

## ***Interactive comment on “Modeling the biogeochemical impact of atmospheric phosphate deposition from desert dust and combustion sources to the Mediterranean Sea” by Camille Richon et al.***

### **Anonymous Referee #2**

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Anonymous Review for “Modeling the biogeochemical impact of atmospheric phosphate deposition from desert dust and combustion sources to the Mediterranean Sea”

#### General comments

In this work, the authors assess how modeled phosphate deposition output from dust and combustion aerosols can affect the phosphate fluxes into the surface waters of the Mediterranean Sea. The oligotrophic Mediterranean is phosphorus stressed, limited, or co-limited in certain regions/species, and atmospheric deposition may be an important

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source of this nutrient. Given high anthropogenic impact on aerosols in this region, and potential future enhancements in surface water stratification, this is a topic worthy of study.

The methodology in this paper was good in most cases, and some of the important uncertainties were discussed very thoroughly. I have pointed out in the specific comments several places where the manuscript requires further explanation of the methodology. My main issue is that, in my opinion, the importance of this study was overstated, and that a few key uncertainties in the findings were downplayed too much (e.g., nutrient co-limitation, the influence of soluble organic P in deposition, non-Redfieldian marine biogeochemical dynamics, and some important model uncertainties).

Because of this latter concern, I suggest the authors proceed in one of two ways: 1) Scale back the conclusions substantially, to focus on the differences between model-estimated P<sub>comb</sub> and P<sub>dust</sub> deposition and their potential implications in a (more clearly-emphasized) highly-simplified Redfieldian ocean, or 2) Maintain the scope that the authors do now, but also present results from non-Redfield experiments with prognostic biogeochemistry (this would probably be a lot more useful for the community than option 1, but would of course be more work).

#### Specific comments

In some cases, the manuscript methodology could benefit from further explanation. For example: I was very confused about how PO<sub>4</sub> was handled in the model. On P.4 l. 108 it is stated that, “The model is run in off-line mode like in the studies performed by Palmiéri et al. (2015), Guyennon et al. (2015), Ayache et al. (2015, 2016a, b) and Richon et al. (2017). PISCES passive biogeochemical tracers are transported using an advection–diffusion scheme. . .” What was meant by the model being run offline? Of the references above, only Guyennon et al. and Richon et al. looked at biogeochemical processes – the others looked at processes involving actual passive tracers that do not behave like nutrients in the real ocean. In Guyennon et

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al., they said, “the coupling between the hydrodynamic and biogeochemical models is offline, i.e., biological retroaction on the physics is not taken into account” – but it appeared to me that biogeochemistry was prognostically calculated in that reference but not in this paper. Even if passive nutrient tracers follow deep-sea observations very well based on an offline model, how can one assess the biogeochemical changes caused by P deposition at the surface as the authors do here, if biogeochemistry is not calculated prognostically? Please clarify. In the Richon et al., 2017 text, this uncertainty was not discussed. Also, if P is a passive tracer, how can it affect Chl a as discussed in section 3.4? Please clarify this point in the text as well, and address any associated uncertainty and implications of the method in the text.

“On a related note, how exactly was surface PO<sub>4</sub> related to Chl a in the model? I did not see this discussed, or any of the associated uncertainties.

Section 3.3 and figure 5: Where does the referred-to surface PO<sub>4</sub> data come from? From the model or from observations?

Section 3.1: How was P deposition estimated from aerosol concentration observations? Was a deposition velocity assumed, and if so, what assumptions were used?

The usage of the terms “total P” and “total phosphorus” in the manuscript are confusing. In most of the literature on atmospheric P deposition, the term total P indicates the sum of all phosphorus in any form (soluble or insoluble, organic or inorganic). On p. 6 l. 172, the authors state, “We investigate the impacts of each source of PO<sub>4</sub> by performing two different simulations: “PDUST” and “PCOMB”; they include, respectively, natural dust only and combustion-generated aerosol only as atmospheric sources of PO<sub>4</sub>. We also performed a “Total P” simulation with the two sources included.” Although it is not completely clear, here the authors seem to me to imply that total phosphorus is actually the sum of phosphate only from dust and combustion sources. On p4 l. 117, the term “total phosphorus” seems to imply the same thing. Then on page 6 line 187, the authors state, “We used the times series of total P measured at 9 different

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stations over the Mediterranean from the ADIOS campaign (Guieu et al., 2010) and the soluble P measured at 2 stations in the South of France from the MOOSE campaign (de Fommervault et al., 2015)”. Here the authors seem to distinguish between soluble and total P, as I would have otherwise expected. Elsewhere in the manuscript, the authors also use the term “atmospheric P” (which to me implies total phosphorus) to mean atmospheric soluble PO<sub>4</sub>. I suggest clarifying these different concepts, and using separate terms for each. Along those lines, I also suggest changing the title in Fig. 6 from “Total P” to something else.

“On a similar vein, P1 l.15: “We examine separately the different soluble phosphorus (PO<sub>4</sub>) sources...” Please keep in mind again that soluble phosphorus and PO<sub>4</sub> are different things. Soluble P includes soluble organic P, which was not discussed much in this manuscript, except as a small note late in the paper in section 4. To avoid confusion, I recommend being clearer about this in the text.

The authors talk about other sources of surface PO<sub>4</sub> (e.g., riverine and oceanic via Gibraltar). Were these data obtained only from the model? Is there literature data with relevant information? If so, that information would be good to put in Table 2 for reference and discussion in section 3.2. If these data are not available, that would be worth mentioning and discussing.

My main concern, as mentioned, was that a few key uncertainties were either not made clear enough or fully addressed. These include:

1) Non-Redfieldian marine biogeochemical dynamics. The authors state on P. 4 l. 102 that: “PISCES is a Redfieldian model: the C/N/P ratio used for biology growth is fixed to 122/16/1.” Many recent studies have discussed the shortcomings of this assumption in the real ocean, particularly in oligotrophic regions like the Mediterranean. A very large body of work shows that Redfield dynamics may be particularly erroneous with respect to P cycling (e.g., work by M. Lomas, R. Letscher, A. Landolfi, etc. (this is not a comprehensive list)). Given that Redfieldian assumptions are unlikely to represent

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actual biogeochemical dynamics in this paper's study region, I feel that the authors must spend much more time discussing this uncertainty. It would be good if they could also more clearly state what meaningful information the results provide, given this large uncertainty. Ideally, they would also run additional model tests under non-Redfieldian assumptions.

2) The influence of soluble organic P in deposition was only touched upon in the manuscript. However, various studies suggest that it could be an important, or even dominant, source of soluble phosphorus to organisms in addition to the PO<sub>4</sub> covered in this study (e.g., Chen et al., 1985; Kanakidou et al., 2012 and references therein). Particularly relevant for this paper is the fact that soluble organic P, in the few cases where it has been measured, appears to be much larger in combustion-sourced aerosols than in dust aerosols (e.g., Longo et al., 2014; Zamora et al. 2013). The authors should discuss the implications of/uncertainties related to not including organic P in their analysis. To make the paper more useful to the community, they may also consider running sensitivity tests estimating the potential impact on their results of including this additional P source.

3) Uncertainties with the model assumptions themselves require further discussion. For example: "The majority of the results focus and rely on modeled ocean surface PO<sub>4</sub> concentrations. However, the majority of the model evaluation focuses on sub-surface ocean PO<sub>4</sub> trends, or surface Chl a trends. There was no in-depth discussion of how well the model compared to surface PO<sub>4</sub> data, or what kind of data were available for this comparison. Moreover, the authors do not discuss how surface Chl a is related to surface PO<sub>4</sub>, either as parameterized in the model, or in actual observations. Relatedly, on P11, l.342 the authors state: "Based on our large scale LMDz-INCA model, we estimate that combustion is responsible for 7 % on average of total PO<sub>4</sub> supply. In comparison, the average contribution of P<sub>dust</sub> to PO<sub>4</sub> supply is 4 % (Table 2)." These are very precise numbers that imply high confidence. What is the certainty in the other P sources? Please rephrase, or discuss further.

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4) Potential effects of nutrient co-limitation on the results. Most of the studies that I know of (although I am not an expert), indicate that phosphorus may be co-limiting along with other nutrient sources. This may also be worth discussing further.

I also had a variety of other, more minor suggestions/concerns:

P2l.27: "The most important aerosol deposition fluxes to the global ocean are induced by sea salt and natural desert dust (Goudie, 2006; Albani et al., 2015) respectively corresponding to material recycling and external inputs." Did the authors mean "most important" here (which is dependent on the process of interest) or something like, "largest by mass"? Please rephrase.

p.2 l. "It is especially important to constrain external sources of phosphorus because it limits productivity in many regions of the oceans." Reference?

p. 2 "The main sources of atmospheric phosphorus for the surface waters of the global ocean are desert dust, sea spray and combustion from anthropogenic activities (Graham and Duce, 1979; Mahowald et al., 2008)." I don't think sea spray should be considered a source, because as the authors stated, it is recycled material.

P2l54: "The Mediterranean Sea is also a hot-spot for climate change impacts (Lejeusne et al., 2010), in part because it is the recipient of aerosols from a variety of different geographical sources." I don't see how being the recipient of aerosols from a variety of geographical sources makes the Mediterranean Sea a hotspot for climate change impacts (was that referenced in the Lejeusne article somewhere)? Suggest rewording.

P.4 l. 95: "These evaluations showed satisfying results." Please be more specific?

p. 4, l. 111: "Biogeochemical characteristics of the latest version of the NEMOMED12/PISCES model are evaluated in Richon et al. (2017)." Am I correct in understanding that the Richon et al., 2017 model setup is very similar and relevant to this work? If so, I recommend that the authors just cite this paper and summarize

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the relevant information on how well the model performs from Appendix A in the text, instead of including Appendix A which just repeats the information in Richon et al., 2017 as far as I can tell. Figures A1 and A2 are already in Richon et al., 2017 almost exactly, so those can also be removed.

Figure A2 (if you decide to keep it): Currently, it's hard to understand this figure since it is not clear what is east and west (although east and west are discussed in the referring text), and since the surface observations are unidentifiable in its present form. Please note the latitudes/longitudes of the points somehow (e.g., by having an insert with a cruise track). Please also label the x-axis (is this distance in m)? Please present the data in a different way so that the reader can see the nutricline information better (e.g., with an insert, following Richon et al., 2017, or by presenting the data on depth/nutrient plots). Again, I suggest just removing this figure entirely and referencing Richon et al., 2017.

P5, l.151: "Another important source of P aerosols in this region is sea spray" I recommend removing the word "source" and with something like "input" since recycled aerosols are not really a new source of P.

P7, l.213: "The underestimation of total P deposition is also likely due in part to our omission of P from other potential sources such as PBAP and sea salt." Estimating deposition velocities from aerosols accurately is a major challenge (e.g., Jickells et al., 2017; Baker et al., 2017; Duce et al., 1991) and it is associated with high uncertainties in deposition fluxed to the ocean surface. I think this would be worth mentioning and keeping in mind as another major uncertainty for this comparison.

Figure 2 caption: please note somewhere that this is model output.

Table 2: Please mention in the Table or the caption that these estimates are model-derived. Also, as mentioned, the caption "Total P" is confusing –please clarify what you mean here – I think this value include riverine P? If so, please title this with something else distinguishable from total P in aerosols, and total sources of soluble PO<sub>4</sub>. Does

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the Krom et al estimate include rivers? Please specify.

Fig. 4: Please define in the caption what the red and black bars indicate (which is where my eye goes first to find this information). Also, it would be useful to have the same numbers in the different regions that correspond to their label in Figure 2. Also, please clarify the units of the bar plots.

P.8, l. 244: "Our previous study showed that June is the period of most significant impacts from aerosol deposition in spite of the low fluxes, due to thermal stratification (Richon et al., 2017). " Please be more specific here - most significant impacts on what?

p.8, l. 248: "The North Adriatic is under strong influence of riverine inputs and atmospheric deposition of P from combustion (Figure 3)". Did you mean Fig.4?

Section 3.2: it might be useful (although not strictly necessary for me to recommend for publication) to know how your model dust observations compare with AOD trends in the region, which are available during your study period.

P9, l. 273: "Atmospheric phosphorus deposition has different impacts on PO<sub>4</sub> concentration depending on the source, the location, and the period of the year." Suggest changing to, "Atmospheric phosphorus deposition has different impacts in the model on PO<sub>4</sub> concentration depending on the source, the location, and the period of the year."

Section 3.3 and figure 5: Please define "maximal relative effects" and "relative impacts" and what a percent of average maximal relative effect means and how it is calculated. Where do you get the surface PO<sub>4</sub> data? From the model or from observations? If in the model, how well does the model reproduce observations?

P9 l. 278: "Figure 5 shows the relative impacts of phosphorus deposition from the two sources (combustion and dust) on surface PO<sub>4</sub> concentration for the month of June. The relative impacts of atmospheric deposition from different sources are varying over

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time. . .” Please specify why you focus on June. You do not show or discuss how the relative impacts vary over time – please do so if you wish to keep this sentence.

Fig. 6: again, what does Total P represent in this instance?  $P_{dust} + P_{comb}$ ? Also, in the discussion of this figure, I think it is important to be much more focused on the uncertainties in your findings – e.g., regarding the relationship between modeled  $PO_4$  and Chl a, Redfieldian assumptions, etc.

P10 l.330: “We performed a Student’s t-test on the grid matrix of relative impacts of  $P_{dust}$  and  $P_{comb}$  over the three regions . . . and found that the mean values are statistically different ( $p$ -value < 0.01). This shows that even though the impacts of  $P_{dust}$  are close to the effects of  $P_{comb}$  in the South Ionian, they are significantly dominant. ” What do you mean by “dominant” specifically? Larger? Just because differences are significant, does not mean that the differences are meaningful. Please clarify (or remove the sentence, since it does not appear to be central to the paper).

P13, l.: “In the coastal Adriatic and Aegean Seas that are under strong influence of anthropogenic emissions, we showed that combustion-derived phosphorus deposition has effects on the biological productivity.” Suggest rephrasing to: “In the coastal Adriatic and Aegean Seas that are under strong influence of anthropogenic emissions, we showed that combustion-derived phosphorus deposition may have effects on the biological productivity” or something similar. I also suggest emphasizing that your idealized experiment results indicate that these effects are likely to be fairly small, although other experiments with more realistic biogeochemistry are necessary to further constrain this problem.

Technical comments

P11, l. 372: “tshe” to “the”

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