evaluated? In addition to the already stated problem of evaluating and upscaling heterogeneity in snow or dust deposition within the watershed, there is the issue of interannual variability in dust fluxes? If the deposition is mainly episodic there can be huge interannual differences, please discuss this point.

33) 438-451. Concerning P budgets, what role can play P diffusion from sediment to water during low oxygen periods (winter ice and snow cover)? What is the role of water flow? Do these sediments freeze during winter? Is there any wash out of sediments during thawing?

34) 452-462. Estimating N fluxes without considering wet deposition is extremely risky,

some estimation based on potential N values for the area should be compared to the dust N flux.

35) 463-476. Both species Achnanthidium minutissimum and Staurosirella pinnata are among the most widespread species in remote cold lakes throughout the world, it is difficult to assess the underlying cause of their relative increase. If total diatom valves fluxes were evaluated some direct evidence on productivity changes would have been available. Interestingly, in this part of the discussion you consider changes in lake level, could these changes have modified the local distribution (and thus local accumulation rates) of the sediment deposition, without changes in the average atmospheric dust deposition?

36) 477-480. You mention droughts during 20th century, is there also evidence of them during the last 3 millennia?

37) 481-512. As I repeatedly mentioned I think is completely inappropriate to discuss nutrient limitation and stoichiometric issues of lake primary production based on C:N ratios from the sediments and bulk sediment total P.

38) Table 1 legend: The following sentence is ambiguous: "There was no significant difference between sediment and bedrock samples for various watersheds in the San Juan Mountains, therefore these samples have been pooled for all analyses". What did you exactly have done? According to the standard deviation of Sr in bedrock samples, differences in concentration are huge. Is there any relationship between concentration and isotopic composition? How many samples did you measured to be sure SD was stable and thus mean value reliable. Sr/Nd ratios are quite different between the three compartments (5.3 sediments, 8.3 dust, 6.2 bedrock), in this feature, bedrock and sediments were more similar. In summary, you need to explain better the number of samples considered in each comparison, why these samples are representative of the whole watershed and then test statistically that the compartments are different.

39) Table 2 and main text: Note that it is "Akaike" not "Akaiki"

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40) Figure 8. Harmonize legend labels with other figures. Update species names according text.

41) References: check typo errors and complete references.

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