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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.

Camille Richon et al.

camille.richon@lsce.ipsl.fr

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We thank the editor and reviewers for their valuable review of our manuscript "Modeling the biogeochemical impact of atmospheric phosphate deposition from desert dust and combustion sources to the Mediterranean Sea". Below are our detailed responses to their questions and comments. The reviewer's comments are in *italic*, the author's answers in plain text, quotes from the manuscript are in quotation marks. We provide with this document a revised version of the manuscript and a track version allowing to visualizing changes from the submitted manuscript.

Jean-Claude Dutay published on July 25 on behalf of the authors a first reply to the reviewer 1: Referee1 is right to consider that having another set of forcings with higher resolution would represent an undeniable improvement for our modeling efforts. Unfortunately this simulation is not conceivable at short term, because such forcings are not available yet. For instance, the ALADIN-Climat model used in our previous study, presently does not simulate Phosphate from combustion (it has only Pdust at the moment), and this product will not be available soon since this development requires time. In order to make progress anyway on our scientific research, we were forced, for a preliminary study to use low resolution forcings, a classical strategy. We comment the limits of this approach in the manuscript and encourage for revisiting our conclusions with more refined forcings in the future. However we consider that our study has revealed some new interesting results, such as the spatial difference for the impact of Pcomb and Pdust and their relative quantification, that represent new information that deserve to be presented to our scientific community, as they have an importance for the modeling and the functioning of the biogeochemical cycle of the Mediterranean sea. We hope as well that this preliminary study will motivate atmospheric regional modeling group for producing more appropriate high resolution aerosols deposition field soon.

The present manuscript "Modeling the biogeochemical impact of atmospheric phosphate deposition from desert dust and combustion sources to the Mediterranean Sea" proposes an analysis of the impact of phosphorus atmospheric deposition comparing different sources: namely desert dust (Pdust) and combustion sources (Pcomb). The idea is very interesting and useful because the two sources are, in principle, characterized by different spatial and temporal distributions. But, the main weakness of the manuscript is, in my opinion, the insufficient skill of the global atmospheric model LMDz-INCA in reproducing correctly the amount of dust deposition fluxes and its spatial and temporal variability for the Mediterranean area. Authors cite

another model, the higher resolution ALADIN-Climat (Nabat et al. 2012), used for a companion paper (Richon et al prog ocean. 2017), which gives higher deposition rate. I think that it is necessary to add also a test with the ALADIN-Climat model (equipped with the proper phosphorus deposition model), in order to have at least an ensemble composed by two members, this would make results more robust. Moreover, the choice of selecting only the year 2005 given the high variability of Pdust, is not clear to me. This high variability is important and its impact should be estimated. Therefore, I suggest that the present manuscript can be published only after major revisions of the simulation protocol.

We thank the reviewer for these comments. As stated in the first reply by Jean-Claude Dutay (see also above), the use of the global model LMDz-INCA was driven by the availability of the model outputs. Indeed, we were not able to find any regional atmospheric model that treats the phosphorus cycle and distinguishes between phosphorus in dust and phosphorus from combustion. When we initiated this study, the only year for which P deposition from combustion had been computed was 2005, this guided our choice for simulating 2005 with NEMO/PISCES. The source of phosphorus from combustion has not yet been included in the ALADIN-Climat model and such development is not expected for a while. Considering these difficulties, we try to provide the best evaluation of deposition fluxes keeping in mind that both models and measurements have some uncertainties and that these induce uncertainties in our results.
We agree with the reviewer that it is difficult to conclude on the exact impact of atmospheric deposition on the biogeochemistry of the Mediterranean with such discrepancy between models and measurements. This is why we will in general try to emphasize more in the manuscript that the scope of this study is not to use models as predictors of the Mediterranean's functioning, but to test the sensitivity of an oligotrophic area such as the Mediterranean to contrasted atmospheric deposition patterns representative of some of the varied aerosol sources surrounding the Mediterranean. We hope that

these first results, along with their limitations, should encourage for more model and measurements development in the coming years.
In this paper, we point out the zonal distribution of atmospheric deposition from contrasted sources and the changes in the biogeochemistry of the different basins they impact. Therefore, even if the flux values are uncertain, there is a clear distinct distribution of phosphate deposition from the 2 considered sources. In this paper, we show that P deposition in the South of the basin (that is likely to come mainly from natural dust) has different impacts than P deposition in the North (that is likely to come from combustion sources).
We added to the discussion section some precisions about the scope and limitations of our study (lines 379-387) "The purpose of this study is to raise questions on the relative importance of the various aerosol sources that border the Mediterranean and their potential impacts on the nutrient supply and biological productivity of the basin. The literature on the Mediterranean aerosols is often centered on Saharan dust deposition which is believed to have the highest impact on the basin's biogeochemistry (e.g. Bergametti et al., 1992; Migon and Sandroni, 1999; Aghnatios et al., 2014). The study aims at shading new light on the other sources and their potential role, but, if Saharan dust does have an impact on the regional climate system and represents a source of particles (e.g. D'Almedia, 1986; Nabat et al., 2012), it may not be so dominant as previously believed as a source of bioavailable nutrients." And lines 402-406: "However, the underestimation of deposition fluxes shown by Figure 1 forces to consider that our results on the relative contributions of the different phosphate external sources are somewhat uncertain. More measurements and developments of the atmospheric modeling must be undertaken in order to make more precise assessments of the importance of atmospheric deposition as a source of nutrients to the oligotrophic Mediterranean."

ABSTRACT lines 18-20: "The impact of the different sources of phosphate on the biogeochemical cycles is remarkably different and should be accounted for in

modeling studies." This sentence is, in my opinion, not clear, "remarkably different" with respect to what? We agree with the reviewer and modified the sentence accordingly, to: "Differences in the geographical deposition patterns between phosphate from dust and the one from combustion will cause contrasted and significant changes in the biogeochemistry of the basin. These different sources should therefore be accounted for in modeling studies."

Pg 4. line113:line 115 : "The model satisfyingly reproduces the vertical distribution of nutrients in the basin and the main productive zones that are the Alboran Sea, the Gulf of Lions and most coastal areas (see appendix)." The comparison/validation shown in the appendix appears quite subjective, no objective statistical indicators are provided. It is complicated to produce reliable statistics over model simulations and difficult to find available data covering our entire simulation period/area. Therefore, with such sparse data, most of the validation in many modeling studies is only qualitative.
We calculated the average and standard deviation of chlorophyll values measured and modeled at the DYFAMED station (Ligurian Sea) for the 1997-2005 period (see Richon et al 2017, Prog. Ocean.). We found that the average measured chlorophyll a in the top 200 meters is 0.290 $\pm$ 0.177 10-3 g m-3 and the average model value is 0.205 $\pm$ 0.111 10-3 g m-3. For PO4, the average measured value is 0.234 $\pm$ 0.085 mmol m-3 and the modeled average is 0.167 $\pm$ 0.179 mmol m-3. This has been added in the manuscript lines 129-133. It should be noted that as suggested by Reviewer 2, we withdrew the appendix because the model evaluation is already shown in Richon et al. (2017, Prog. Ocean.) and shows the same figures and results.

Pg6, line 198: Pg 7, line 200: "We were able to compare the dust deposition flux modeled with LMDz–INCA used to derive Pdust deposition over the ADIOS sampling period with the measurements. The comparison is shown in Table 1. The dust fluxes produced by the model are realistic." I plotted results reported in Tab 1. See figure attached (x-axis stations, y-axis dust deposition, units are g m-2yr-1). In my

opinion dust fluxes produced by the model (brown line; MODEL "ADIOS period") compared to data (blue line; DATA "ADIOS period") are very different. In particular there is a strong underestimation of the model, about an order of magnitude, and the spatial variability across stations is absent in the model. So the sentence "The dust fluxes produced by the model are realistic", should be substituted by something like "model presents a strong underestimation compared to data and it is not able to represent the spatial variability of the data". Clearly, as stated by Authors, the dataset available is not enough, and continuous time series at different stations should be used to corroborate the model. But, given this situation, the usage of another atmospheric model, for example the ALADIN-Climat is, in my opinion, mandatory. A higher resolution model would allow for more robust results in terms of spatial gradients of dust deposition also.
In Figure 1 of the article, we provide the comparison of total phosphorus deposition (from dust and from combustion) from LMDz-INCA with total phosphorus deposition measured at different stations. Although we stress that model and observation data are not from the same year, this Figure helps evaluate the modeled fluxes against the rare measurements we found in order to point out the uncertainties of the model. In the Table 1, the dust deposition fluxes for the period 2001-2002 corresponding to the ADIOS campaign are based on model outputs with a lower resolution (1.27° in latitude by 2.5° in longitude) than those for the year 2005 (0.94° in latitude by 1.28° in longitude). As stated by Bouet et al. (2012), dust emission (and hence its deposition) is highly sensitive to model resolution. Therefore, the coarse resolution of the dust model used in table 1 for 2001-2002 may explain the underestimation and the lack of spatial variability from the model. We also noted that comparison appears better (within a factor of 2) at the 4 stations of the eastern Mediterranean (Cyprus, Greek Islands and Turkey). We added this precision in Section 3.1 (lines 234-243).
Comparison of LMDz-INCA phosphorus deposition fluxes to measurements on the global scale are provided by Wang et al (2017) Global Change Biology (see Figure 2 below), it showed that the normalized bias observed at the global scale is coherent with the underestimation we observe in the Mediterranean.

For comparison, we show in the following table taken from Richon et al. (2017, Prog. Oceanog.) the comparison of dust deposition from the ALADIN-Climat regional model with the measured values during the ADIOS campaign.
This Table illustrates that atmospheric deposition fluxes of mineral dust produced by ALADIN-Climat are higher than those from LMDz-INCA. However, the spatial variability of ALADIN is also low compared to point observations, and the fluxes are overestimated. These results seem to show that the 50 km resolution of the regional model ALADIN-Climat is still not enough to reproduce the spatial variability observed in the measurements. Moreover, the model ALADIN-Climat does not include sources of atmospheric P other than desert dust at the moment, which explains that it is not used in our present study.

Pg 7, lines 221: lines 226: also in this case the estimates for the total deposition flux by the model seem low. In a recent paper [Powley et al. (2017), Global Biogeochem. Cycles, 31, 1010-1031] Authors report atmospheric deposition rates of 0.16 109 mol/yr WMS and of 0.38 109 mol/yr EMS (see their Tab. 3). In the present manuscript the estimates are much lower. Given the lack of data it is difficult to reach a conclusion, but anyway this discrepancy raises the question of how robust is the discussion on spatial gradients if the average values present such an uncertainty. We thank the reviewer for this reference. Powley et al. use different estimations of total P deposition for their assessment, among which, the ADIOS campaign data included in our paper. The average values of Powley et al. over the basins are higher than our estimates. However, the measurements used include total P deposition (bulk deposition from all sources) whereas our model estimates do not include biogenic and volcanic sources. Moreover, the extrapolation of the deposition fluxes measured in a few localities to a basin scale average deposition flux as in Powley et al. (see their supplementary material) may lead to high uncertainties in the estimates. In particular, this method may not represent the important gradients in deposition from coastal to pelagic areas. We are conscious that neither the model, nor the measurements

can be representative of the full temporal and spatial variability of the fluxes, and that this variability is probably underestimated by both models and extrapolations of measurements. We agree, however, that is it important to consider all estimation methods in order to get a picture as precise as possible of atmospheric deposition over the Mediterranean.

Pg 8, line 239 "However, riverine inputs are the dominant external source of phosphate for almost all Mediterranean regions" Given the uncertainty on phosphorus deposition, and apparently its underestimation, this sentence appears not demonstrated. We modified this sentence to: "According to our model result, which remain highly uncertain, the riverine inputs computed from the PISCES model would constitute the main phosphate source to the Mediterranean Basin. They account for over 85

Pg 11, line 363: line 365 : "The atmospheric model LMDz–INCA has a low resolution given the regional Mediterranean scale: Pdust deposition forcing has 280x193 grid points globally and 500 grid points covering the Mediterranean, and Pcomb forcing has 144x143 grid points in total and 200 grid points covering the Mediterranean. These forcings reproduce well the average deposition patterns at the basin scale but may not be reliable when analyzing small scale deposition patterns." The statement that the global forcing reproduces well the average deposition should be somehow proved. In a recent article, Wang et al. (2017) compare the deposition fluxes of total phosphorus (from dust and combustion) from the LMDz-INCA model with measurements from all regions of the world. The following Figure (Figure 3c in their article) shows a good correlation between modeled and measured fluxes in spite of some underestimations in many regions, among which, the Mediterranean. We change the sentence to "These forcing reproduce realistic deposition patterns at the global scale, in spite of generally underestimating the measured fluxes. . ."

Pg 12, line 381:line 384 : "Natural dust emissions, transport and deposition to the Mediterranean are shown to be highly variable from a year to the next (e.g. Moulin et al., 1997; Laurent et al., 2008; Vincent et al., 2016) so that the relative contributions of Pcomb and Pdust may also vary." Authors focused on an average (or median) year, namely the 2005. But given the high inter-annual variability, at least of Pdust, what is the meaning of such a choice? It would be better to consider many years and analyse the temporal variability to have a better quantification of the reality? What is the role of extremes? As stated in the methods section, 2005 is the only available year for Pcomb deposition. We are conscious that reducing the simulation period to 1 year prevents us to conclude on inter annual variability effects. Naturally, conducting similar experiments with several deposition years would be necessary. Unfortunately, daily atmospheric deposition of combustion-derived phosphorus was only simulated for the year 2005. We consider that, given the high spatial and temporal variability of deposition fluxes, a monthly resolution of deposition, as available for other years (Wang et al. 2017, GCB) would be a too strong and unnecessary limitation in simulating the biological response. We added this sentence in the manuscript lines 151-154.
The monthly deposition of phosphorus from combustion has recently been simulated over the 1997-2012 period by Rong Wang. The following plot represents the yearly bioavailable phosphate deposition (in kg year-1) over the entire Mediterranean for the 1997-2012 period. In blue is the phosphate from combustion and in green the phosphate from dust. We can observe that 2005 is not an exceptional year in terms of deposition flux and that in spite of some inter-annual variability, mass deposition of phosphate from combustion seems to be, at the basin scale, always dominant over dust-derived phosphate deposition for the period at our disposal. We added a sentence in the discussion (line 390): "The relative dominance of combustion over dust as a source of phosphate for the Mediterranean seems to be confirmed by the analysis of yearly deposition fluxes of Pdust and Pcomb over the 1997-2012 period (not shown)."
Even though these deposition fluxes are only based on monthly values, we can

conclude that, on a yearly basis, combustion seems to be, according to this model, dominant over dust as a source of phosphate. However, the low temporal resolution of Pcomb deposition does not allow us to conclude on the importance of short term events such as Saharan dust outbreaks that may lead to local dominance of P deposition from dust.

Please also note the supplement to this comment:
https://www.biogeosciences-discuss.net/bg-2017-242/bg-2017-242-AC1-supplement.pdf

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Station	ADIOS	ALADIN (ADIOS period)	ALADIN (1982 - 2012)
Cap Spartel, Morocco	6.8 (108)	15 (135)	19 (42)
Cap Béar, France	11 (120)	18 (113)	15 (46)
Corsica, France	28 (275)	20 (116)	19 (55)
Mahdia, Tunisia	24 (127)	62 (124)	45 (52)
Lesbos, Greece	6.0 (101)	27 (115)	42 (79)
Crete, Greece	9.0 (199)	24 (129)	42 (78)
Akkuyu, Turkey	10 (99)	23 (119)	26 (79)
Cavo Greco, Cyprus	4.1 (63)	27 (120)	35 (80)
Alexandria, Egypt	21 (74)	30 (142)	31 (77)

Table 3: Dust deposition fluxes (g m^{-2} yr^{-1}) measured during the ADIOS campaign (derived from Al measured deposition fluxes considering that dust contains 7 % of Al), simulated by the ALADIN–Climate model (June 2001 - May 2002) and values simulated by ALADIN–Climate model on the whole period available (1982–2012). Values in brackets indicate the relative standard deviations of monthly fluxes calculated as (standard deviation)*100/mean.

Fig. 1. Dust deposition fluxes from the ALADIN-Climat model compared to the ADIOS campaign measurments (Table from Richon et al. 2017, Prog. Ocean.)

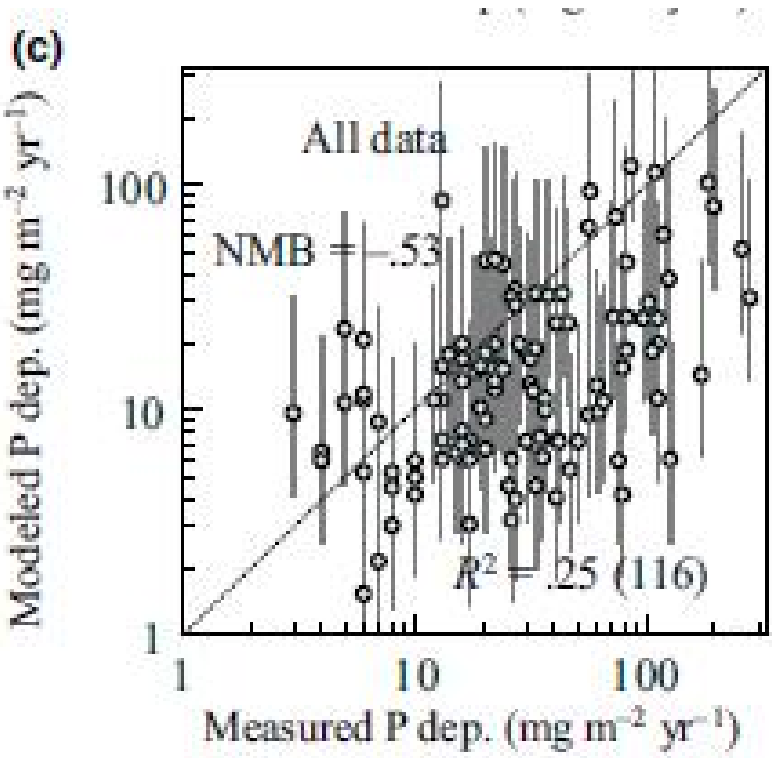

Fig. 2. Comparison of modeled and measured P fluxes for the global LMDz-INCA model. Figure from Wang et al. (2017, Global Change Biology)

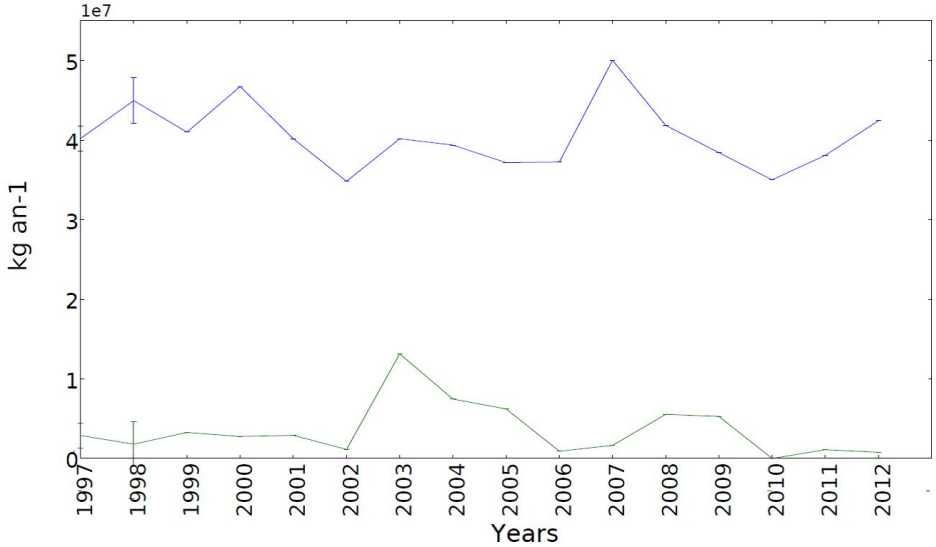

Fig. 3. Total deposition of soluble phosphate from combustion (blue line) and natural dust (green line) over the entire Mediterranean basin for the 1997-2012 period.