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Interactive comment on “A fine fraction of soil used as an aerosol analogue during the DUNE experiment: sequential solubility in water with step-by-step decreasing pH” by C. Aghnatos et al.

Anonymous Referee #2

Received and published: 10 April 2014

Review for the paper submitted by C. Aghnatos, R. Losno and F. Dulac for Biogeosciences Discuss., 11, 2623–2637, 2014

1. Does the paper address relevant scientific questions within the scope of BG?

Yes. The paper address a scientific question related to the biogeochemical cycling of elements.

2. Does the paper present novel concepts, ideas, tools, or data?

It does provide some novel data on the associations of solubilities in different elements in a Saharan soil sample. Such data are worth publishing.

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3. Are substantial conclusions reached?

The importance of the conclusion is unclear.

4. Are the scientific methods and assumptions valid and clearly outlined?

The methods need clarification. The assumptions have to be fully supported by a proper literature review

5. Are the results sufficient to support the interpretations and conclusions?

In most cases, yet but some interpretations are inaccurate or wrong.

6. Is the description of experiments and calculations sufficiently complete and precise to allow their reproduction by fellow scientists (traceability of results)?

Clarifications or more information in some details are needed. See below and also the comment by the first reviewer.

7. Do the authors give proper credit to related work and clearly indicate their own new/original contribution?

No. A much better literature review and reference to literature is needed to better interpret the data presented.

8. Does the title clearly reflect the contents of the paper?

Yes.

9. Does the abstract provide a concise and complete summary?

Yes, but there are clearly inaccurate or wrong statements.

10. Is the overall presentation well structured and clear?

The structure is ok but the presentation needs substantial improvement.

11. Is the language fluent and precise?

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Needs improvement.

12. Are mathematical formulae, symbols, abbreviations, and units correctly defined and used?

OK

13. Should any parts of the paper (text, formulae, figures, tables) be clarified, reduced, combined, or eliminated?

No.

14. Are the number and quality of references appropriate?

No. More references are needed to interpret the data.

Overall, this short paper presents some interesting data on the associations of solubilities of different elements in a Saharan dust sample at different pHs. Such data are novel and worth publishing. However, substantial improvements are needed to meet the high quality standards of Biogeosciences. A general comment is how the outcome of this paper supports the overall aim of the DUNE project as this paper is part of DUNE special issue.

Detailed comments:

P2624, line 2: “sieved at 20um”, this is the only place where it mentioned 20um sieving. The methodology has been briefed in the Methodology section. Is it a wet sieving or dry sieving? What are the implications of wet sieving for more soluble elements such as P? This needs to be discussed in the main text.

P2624, line 6-7: This is the only place where the authors have chosen different pHs to simulate the atmospheric processing of dust. A substantial reference to literature is needed in the text to justify the chosen conditions. The statement that “the lowest of which could be reached during cloud conditions” is most likely wrong. Low pH is not commonly seen in cloud conditions and may only occur when there is serious pollution

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in the fog droplets. Such conditions have nothing to do with dust though. There are some French papers that you may refer to:

Deguillaume, L., Charbouillot, T., Joly, M., Vaitilingom, M., Parazols, M., Marinoni, A., Amato, P., Delort, A.M., Winatier, V., Flossmann, A., Chaumerliac, N., Pichon, J.M., Houdier, S., Laj, P., Sellegri, K., Colomb, A., Brigante, M., Mailhot, G., , 2014. Classification of clouds sampled at the puy de Dome (France) based on 10 yr of monitoring of their physicochemical properties. *Atmos. Chem., Phys.*, 14, 1485–1506, 2014. Deguillaume, L., Leriche, M., Desboeufs, K., Mailhot, G., George, C., Chaumerliac, N., 2005. Transition metals in atmospheric liquid phases: sources, reactivity, and sensitive parameters. *Chem. Rev.* 105(9), 3388–3431.

P2624, line 9: “strength”, it is not clear what is the meaning of this word and the sentence here?

P2624, line 11-12: “. . .with a maximum solubility at pH 3”. This statement is inaccurate and misleading. Because the authors have used a sequential extraction, it is wrong to say that the maximum solubility is at pH 3. What if the sample is left at pH 1 or 2 without the pH 3 step? It is most likely that the maximum solubility is at pH 1 or 2 rather than 3 for all of these elements if not using a sequential leaching, which most previous studies did not use. It is clear that it is appropriate to group the elements into two categories but the statement has to be precise to avoid misunderstanding. Similar comment applies to that related to Fe and Al.

P2624, line 15: “unexpected high liability”; why this is “unexpected”? This needs to be justified in the discussions in context of literature.

P2624, line 18: “finest”; There are much finer and lots of nanoparticles in the soil dust. <20um cannot be defined as finest?

P2624, line 20: “true” should be “natural”

P2624, line 22: Only three types of minerals in this sample? There must be some other

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minerals. If this statement is to stay here, it needs to be re-worded, taking account that these three mineral types are the dominant ones.

P2625, line 1: “soluble”; this needs to be defined. The following methodology suggests that this fraction is $<0.2\mu\text{m}$ so not really “soluble”. It is more appropriate to say “dissolved” here.

P2625, line 3: “Basically. . .”; the reviewer is completely confused with the statement here? What does basically mean here? Does it mean a common “practice” in the community? P2625, line 10: Processed (P, R1, R2); these notations are confusing so need to be fully detailed.

P2625, line 25: “fine fraction”: as commented above, the method of sieving and where exactly the sample was from need to be briefed here.

P2625, line 26: How the samples were deposited to the filter? Re-suspended in the air and filtered? Or just pour the soil into the filtration device?

P2626, line 5: It is crucial here to justify why the different pHs are used with support from literature.

P2626, line 8: pH 6.5; Is it the measured pH in filtered solution? Does the pH change among different leaching? It reads like that the pure water has a pH of 6.5, which is wrong. Later sections also mentioned pH of 6.7 with pH 5 leaching? Why pure water would make a lower pH than pH 5?

P2626, line 9: What acid was used? Why this particular pH is chosen? 10^{-5} M acid does not necessarily mean 10^{-5} M H^+ (pH 5).

P2626, line 21-25: Define the detection limit here? From what was described below (page 2627, line 3-5), this DL appears to be the instrument DL rather than the protocol DL. In any case, this needs to be clarified. It is suggested that a table is added here to detail the DL in terms of both ppb and the percentage to that in dust used here.

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P2627, line 18: A review paper by Formenti et al. (2011), Recent progress in understanding physical and chemical properties of African and Asian mineral dust. Atmos Phys Chem should be a good reference here as well.

P2627, line 20 and Table 1: the Ca in the dust is extremely high? Is this soil from the lake deposit? How representative are these type of dust to the western Mediterranean? For example, what are the frequencies of such dust reaching the western Mediterranean? A review paper by Formenti et al. (2011) may also provide some useful data.

P2628, line 5: It appears that the high Ca has resulted in significant buffering. This raises a question over the methodology used in this paper. Such a high buffering capacity would make the dust from this source very unlikely to be acidified to low pH such as 1 during its transport from the source to the western Mediterranean. How realistic is the leaching conditions?

Another issue here is that if the pH has been buffered to 6.7 for the so-called pH 5 solution, the applicability of the results to other dust is compromised because other dust might have such a high buffer capacity so dissolution behaviour of elements might be different.

P2628, line 10: pH of 1.1; this is not surprising due to design of the pH meter P2628, line 25: Phosphate may be taken by calcium carbonate in seawater and fresh water but here it is dust. A major problem in this manuscript is the interpretation of the P data. The close correlation of P with Ca solubility is more likely caused by the Ca present as apatite rather than P being absorbed onto calcite. Apatite is a major species of P in the Saharan dust (Nenes et al., 2011). Also, Fe oxides also have a strong tendency to absorb phosphate.

Nenes, A., Krom, M., Mihalopoulos, N., Van Cappellen, P., Shi, Z., Bougiatioti, A., Zampas, P., Herubt, B., 2011. Atmospheric acidification of mineral aerosols: A source of bioavailable phosphorus for the oceans Atmospheric Chemistry and Physics, 11,

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6265–6272, doi:10.5194/acp-11-6265-2011.

P2629, line 15: The discussion on Si was irrelevant because Si was not measured here.

P2629, line 13: Low solubility of Fe in dust at pH >3 has been reported previously such as Mackie et al. (2005) and Desboeufs et al. (2001). These references need to be taken into account for discussions.

Mackie, D. S., Boyd, P. W., Hunter, K. A., and McTainsh, G.H., 2005. Simulating the cloud processing of iron in Australian dust: pH and dust concentration. *Geophys. Res. Lett.* 32, L06809, doi:10.1029/2004GL022122.

Desboeufs, K.V., Losno, R., Colin, J.L., 2001. Factors influencing aerosol solubility during cloud processes. *Atmos. Environ.* 35, 3529-3537.

P2629, line 18: The statement that “part of Al and Fe could be released by carbonate at pH 3” needs to be justified. No evidence or literature has been provided to judge the credibility of the statement.

P2629, line 10: “but excluding oxides. . .”; what are the mineralogy of Fe oxides in this particular sample? What size of these Fe oxides? What crystallinity of these Fe oxides? All these parameters could affect whether Fe oxides will dissolve at low pH (Cornell and Schwertmann, 2003). Unless such parameters are understood, it may not be appropriate to be conclusive that “no Fe will be dissolved from oxides”.

Cornell, R. M., Schwertmann, U., 2003. *The Iron Oxides: Structure, Properties, Reactions, Occurrence and Uses*, Wiley-VCH Publishers, New York.

P2630, line 8: How the total P was measured? The P solubility was calculated but Table 1 did not show P so it has not been measured by XRF.

P2630, line 18: This sentence is difficult to understand. Re-word.

P2630, line 19-21: This sentence cannot be part of a conclusion. It is a citation of

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previous results.

P2631, line 8-9: “carbonate as a source of bioavailable nutrients”; What type of nutrients in this case? Is it P?

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