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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Phosphorus solubility in aerosol particles related to particle sources and atmospheric acidification in Asian continental outflow" *by* Jinhui Shi et al.

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Responses to Reviewer 1# Comments

Reviewer #1 (Comments to Author):

General Comments: I applaud the aim of this manuscript and I feel once it is modified slightly, that it will make an important contribution to the literature. I should say that a couple of years ago, we tried to do exactly the same data treatment using a data set collected in Crete. We had in total ∼100 data points and we were unable to find significant patterns. This manuscript has 170 data points and has managed (just) to see

some real patterns albeit the correlations they find are often statistically significant but with correlation coefficients of ∼0.3!. In other general words, while the conclusions are interesting, they are not actually very strong. In particular the authors seem to divide the particles into anthropogenic or dust only. They do not include the importance of acid processing of inorganic particles (dust or anthropogenic) as wet aerosols associated with clouds (high relative humidity) as a potentially important process. In fact they do discuss this in the text and state on page 10 that "Unfortunately , we were unable to quantitively distinguish the contributions of aerosol source and acidification to phosphorus solubility at this stage." Yet the text elsewhere minimises the possible contribution of acidification and emphasises instead anthropogenic particles which had high P solubility at source. This reviewer feels the manuscript would benefit by taking a more even balance between these two possibilities. As a final general point, we have just published a paper in Global Biogeochemical Cycles (Herbert et al., 2018), which the authors obviously could not have seen. However it does predict that acid processes in China could be an important source of. Bioavailable P as a plume which passes over location such as Qingdoa and on to the western Pacific.

Herbert, Herbert R. J., Krom, M.D., Carslaw, K.S., Stockdale, A., Mortimer, R.J.G., Benning, L.G., Pringle, K., Browse, J., (2018) Quantifying the effect of atmospheric acid processing on the global deposition of bioavailable phosphorus from dust. Global Biogeochemical Cycles. (5.79) https://doi.org/10.1029/2018GB005880

Response: We thank the reviewer for the careful evaluation and helpful comments to improve the quality of the manuscript. We revised the manuscript and addressed the reviewer's comments, e.g., we make clearer the idea that atmospheric acidic processes associated with anthropogenic pollutants may transform unreactive P to bioavailable P, we cite Herbert et al., (2018) to support the idea, and etc.. In the special comments, there are three questions concerning these aspects. Please see the detailed responses to these comments below.

Specific comments: Line 16 of Abstract and elsewhere: The convention for what is

called in this manuscript DP, is actually TDP (Total Dissolved P). That is the P measured after persulphate oxidation in solution. Please change to TDP throughout.

Response: Revised as suggested.

Line 24: The authors suggest that humidity plays an important role in converting refractory P to bioavailable P. The most likely mechanism is that suggested in Nenes et al., (2010) which is the acidification of particles as they cycle from clouds, where the pH is rather high, to wet aerosols (where the pH is very low) and back again (see Stockdale et al., (2016).

Response: We added "This was likely caused by the acidification of particles as they cycled from cloud droplets to wet aerosols and back (Nenes et al., 2010; Stockdale et al., 2016)." in the Section 3.3.3 (Page 11, Lines 19-20).

Introduction Line 5 Add in offshore areas and regions where P limits. . ... General: Even in systems where N is the immediate limiting nutrient P can increase phytoplankton growth by moving the entire system to higher productvitiy.

Response: Revised as suggested.

Introduction page 2 line 9: The authors should comment/introduce the idea that anthropogenic processes can include the production of atmospheric acids, which can cause previously unreactive p to become bioavailable DIP. They discuss this possibility at length towards the end of their manuscript.

Response: In the revision, we added "In addition, atmospheric acidic processes associated with anthropogenic pollutants may transform unreactive P to bioavailable P. Recent model studies predict that acid dissolution process increases the fraction of bioavailable P from $\sim$10% globally at labile pools to 42% in the Pacific Ocean, with the mean value of 22% in global marine atmosphere (Herbert et al., 2018).", and we cite Herbert et al., (2018) in the Introduction (Page 3, Lines 8-11).

Methods page 4 line 7 Remove 'in number of particles' and Replace monitored with

measured.

Response: Revised as suggested.

Page 5 line 19: What was the assumed value of Al in mineral dust that allowed the authors to assume that the particles were 8% by mass? I may have misunderstood what was written, in which case the authors should make it clearer.

Response: The description was revised to "The particle mass loading was estimated from Al contents by assuming that all aerosol Al was derived from mineral dust, which comprises 8 % mineral aerosol mass (Taylor, 1964)." (Page 5, Lines 23-25).

Page 5 line 25 What is 'floating' dust? A dust storm?

Response: Floating dust is a kind of dusty weather but is not in the stage of dust storms. It usually occurs after the passage of dust storms when there are still a considerable dust particles (floating) in the air. It is not a dust storm. We replaced "floating dust" with "dusty weather" in the revision.

Page 6 line 11 (and various other places including table 1) Aqaba is spelt wrongly. It is a b and not a d

Response: The typo was corrected.

Page 6 lines 31-34: If TP had high correlations with major elements (dust) and with heavy metals (anthropogenic) at the same time, is that not ambiguous?

Response: There are overlaps of mineral elements and heavy metals between natural dust (from desert) and anthropogenic particles (mainly from coal burning)

Page 7 line 3: The actual correlation data is not given (or at least not given here). This reviewer is a little confused as to what the authors mean by 'higher correlations' and whether that also means lower p values.

Response: The values were given in the supplementary materials. Please see Table

S1 in the Supplement.

Yes, “higher correlation” means having a larger correlation coefficient (r value) and a smaller p value. To avoid confusion, we modified the descriptions. Please see Lines 1-6 on Page 7.

Page 7 line 13 Are the authors convinced that soil dust (from deserts?) are an important source of DOP?

Response: Yes. Our results indicate that soil dust is one of substantial sources of DOP, in comparison with that in anthropogenic particles, and this is also partly the reason for the correlation between TDP and mineral elements. In addition, similar result was also reported by Myriokefalitakis et al. (2016), who estimated that the contribution of soil dust source to DOP was approximately 25 % on the global scale.

Page 7 Line 25 The authors might consider quoting Carbo et al., (2005) which presents the P solubility data for the Eastern Mediterranean in a more comprehensive manner than Herut et al. (2002). Carbo, P., Krom, M.D., Homoky, W.B., Benning, L.G., Herut, B., 2005. Impact of atmospheric deposition on N and P geochemistry in the southeastern Levantine basin. Deep-Sea Research II Volume 52: Nos 22-23, 3041-3053.

Response: We add Carbo et al (2005) in the revision.

Page 8 line 24: The data in that graph is non-linear

Response: Yes, they are non-linear, likely because of multiple reasons or mechanisms. If the data number is adequate to categorize them into different groups for a statistically meaning investigation, more detailed mechanisms could be addressed. Here, we can only show the trend, and the data show a statistically rough linear correlation ($y=0.013x+75.8$, $R2=0.54$, $p<0.01$).

Page 8 line 27 And because anthropogenic P is more likely to have interacted with pollutant gases to produce more bioavailable P

Response: We added this information “The reason is that anthropogenic P tends to associate loosely with particulates, dissolve more readily than mineral P, and, consequently, interact easily with acid gases to produce more bioavailable P (Herut et al., 2002; Baker et al., 2006a; 2006b; Anderson et al., 2010; Hsu et al., 2014; Herbert et al., 2018).” in the revised version. Please see Page 8, Lines 27-30.

Page 9 line 4 Remove obviously

Response: Removed in the revision.

Page 9 line 14 I had the same problem with Sholkovitz’s paper too. It ignores the possibility that anthropogenic acids can interact with mineral dust to produce bioavailable P (or Fe). The authors of this article suggest this might be an important process themselves in line 31 “which more efficiently serves as a sink . . ..derived elements.” And later on page 10 “Unfortunately we were unable to quantitatively distinguish the contributions of aerosol source and acidification to phosphorus solubility at this stage”. That means both should be retained as possible sources. In reality the answer is probably that both more soluble P in anthropogenic particles at source and more P made soluble by acid processes in air masses from polluted sources occur and are in different proportions in different air masses.

Response: We totally agree with the reviewer. We discussed the contribution of aerosol sources to and the effects of atmospheric acidification process on P solubility, and also pointed out that both of them were two important factors/processes influencing the P solubility in the mentioned paragraph and pervious descriptions in the manuscript. We considered additional descriptions but feel such an addition would make the manuscript tedious. In order to avoid misunderstanding, we removed the sentence “Unfortunately we were unable to quantitatively distinguish the contributions of aerosol source and acidification to phosphorus solubility at this stage.” in the revision.

Page 11 line 29 There seems to be a mistake in the first half of the line. I read it several times and could not decide what was meant.

Response: We revised the sentence as “On average, the P solubility was approximately 13 % in the aerosols of Panth/TP < 50 %, while the value was approximately 21 % in the aerosols of Panth/TP > 50 %.” (Page 11, Lines 26-28).

Page 11 line 34 How did the authors define ‘acidification degree of 150 nmol nmol-1? nmoles of what?

Response: We define the acidification degree with the molar ratio as [2nss-SO42-+NO3-]/TP. We add the information in the revision (Page 10, Line 22).

Page 12 line 3 Remove obviously

Response: Removed in the revision.

Page 12 Conclusions Very well written and this reviewer entirely agrees with the conclusions.

Thank you very much for your careful reading and helpful comments.

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2018-892/acp-2018-892-AC1-supplement.pdf

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