

We thank the referee for the careful reading of the manuscript and the fruitful comments that helped improving the presentation of our study. We have addressed all of them as described in the following point-by-point replies to the referee's comments.

Major points:

1) This is the first study which explicitly models the evolution of the state of the phosphorus in the atmosphere, which is an important innovation. I think you should highlight this in the abstract, introduction and conclusions.

We thank the reviewer for recognizing the value of our study. The third and fourth sentences of the abstract now read: "The P solubilisation from mineral dust under acidic atmospheric conditions is also parameterized in the model and is calculated to contribute about one third ($0.14 \text{ Tg-P yr}^{-1}$) of the global DP atmospheric source. To our knowledge, this is the first global study that explicitly models the evolution of phosphorus speciation in the atmosphere."

Furthermore a sentence has been added in the last paragraph of the introduction "To our knowledge, this is the first study that accounts for both inorganic and organic forms of P and their evolution in the atmosphere".

The first sentence in the conclusion has been accordingly modified as follows: "Primary TP and DP emissions accounting for both inorganic and organic P and for the atmospheric processing of P are taken into account for the first time in the state-of-the-art atmospheric chemistry transport global model TM4-ECPL".

2) More careful consideration of size. As shown by the contrast between Wang et al., 2014 and [J Brahney et al., 2015], there is a large sensitivity in the budgets of P to assumptions about aerosol size. Please discuss in the methods section the sizes you are considering within the model. Please also discuss the deposition data, and whether it includes sizes of particles you don't include, and so some part of that mass should be neglected (e.g. Brahney et al., 2015 discussion of sizes).

In our reply to reviewer #1 comment #8, we provide details on the size distribution we consider in the model for the P related tracers. Note we use additional model tracers to represent phosphorus in our model and modal approach to account for the size of the P-containing particles in fine and coarse modes with the mean mass diameters to be dependent on the source categories. Details are provided in our reply to reviewer #1 (comment 8). During atmospheric transport there are major changes in the size distribution of dust as a consequence of the stronger removal of larger particles due to gravitational settling. All details about the modal sizes used in the model to represent P-aerosols are already in the methods section in the source respective P-emissions sub-sections. However a short summary on the treatment of the P-aerosols in our model has been added at the end of the introduction of section 2 on methods. There, we also clarify the number of additional model tracers used to represent P in our model (32) as well as the fact that the size distribution of P-containing aerosols is changing in the model as a result of emissions, atmospheric transport and removal processes.

Furthermore, deposition data in particular wet or bulk dry deposition data include all sizes of aerosols, not only fine and coarse used in TM4 but also super coarse that are deposited close-by their sources, practically in the same grid box of the model where they are emitted.

In the introduction we have modified the 7th paragraph of the discussion on the P emissions to provide notions on the size distribution assumption in each source estimate: "the estimates of global strength of the primary P combustion source vary by about an order of magnitude on the global scale, due to the consideration of different forms of the emitted P (i.e. residual or P-containing ash, gaseous or particulate P produced during combustion processes; Wang et al. (2014)) and different size distributions in the emitted P-containing particulate matter. Mahowald et al. (2008) using observed mass ratios of P to Black Carbon (BC) for fine ($<2 \mu\text{m}$) and coarse ($2 \mu\text{m} \leq \text{mean particle diameter} < 10\mu\text{m}$) particles, calculated emission fluxes from biomass burning and anthropogenic fuel (i.e. fossil fuel and biofuel) combustion of $0.03 \text{ Tg-P yr}^{-1}$ and $0.05 \text{ Tg-P yr}^{-1}$, respectively. Tipping et al. (2014) estimated a global atmospheric P emission flux of 3.7 Tg-P yr^{-1} by combining observed deposition rates over land together with modelled deposition rates over the ocean. This emission flux, also accounts for P deposition fluxes of larger particles (e.g. primary biological material, hereafter PBAP, in the aerosol mode $\gg 10\mu\text{m}$) that are mainly deposited very close to their source region and thus not long-range transported. On the other hand Wang et al. (2014), by assuming that combustion processes emit significant amounts of P as large particles $> 10\mu\text{m}$ (hereafter as super-coarse particles) calculated that P

emissions from biomass burning and anthropogenic combustion processes can contribute about 0.7 Tg-P yr⁻¹ and 1.8 Tg-P yr⁻¹ respectively. In contrast to that study, which was more focused on the impact of anthropogenic combustion on the global P source, Brahney et al. (2015) extended the methodology of Mahowald et al. (2008) in a more explicit aerosol size manner by taking into account also the naturally emitted super-coarse P-containing particles (i.e. dust, PBAP and sea-salt). Brahney et al. (2015) showed that considering this super-coarse fraction as an additional P source, the estimated deposition fluxes close to the source areas where large particles are emitted (e.g. Tipping et al., 2014) can be significantly improved.”

Furthermore, in section 2.1.1 we have added the following discussion:

“Note also, that recent studies indicate that dust super-coarse particles can be very important for the biogeochemistry over land, since they can represent the dominant fraction of dust close to source regions (Lawrence and Neff, 2009; Neff et al., 2013). Brahney et al. (2015) modelling study that focused on the atmospheric phosphorus deposition over global alpine Lake, based on Neff et al. (2013) observations, estimated that only 10% of the mass that travels in the atmosphere is within the <10 µm size fraction. In our study we do not account for super-coarse dust particles because due to their short atmospheric lifetime, they are emitted and deposited in the same model grid box (Brahney et al., 2015). This omission is not expected to have significant impact on our results, since the present work is focused on the P-solubilisation mechanisms occurring via atmospheric long-transport mixing and on the bioavailable P deposition over the marine environment.”

Therefore the size distribution of the emissions is very important for the model evaluation. Section 2.3 on observations used for model evaluation has been improved to provide information on the sizes of observed aerosols. Similarly such information is provided in section 3.2 on model evaluation. The discussion in this section has been modified to present model evaluation distinguishing for aerosol sizes when available. Figures S4 and S6 have been modified to present size segregated comparisons. In the present study we do not account for super-coarse dust or sea salt aerosol, while we consider the emissions of pollen that are super-coarse aerosols. Therefore, it is expected that deposition fluxes close to dust source areas are underestimated by the model. Due to the small contribution of sea salt to the P-aerosol budget, the omission of sea salt super coarse aerosol can affect local comparisons but overall does not introduce more than a 3% underestimate of DP flux over the ocean (relevant comment has been added in section 2.1.4 and 3.2).

At the end of section 2.1.4:” The omission of the super coarse sea salt aerosol might affect our estimates of P deposition to the ocean. Brahney et al (2015) evaluated this source at 0.0046 Tg-P yr⁻¹, an amount that introduces a 3% underestimate to the here calculated present-day P deposition flux to the oceans.

In the 3rd paragraph of section 3.2: “The omission of super-coarse marine DP sources associated with sea-salt particles can explain some discrepancies between model results and observations only when these later concern bulk aerosols in oceanic regions (so they could include super-coarse particles), which is the case for wet or dry deposition samples. As discussed in Sect. 2.1.4, this omission can affect local comparisons but overall does not introduce more than a 3% underestimate of DP flux over the ocean. In many cases, aerosol samples have been collected with inlet devices that enable collection of specific fractions of aerosols and eliminate super-coarse particles. When bulk aerosols have been collected, then the presence of super-coarse aerosols might introduce discrepancies between model results and observations. Overall the model performs better for DP dry deposition fluxes over the oceans than over land, indicating a possible underestimate in the continental source of P. “

3) More description of the observations and how you are comparing to them. Please add a section of the methods talking about your observations. It’s unclear in your scatter plot where the data comes from and how you are comparing the deposition data (e.g. are the sizes consistent?). Could you show on one of your plots where the data comes from in these scatter plots (a little x?), and maybe show your observations from the cruises in a different color than the observations from the station on the scatter plot? Please discuss a little bit the differences in these observations and their value for your comparison (e.g. temporal variability vs. cruises showing spatial+temporal variability). Also, please include a description of your metric within the methods section (not a reference to another paper in the results section).

Following reviewer's 1 comment 18, we have added two maps in Figure 4a,b that show the location of the measurements. Different measurements (from cruises, stations) have been marked in different colors in the scatter plots. A subsection 2.3 on Data for model evaluation has been added in section 2, where the description of the normalized mean bias (NMB) used to compare model results with observations is now provided. In addition Tables S1 and S2 have been added in the supplementary material to provide information on the species, size, date, location and reference of the observations used for the model evaluation. As we have now performed a simulation for an 11-years period (2000-2010) that covers most of the observational data, the observations are compared to model results that correspond to the day of the observations. In addition, "observations are also spatially averaged inside the same model grid box" (as is mentioned in section 3.2.)

4) Compare % soluble observations vs. model? You get really high solubilities far away from the sources. Is there any evidence of this? Perhaps if you compare % solubility instead of soluble P amount, it might make your case more compelling that you are doing the solubility right? In a sense the P amounts are dominated by getting the P sources, and the right size comparisons, but the solubility, which is the real innovation in this study, might be better explored by the % solubility in the obs. Vs. model? Even if there is no evidence of these high solubilities, you are underestimating soluble P, so maybe underestimating % solubility close in, so maybe we should believe these high solubilities?

Unfortunately to our knowledge only few observations exist with simultaneous measurements of soluble and total P that provide information on the solubility of P that could be used for such evaluation and most of these are not open ocean data. In addition, the P solubility shown in Figure 7a is computed as the fraction of the sum of the soluble organic and inorganic P to the total P.

The following discussion has been added after the first sentence in section 3.5 on P solubility:

"Over such remote oceanic regions, high solubility fractions are calculated due to low P-containing aerosol mass concentrations, that occur via the long-range transport of fine particles from distance source regions, and the P which is associated with more aged aerosols and thus a greater fraction is present in the soluble mode; either as DIP via mineral acid solubilisation processes or DOP via atmospheric oxidation of P-containing organic aerosols and as PBAPs. Vet et al. (2014) in their review paper for nutrients deposition, also mentioned that the P solubility fractions of wet-only samples on coastal and inland sites have been measured to range from 30% to 90%, reflecting thus the effects of combustion, biomass burning, and phosphate fertilizers on airborne phosphorus concentrations. Anderson et al. (2010) reported that only 15-30 % of P in atmospheric aerosols at the Gulf of Aqaba was water soluble phases or relatively soluble to be bioavailable to the ecosystems. In the Mediterranean the measured median solubilities of the inorganic fraction of P in aerosols (ratio of PO_4^{3-} to total inorganic P) range between 20% and 45% in the East Mediterranean with the lowest values in dust influenced air masses and the highest values in air masses from the European continent (Markaki et al., 2003; Herut et al., 1999) and have been reported to be around 38% in the West Mediterranean (Markaki et al., 2010). However, simultaneous observations of TP and DP deposition fluxes are required to evaluate the solubility fraction of P (both organic and inorganic) over remote oceans and thus to understand the atmospheric fate of P. There are only a few aerosol data available in the literature for the marine atmosphere (Graham and Duce, 1982; Baker et al., 2006a; Baker et al., 2006b; Zamora et al., 2013) that provide hints on the total P solubility. These data indicate P solubilities ranging overall between 0.01% and 94%, with the lowest values corresponding to dust influenced air masses and the highest to sea-salt influenced air masses. Over the northern hemisphere Atlantic ocean P solubilities in aged Saharan dust aerosols have been measured to range from 0.01 to 37% during oceanographic cruises (Baker et al., 2006a; Baker et al., 2006b). At Barbados island median solubilities of P on dust of about 19% and of sea-salt aerosol of about 94% have been reported (Zamora et al., 2013). In the southern Atlantic atmosphere P-solubilities in aerosols of up to 67% (median 8% for dust aerosol and 17% for southern Atlantic aerosol; Baker et al., 2006a) and of up to 87% (median 32%; Baker et al., 2006b) have been reported. These studies but one report P solubility as the ratio of PO_4^{3-} to TP, thus neglecting the organic fraction which has been measured to be about 28-44% (Zamora et al., 2013). Although these observations support high P solubilities in aged aerosols or aerosols impacted by non-dust sources supporting the findings of our modelling study, only the work by (Zamora et al., 2013) could be compared to the here simulated total P solubility (Fig. 7a). They indicate that the model simulated total P solubility is at the upper edge of observed P solubilities.

5) You make the case that your results suggest a more important soluble P sources from biogenic aerosols. Why do you get a larger source than previous studies? Is it because you assume more of the biogenic are bioavailable, or are your sources larger? Just add a sentence or two on this.

There was a previous explicit global estimate of this source. The Kanakidou et al. (2012) estimate of OP from PBAPs is of the same order of magnitude with the present estimate. However, that study did not compare with the DIP deposition. In addition Mahowald et al. (2008) estimated total PBAP emissions at $0.164 \text{ Tg-P yr}^{-1}$ and considered this amount to be by 50% soluble P ($0.082 \text{ Tg-P yr}^{-1}$ DIP), while the dust soluble P estimate was $0.115 \text{ Tg-P yr}^{-1}$ i.e. of the same order of magnitude with the total PBAPs emissions. So the results are very similar, in our study we are just focusing on the importance of this finding that however needs to be consolidated with additional new observations because both the PBAPs sources and the dust-P solubilisation kinetics are uncertain. In addition here we consider that all biological material is bioavailable.

Details:

1) “primary and secondary sources of P” Are there any secondary sources of P in your approach?

With ‘primary sources’ we meant the P (either TP or DP) emissions while with ‘secondary sources’ we meant the DP released in the atmosphere due to solubilisation processes. To avoid, however, misunderstanding we changed this as remove this part of the sentence and replace it simply by ‘*P sources*’.

2) “Okin et al. (2011) evaluated the impact of Fe and P atmospheric deposition to the ocean in increasing N₂-fixation and found that Fe deposition is more important than P deposition in supporting N₂-fixation, while they pointed out the large uncertainty in the bioavailability of atmospherically deposited P.” There are also ocean biogeochemical model studies which show this same results either: [A Krishnamurthy et al., 2010] or [R Wang et al., 2015] which also suggest that atmospheric deposition of P doesn’t matter because of large P reservoirs in ocean.

It is true that the deep ocean is a major source of P for the surface seawater. However depending on season, the water stratification can minimize the impact of the deep water to the upper layers, This is mainly occurring in summer and it is during that period that the atmospheric deposition of P is expected to have the largest impact on the marine ecosystems.

Notions are provided in the abstract: “...in summer when atmospheric deposition impact on the marine ecosystem is the highest due to ocean stratification.”

In the last paragraph of section 3.4: “The maximum DP deposition flux in summer occurs when ocean stratification also maximizes thus leading to the highest impact of atmospheric deposition to the marine ecosystems (Christodoulaki et al., 2013).”

And almost at the end of section 4.2: “It is also noteworthy that the bioavailable P deposition flux from bioaerosols maximizes in summer (Fig. S8e-h) when ocean stratification is also the strongest, thus leading to the highest impact of atmospheric deposition to the marine ecosystems (Christodoulaki et al., 2013).”

Furthermore, there are regions of the global ocean, like the Mediterranean that are P-limited. We have added a comment at the end of the first paragraph of the introduction:

“However, in some regions like the Mediterranean, primary productivity is found to be limited by P-availability to the marine ecosystems (Krom et al., 2005). Furthermore, Brahney et al. (2015) and Du et al. (2016) found that human driven imbalanced atmospheric deposition of N and P might have induced or will induce P-limitation to the ecosystems (global alpine lakes and large areas of China’s forests, respectively).”

3) Wang et al. (2014) taking into account the potential volatilized-P produced during combustion processes, calculated about 30 times higher global atmospheric P emissions from

biomass burning and anthropogenic combustion processes (0.7 Tg-P yr⁻¹ and 1.8 Tg-P yr⁻¹ respectively).” This is not accurate.

“Primary P sources from combustion processes of anthropogenic and biomass burning origin are estimated to contribute significantly to global P fluxes in the atmosphere (Mahowald et al., 2008; Tipping et al., 2014; Wang et al., 2014; Brahney et al., 2015). However, the estimates of global strength of the primary P combustion source vary by about an order of magnitude on the global scale, due to the consideration of different forms of the emitted P (i.e. residual or P-containing ash, gaseous or particulate P produced during combustion processes; Wang et al., 2014) and different size distributions in the emitted P-containing particulate matter. Mahowald et al. (2008) using observed mass ratios of P to black carbon (BC) for fine (<2 μm) and coarse (2 μm ≤ mean particle diameter < 10 μm) particles (Mahowald et al., 2005), calculated emission fluxes from biomass burning and anthropogenic fuel (i.e. fossil fuel and biofuel) combustion of 0.03 Tg-P yr⁻¹ and 0.05 Tg-P yr⁻¹, respectively. Tipping et al. (2014) estimated a global atmospheric P emission flux of 3.7 Tg-P yr⁻¹ by combining observed deposition rates over land together with modelled deposition rates over the ocean. This emission flux estimate however, also accounts for P deposition fluxes of larger particles (i.e. primary biological material in the aerosol mode >> 10 μm) that are mainly deposited very close to their source region and thus not long-range transported. On the other hand Wang et al. (2014), by assuming that combustion processes emit significant amounts of P as large particles > 10 μm (hereafter as super-coarse particles) calculated that P emissions from biomass burning and anthropogenic combustion processes can contribute about 0.7 Tg-P yr⁻¹ and 1.8 Tg-P yr⁻¹ respectively. In contrast to that study, which was more focused on the impact of anthropogenic combustion on the global P source, Brahney et al. (2015) extended the methodology of Mahowald et al. (2008) in a more explicit aerosol size manner by taking into account also the naturally emitted super-coarse P-containing particles (i.e. dust, primary biological material and sea-salt). Brahney et al. (2015) showed that considering this super-coarse fraction as an additional P source, the estimated deposition fluxes close to the source areas where large particles are emitted (e.g. Tipping et al., 2014) can be significantly improved. “

4) Tipping et al., 2003 put together a compilation of deposition in ecosystems, and indicated that the observations suggest this deposition dominated by locally generated primary biogenic material, in the aerosol mode >10 μm which is not long range transported. Wang et al., 2014 used the mismatch in size between their <20 μm modeled aerosols and the observations in the source regions and assumed that this mismatch was only from combustion sources. Thus there is a serious methodological problem in the Wang et al., study, and they don't bother to compare against the available concentration data which would have revealed this problem (as you do here), nor the observation-based source apportionment in Mahowald et al., 2008, which was consistent with the much smaller combustion sources. Instead one should say perhaps: Wang et al. (2014) taking into account the potential volatilized-P produced during combustion processes by assuming that all mismatches between observed deposition (<1000 μm aerosols) and modeled-long-range transported (<20 μm) deposition was due to combustion, estimated about 30 times higher global atmospheric P emissions from biomass burning and anthropogenic combustion processes (0.7 Tg-Pyr-1 and 1.8 Tg-P yr-1 respectively).” Or simply don't refer to that paper here or mention it in passing, since it is deeply methodologically flawed. [J Brahney et al., 2015] discusses how to compare to the Tipping et al., data in a more explicit aerosol size manner, and extends the Mahowald et al., 2008 study, showing that one can match deposition and concentration observations at the same time.

Matching atmospheric deposition fluxes and concentrations at the same time is also what we try to do in the present study focusing on coastal and oceanic regions. See also our reply to reviewer's detailed comments point 3.

5) “where EDu is the on-line calculated dust emissions in the model, F880 is a factor applied to adjust the P emissions to the global mean P content of mineral dust in the model domain of 880 ppm per weight as observed by Zamora et al. (2013), and EP is the resulted inorganic P emissions from mineral. P-containing minerals associated with dust particles are emitted in the fine and coarse mode with mass median radii (lognormal standard deviation) of 0.34 μm (1.59) and 1.75 μm (2.00), respectively. The apatite emissions from mineral dust calculated for the year 2008 amount to 1.034 Tg-P yr⁻¹ with 10% of it (0.103 Tg-P yr⁻¹) in the dissolved form (Table 1).” How does this approach compare to the size resolved methods used in [J Perlwitz et al., 2015] for this mineral?

Perlwitz et al. (2015) study focused on Fe-containing minerals. For the present study, we did not account for different P content in different dust minerals since that information was not available in the database that we have used or between soil and aerosols. Although the repetition of Perlwitz et al. (2015) methods for apatite minerals is out of the scope of this study, we added a comment in the manuscript in section 2.1.1:

“In a recent iron modelling study however (Perlwitz et al., 2015), a significant effort has been made to model the mineral composition of dust considering the differences from the original soil composition. Perlwitz et al. (2015) have found significant overestimate (a factor of 10-30) mainly in the fine aerosol emissions that are the smallest part of dust emissions (e.g. about 7% of the total emissions in our model) and an underestimate in the larger particles emissions both for total dust and for individual minerals when the mineralogy of dust aerosol is assumed to be the same as that of the soil. However for the present study, we did not account for different P content for dust particles in the fine and the coarse mode, since the global soil mineralogy dataset used (Nickovic et al., 2012) does not provide any information of P content in silt and clay soil particles separately.”

6) Section 2.0: model description; can you describe your aerosol size bin or modal structure for the primary aerosols in P?

We use modal scheme and this is clarified in the introduction of section 2.0, see also our detailed reply to the comment 8 of reviewer 1.

7) Section 2.1.3: how do you include bits of insects and plants that would be part of PBAP? How important is the neglect of these terms to your budgets do you think?

PBAPs from insect fragments and plant debris are neglected in the present study. Omission of these super coarse particles is expected to lead to an underestimate in the PBAPs contribution to P deposition that requires to be evaluated with targeted observations. This is now clearly stated in the beginning of section 2.1.3.

We also added the following sentence in section 4.3: “Note that as mentioned in section 2, PBAPs from insect fragments and plant debris are neglected in the present study. Thus the contribution of PBAPs to bioavailable P deposition is here underestimated.”

8) Please fix English by adding preposition (e.g. of): “(i.e. Nigeria downwind the Sahara Desert, Pakistan downwind the Thar Desert and China downwind of the Gobi desert).”

Done.

9) In Fig. 4b, PO₄ deposition fluxes (wet and dry deposition) from the Vet et al. (2014) compilation and from observations at Finokalia Station (Mihalopoulos and co-workers, unpublished data) are also compared with the model derived fluxes for the PRESENT simulation.” What is the size distribution of the PO₄ in the deposition? Is it the same size as the modeled boxes? I also think you should present the data you are going to compare against as a section in the methods, and describe the characteristics of the data, especially as some of the data is from unpublished sources. We also need to know where this data comes from physically: is it all in Greece, or elsewhere?

A subsection 2.3 describing the data for model evaluation has been added in section 2, and the description of all data is now provided in the supplementary tables S1 and S2. In addition, Figures 4, S4, S5, S6 have been modified for clarify. Size segregated comparisons are now shown in these figures. Figures 4a and 4b illustrate the global distribution of the locations with aerosol concentrations and deposition fluxes data used for the model evaluation respectively.

1.1 The subsection 2.3 follows: “Observation data for model evaluation

The evaluation of the global atmospheric P cycle for the present study has been performed based on available observations of aerosol concentrations (Table S1) and deposition fluxes (Table S2) from various locations around the globe (cruises and land-based stations). The methodological details of the observations used for this study are well documented in the literature and thus are not reviewed here in detail. For DP concentrations in ambient aerosols, we compiled cruise observations of PO₄³⁻ over the Atlantic Ocean (50°N–50°S) from Baker et al. (2010), over the Western Pacific (25°N–20°S) from Martino et al. (2014) and over the Eastern Tropical North Atlantic Ocean (58°S–35°N, 14°–38°W)

from Powell et al. (2015). For these oceanic cruise observations, samples were either collected separating into fine- (aerodynamic particle diameter < 1µm) and coarse-mode (1µm < aerodynamic particle diameter) particles using cascade impactors that may include or exclude particles with diameters larger than 10 µm, or using a single bulk filter. We additionally use average PO₄³⁻ concentrations (aerodynamic particle diameter < 10µm) from cruise measurements over Bay of Bengal and the Arabian Sea (Srinivas and Sarin, 2012). Finally, we also took into account land-based TP and PO₄³⁻ aerosol concentrations measurements from two sites in the Mediterranean i) from the Finokalia monitoring station (35°20'N, 25°40'E) located in the Eastern Mediterranean (Crete, Greece) and ii) from Ostriconi (42°40'N, 09°04'E) located in the Western Mediterranean (Corsica, France). The samples at both sites were collected either separating for the fine- (aerodynamic particle diameter < 1.3 µm) and the coarse-mode (10 µm > aerodynamic particle diameter > 1.3 µm) (Koulouri et al., 2008; Mihalopoulos and co-workers, unpublished data) or as bulk (Markaki et al., 2010). Details about the characteristics of these Mediterranean sampling sites can be found in Markaki et al. (2010), while the methodology for aerosol sampling and analysis is described in detail in Koulouri et al. (2008).

Although P deposition fluxes data are rather limited on a global scale, for the present study we use the wet and dry deposition fluxes (both for TP and DP) compiled by Vet et al. (2014) (R. Vet, personal communication, 2016). For wet deposition of DP, we use available filtered (i.e. analyzed as orthophosphates with no digestion as DIP) and unfiltered (i.e. analyzed as orthophosphates following digestion as total DP) annual measurements (Fig. 8.2 in Vet et al., 2014). For the TP wet deposition measurements we use annual wet deposition measurements (Fig. 8.3 in Vet et al., 2014) of unfiltered samples. The compilation of the phosphorus dry deposition fluxes by Vet et al. (2014) is based on airborne phosphorus (TP and PO₄) concentrations from around the world and gridded annual dry deposition velocities from the Mahowald et al. (2008) modelling study (Fig. 8.6 and Fig. 8.7 in Vet et al., 2014). The size distribution used in these dry deposition calculations, is the same as in the modelling study by Mahowald et al. (2008), thus the derived dry deposition fluxes account for particles with diameter up to 10 µm. Finally, we also take into account DP wet and dry deposition observations from the Finokalia Station in the Eastern Mediterranean (Markaki et al., 2010; Mihalopoulos and co-workers, unpublished data), based on rain water samplings (wet only collector) and glass-bead devices respectively. Further details on the methodology of the deposition measurements at Finokalia can be found in Markaki et al. (2010).”

10) “(MNB; see definitions of statistical parameters in Myriokefalitakis et al. (2015))” You also need to describe your methods in the methods section: it is not ok to refer us for basic information to another paper.

We have now included the definitions of this statistical parameter – mean normalized bias (MNB) in section 2.3.

11) Figure 7: maybe you want to reformat so that there won't be too much white space in the final figure for this?

We have reformatted the figure as suggested.

12) The present-day P solubility of deposited aerosols (hereafter SP = %DP/TP) is calculated to vary spatially significantly (Fig. 7a),” vary spatially significantly is awkward: please rephrase and only use significantly if you mean statistically significantly.

We rephrased by removing ‘significantly’

13) For your past and future estimates: Your P is strongly dependent on dust, and yet you don't include any changes in dust. I don't think you need to add much here, but just some statements that dust appears to vary strongly and perhaps be sensitive to humans climate change and/or land use [P Ginoux et al., 2012; N Mahowald et al., 2010; J Prospero and P Lamb, 2003], and thus that would also be an important drivers of changes in P and SP.

We agree with the reviewer that past and future dust sources may be changed due to global change. In our model, P atmospheric cycle is strongly depended on dust outbreaks, since according to our calculations about 50% of the deposited bioavailable P is originated from soils for the present atmosphere. As recommended we added the following sentence in section 4.1. of the manuscript: “Although for this work we don't account for any changes in atmospheric dust emissions for PAST and FUTURE simulations, several studies suggest that dust may vary strongly and perhaps be sensitive to anthropogenic climate change and land use (Ginoux et al., 2012; Mahowald et al., 2010; Prospero and Lamb, 2003) and thus could also be an important driver of changes in the atmospheric P cycle.”

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