Letter to the Editor

Dear Dr. Grégoire,

Please find a revised version of the manuscript entitled: ”Modeling the biogeochemical impact of atmospheric phosphate deposition from desert dust and combustion sources to the Mediterranean Sea”, along with a track version that helps visualize the latest changes.

As suggested by reviewers, the manuscript was modified with respect to two specifics points: first, we improved the evaluation of model performances by adding statistical indicators in Figure 1, Table1 and Appendix A;

second, we scaled back our conclusions concerning the effects of phosphate deposition from combustion (Pcomb). Up to now, there are at least two major data sets of Pcomb. Mahowald et al. (2008) had published the first map of P deposition from combustion sources based on a bottom-up P emission inventory, which leads to a general agreement of modeled surface P concentrations with measurements but a large underestimation in the modeled P deposition. Wang et al. (2015, 2017) revised the emission factors from all fuel types burned during combustion to come up with a new global inventory of phosphorus emitted from coal, biofuels and biomass burning. This inventory amounts to an emission of P from these 3 sources of 1.96 Tg P yr-1 for the year 1996 (Wang et al., 2015), which is 28 times the inventory of 0.070 Tg P yr-1 compiled by Mahowald et al. (2008) for the same year. The same authors (Wang et al.) evaluated both P surface concentrations and P deposition and showed no systematic bias against measurements taken at the global scale. Inter comparing P deposition from atmospheric models is an interesting issue, but the goal of our study was to initiate investigation on the effects of different P atmospheric sources over the Mediterranean. The limited time period with daily deposition fields of Pdust and Pcomb limits our oceanic simulations to the sole year 2005. However, to bring these 1-year results in the context of 16 years of simulated monthly deposition fluxes, we added in Figure 5 the spatial repartition of Pdust and Pcomb deposition in the Mediterranean over the 1997-2012. This Figure, together with the time series of P deposition provided in our responses to the reviewers strengthen our conclusion on the domination of Pcomb as a source of atmospheric phosphate over the Mediterranean.

Even with the presence of the results from these new figures, we are conscious of the limitations due to the lack of measurements to validate the atmospheric deposition models over the 16-year period. It is therefore difficult to conclude on the nature of the dominant source over the Mediterranean. We tried to point out these uncertainties in our manuscript. We stress that this work consists of a first approach on the modeling of biogeochemical effects of phosphorus deposition from various types of sources over the Mediterranean. We believe these results call for more studies on atmospheric deposition measurements and modeling, and also on biogeochemical oceanic model developments to better constrain, and maybe challenge, our first observations.

Finally, our results suggest that the Mediterranean biogeochemical response to P deposition is primarily dependent on nutrient limitations in the water. Therefore, the effects of P deposition, independent of its source, will likely be negligible in regions that are not P-limited, although there may be some inter-annual variability in atmospheric deposition that we do not account for.

We sincerely hope that these improvements will answer all of the editor's and reviewers' questions.

Thank you for your implication,

Best wishes,

Camille Richon, on behalf of the co-authors.

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Modeling the biogeochemical impact of atmospheric phosphate deposition from desert dust and combustion sources to the Mediterranean Sea

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Abstract. ~~We used~~ Daily fields of phosphate deposition from natural dust, anthropogenic combustion and wildfires ~~simulated for the year 2005 by a global atmospheric chemical transport model (LMDz–INCA) as additional sources of external nutrient for~~ were used to assess the effect of this external nutrient on marine biogeochemistry. The model used is a high resolution (1/12°) regional coupled dynamical-–biogeochemical model of the Mediterranean Sea ~~. In general, dust is considered as the~~ (NEMOMED12/PISCES). The input fields of phosphorus are for 2005, which is the only available daily resolved deposition fields from the global atmospheric chemical transport model LMDz–INCA. Traditionally, dust has been suggested to be the main atmospheric source of phosphorus, but the LMDz–INCA model suggests that combustion is dominant over natural dust as an atmospheric source of phosphate (PO$_4$, the bioavailable form of phosphorus in seawater) for the Mediterranean Sea. According to the atmospheric transport model, ~~anthropogenic~~ phosphate deposition from combustion (*Pcomb*) brings on average 40.5 10^{-6} mol PO$_4$ m^{-2} year^{-1} over the entire Mediterranean Sea for the year 2005 and is the primary source over the northern part (e.g., 101 10^{-6} mol PO$_4$ m^{-2} year^{-1} from combustion deposited in 2005 over the North Adriatic against 12.4 10^{-6} from dust). Lithogenic dust brings 17.2 10^{-6} mol PO$_4$ m^{-2} year^{-1} on average over the Mediterranean Sea in 2005 and is the primary source of atmospheric phosphate to the southern Mediterranean basin in our simulations (e.g., 31.8 10^{-6} mol PO$_4$ m^{-2} year^{-1} from dust deposited in 2005 on average over the South Ionian basin against 12.4 10^{-6} from combustion). We examine separately the ~~different soluble phosphorus (PO$_4$)~~ two atmospheric phosphate sources and their respective fluxes variability and evaluate their impacts on marine surface biogeochemistry (phosphate ~~concentrations~~concentration, Chl *a*, primary production). The impacts of the different phosphate deposition sources on the biogeochemistry of the Mediterranean are found localized, seasonally varying and small, but yet statistically significant. ~~The impact of the different sources of phosphate on the biogeochemical cycles is remarkably different and should~~ Differences in the geographical

deposition patterns between phosphate from dust and from combustion will cause contrasted and significant changes in the biogeochemistry of the basin. We contrast the effects of combustion in the northern basin (*Pcomb* deposition effects are found 10 times more important in the northern Adriatic, close to the main source region) to the effects of dust in the southern basin. These different phosphorus sources should therefore be accounted for in modeling studies.

1 Introduction

Atmospheric deposition is an important source for bioavailable nutrients to the remote oceanic waters (e.g. Jickells, 2005; Mahowald et al., 2009). Aerosols not only include nutritive elements such as nitrogen and phosphorus, which are the main limiting nutrients for marine biology, but also trace metals (Dulac et al., 1989; Heimburger et al., 2012), among which copper has toxic effects on some phytoplankton species (Paytan et al., 2009). Aerosols can even be associated with living organisms such as viruses, fungae and bacteria (Mayol et al., 2014). The most important aerosol mass 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. In terms of nutrient fluxes, silica, nitrogen, iron and phosphorus are most abundant among the deposited nutrients (Guerzoni et al., 1999). Nitrogen, phosphate and iron are the three most important deposited elements measured in the Gulf of Aqaba, which is under the influence of both natural and anthropogenic aerosols (Chen et al., 2007). It is especially important to constrain external sources of phosphorus because it limits productivity (either as a primary, or secondary limiting nutrient) in many regions of the oceans (Moore et al., 2013). 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).

The Mediterranean Sea is highly oligotrophic and the intense summer vertical stratification leads to rapid nutrient depletion in surface waters (Bosc et al., 2004). During that season, the atmosphere is the only nutrient source for most of the Mediterranean surface waters (Markaki et al., 2003). Many studies discuss the impacts of atmospheric nutrient deposition to the oligotrophic Mediterranean Sea surface waters (Guerzoni et al., 1999; Markaki et al., 2003; Gallisai et al., 2014). Monitoring and experimental studies have shown that deposition of great amounts of aerosols significantly impacts surface biogeochemistry over this basin (see Herut et al., 2005; Guieu et al., 2014). Ridame et al. (2014) showed that extreme events of Saharan dust deposition can double primary production and Chl *a* concentration. In particular, the soluble fraction of this aerosol provides the main limiting nutrient to the Mediterranean: inorganic phosphorus (Bergametti et al., 1992; Krom et al., 2010; Tanaka et al., 2011).

Until now, Saharan dust was believed to be the most important atmospheric source of nutrients to the oligotrophic Mediterranean (e.g., Guerzoni et al., 1999). But the Mediterranean region is one of the most densely populated areas of the world and many of the surrounding countries historically developed their capital cities along its coasts. The recent development of many of the Mediterranean countries has led to high anthropogenic footprint over ecosystems and climate through increased population and industrial activities emitting aerosols (Kanakidou et al., 2011). 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~~. Moreover, aerosols are deposited to the Mediterranean Sea from a variety of different geographical sources. The impacts of aerosol deposition on the Mediterranean region are not fully understood and they may change in the future as a result of climate change impacts on land and sea. The Sahara and Middle East are important sources of natural lithogenic dust (e.g., Ganor and Mamane, 1982; Bergametti et al., 1989; Al-Momani et al., 1995; Vincent et al., 2016) whereas the surrounding cities and highly industrialized areas are sources of atmospheric pollutants emitted by biofuels for heating and fossil fuel burning (Migon et al., 2001; Piazzola et al., 2016). The 85 million hectares of forests around the basin associated to the Mediterranean dry summer climate are also an occasional intense aerosol source due to wildfire emissions (Kaskaoutis et al., 2011; Poupkou et al., 2014; Turquety et al., 2014), providing for instance soluble iron to the Mediterranean (Guieu et al., 2005).

Modeling represents an interesting approach to investigate the impact of atmospheric nutrient deposition on oceanic biogeochemical cycles. Richon et al. (2017) use a regional coupled dynamical–biogeochemical high resolution model of the Mediterranean Sea to study the impacts of N deposition from natural and anthropogenic sources and phosphate from dust on the biogeochemistry of the Mediterranean Sea. In the present study, we extend this investigation of phosphate deposition effects by further considering the contribution of P from combustion sources from anthropogenic activities and wildfires, and comparing the effects of desert dust and combustion inputs of phosphate on the marine surface nutrient and biogeochemical budgets.

2 Methods

We use mass deposition outputs from the global atmospheric model LMDz–INCA (Hauglustaine et al., 2014; Wang et al., 2015a) as external sources of phosphate in the regional high resolution coupled NEMOMED12/PISCES model. We consider separately phosphorus from desert dust and from combustion in order to isolate their respective effects as nutrient sources.

2.1 The oceanic model

We use the regional oceanic model NEMO (Madec, 2008) at a high spatial resolution of 1/12° over the Mediterranean (MED12). The 1/12° ~~ORCA~~ grid resolution is stretched in latitude and ranges between 6 km at 46° N and 8 km at 30° N. This fine–scale resolution enables us to represent important features of the Mediterranean circulation that are small eddies. This grid has 75 unevenly spaced vertical layers, with depth ranging from 1 to 134 m from the surface to the bottom, and 10 levels in the first 100 m. The oceanic domain covers all the Mediterranean and a part of the Atlantic between the Strait of Gibraltar and 11° W called the buffer zone. The regional NEMO–MED dynamical simulation used in this study to force the PISCES model is NM12–FREE, evaluated in Hamon et al. (2015). Atmospheric forcing conditions are prescribed from the ALDERA dataset (Hamon et al., 2015). Temperature and salinity are relaxed monthly to climatologies in the buffer zone (Fichaut et al., 2003). This simulation reproduces well the general circulation and variability of the water masses characteristics. However, the authors identify some shortcomings: transports through the Strait of Gibraltar are underestimated by about 0.1 Sv, the circulation and mesoscale activity in the western basin (Algerian current, Northern current) are underestimated, and positive temporal drifts in the heat and salt content occur in the intermediate layer all over the Mediterranean Sea. This may lead to overestimation in temperature and salinity in intermediate waters after long simulation periods (hundreds of years). The ability of the model to reproduce the general circulation of the water masses was also evaluated in a similar configuration with CFC (Palmiéri et al., 2015), neodymium (Ayache et al., 2016a), tritium–helium–3 (Ayache et al., 2015) and ^{14}C (Ayache et al., 2016b). These evaluations showed ~~satisfying results~~that the NEMO model is able to produce satisfying results when studying characteristics such as age–tracer of water masses or passive tracer transport.

The biogeochemical model PISCES (Aumont et al., 2015) is coupled to the physical model. PISCES is a Monod–type model (Monod, 1958), in which biological productivity can be limited by the availability of nitrate, ammonium, silicate, iron and phosphate. The concentration of nutrients is linked to phytoplankton productivity and chlorophyll *a* production according to the equations described in Aumont et al. (2015). Phytoplankton growth rate is dependent on nutrient concentrations via the growth limiting factors (see Aumont et al., 2015, for detailed equations). Only two biological levels are explicitly represented in PISCES: phytoplankton (autotrophic) and zooplankton (heterotrophic). Each plankton type is composed of two size classes: nanophytoplankton and diatoms; microzooplankton and mesozooplankton. PISCES is a Redfieldian model: the C~~/N/~~:N:P ratio used for ~~biology~~ plankton growth is fixed to 122~~/~~:16~~/~~:1. The ~~latest developments of the PISCES model are described in Aumont et al. (2015). The~~ regional coupled configuration NEMOMED12/PISCES was developed by Palmiéri (2014) and Richon et al. (2017). We prescribe riverine nutrients ~~inputs~~ input fluxes from the estimation of Ludwig et al. (2009) ~~.~~ that accounts for the nutrient fluxes from 239 rivers around the Mediterranean and Black Sea obtained from measurements and model data. The estimations of riverine fluxes are not available after 2000. Therefore, we use the riverine fluxes from the year 2000 in our study. Nutrient concentrations in the buffer zone are relaxed to the monthly climatology of the World Ocean Atlas (WOA) (Locarnini et al., 2006). Nutrient fluxes from the Atlantic are computed in the model as the product of the nutrient concentrations in the buffer zone times the water fluxes through the Strait of Gibraltar.

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 driven by dynamical variables (velocities, pressure, mixing coefficients...) previously calculated by the oceanic model NEMO. Biogeochemical variables are prognostically calculated instead of being read from forcing files. Biogeochemical characteristics of the latest version of the NEMOMED12/PISCES model are evaluated in Richon et al. (2017). The model ~~satisfyingly reproduces the vertical distribution of nutrients in~~ NEMOMED12/PISCES is run in the same configuration than in Richon et al. (2017) in which an evaluation of the model can be found. In particular, NEMOMED12/PISCES produces well the West–to–East gradient of productivity when compared to satellite chlorophyll *a* estimates, and simulates the ~~basin and the~~ main productive zones ~~that are~~ located in the Alboran Sea, the Gulf of Lions and most coastal areas ~~(see appendix).~~ but with a lower amplitude (see Figure 1 and Table 1). We filtered out chlorophyll *a* values in coastal areas (Bosc et al., 2004). The vertical distribution of nutrients is globally satisfyingly simulated, despite a too sharp nutricline in the intermediate waters in some sub–basins (e.g. Alboran, South Ionian).

2.2 ~~Atmospheric deposition of phosphate~~

~~We use atmospheric~~ Additional statistical indicators provided in Appendix A show a good reproduction of the nutrients and the chlorophyll *a* vertical concentrations in different Mediterranean regions. However, the model fails to reproduce surface and intermediate phosphate concentrations in some regions such as the South Levantine. It is important to note that the nutrient concentrations in surface waters, especially phosphate concentrations in the ultra–oligotrophic eastern basin, are often below the detection limit of measuring devices. Therefore, the negative bias in model surface concentration estimates may be linked with measurement uncertainties. Richon et al. (2017) calculated the average and standard deviation of chlorophyll *a* values measured and modeled at the DYFAMED station (Ligurian Sea) for the 1997–2005 period and 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 PO$_4$, the average measured value is $0.234 \pm 0.085\ 10^{-3}$mol m^{-3} and the modeled average is $0.167 \pm 0.179\ 10^{-3}$mol m^{-3}. We report in Appendix A additional figures on the comparison of modeled and measured PO$_4$ and NO$_3$ concentrations. Despite some unavoidable shortcomings, the performances of the model are reasonable for conducting our scientific investigation.

2.2 Atmospheric deposition of phosphate

The objective of this study is to use consistent atmospheric phosphate inputs from contrasted sources simulated by the same atmospheric model. Hence, we selected daily atmospheric deposition fields of total phosphorus ~~from two different emission sources, namely~~ (P) from natural dust and combustion both simulated with the LMDz–INCA chemistry–climate global model (Wang et al., 2015b).

~~For the two sources,~~ This global model has a rather low spatial resolution of 0.94° in latitude × 1.28°in longitude. The daily deposition fluxes ~~are simulated globally for 2005.~~ from these two sources have been simulated globally solely for a one year period (2005). The form of deposited ~~phosphate~~ phosphorus in the model is the same for all sources and is considered to be soluble phosphate (PO_4^{3-}) which is the bioavailable form of phosphorus in PISCES (Aumont and Bopp, 2006). We considered that given the high spatial and temporal variability of atmospheric deposition fluxes, a monthly resolution of deposition, as available for other years (Wang et al., 2017), would be a too strong and unnecessary limitation in simulating the biogeochemical response.

~~The first source of phosphorus we include in this study is natural dust~~In this study, we first include natural desert dust as a source of phosphorus. In the atmospheric model, ~~natural~~ desert dust emissions are computed every 6 hours using the European Centre for Medium-Range Weather Forecasts (ECMWF) wind data interpolated to the LMDz grid. Following Mahowald et al. (2008), we consider that only 10 % of ~~P in~~ phosphorus from desert dust is bioavailable PO_4 (hereafter named *Pdust*). This solubility value ~~is an average that is also~~ has also been used by other authors (see also Anderson et al., 2010). Izquierdo et al. (2012) report an average solubility of phosphorus of about 11 % in African dust–loaded rains in North–East Spain. ~~The resolution of the deposition fluxes of *Pdust* from LMDz–INCA is 0.94° in latitude × 1.28in longitude. These fields are daily averaged.~~ In addition, the daily total dust deposition simulated with similar forcings on a 1.27°in latitude by 2.5°in longitude grid is available for the period 1997–2012. We used this time series of total dust deposition to compare the year 2005 with the average inter–annual deposition flux~~(not shown)~~. We found that the yearly average deposition of dust ~~in~~ from 2005 is close to the multi–year ~~deposition average~~ depositional average (not shown).

The second source of atmospheric phosphorus is combustion. Here, the term combustion entails anthropogenic combustion from energy production, biofuels, and wildfires emissions (hereafter named *Pcomb*). The *Pcomb* deposition fields from LMDz–INCA used here have a coarser resolution than for *Pdust* of 1.27°in latitude by 2.5°in longitude. In the atmospheric model, phosphorus emissions from combustion due to anthropogenic activities are assumed constant throughout the year 2005, only wildfire emissions vary monthly based on the GFED 4.1 data set for biomass burning (van der Werf et al., 2010). According to this data set, wildfire emissions around the Mediterranean for 2005 are close to the inter–annual average for the period 1997–2009. Wang et al. (2015a) estimated global *Pcomb* emissions based on the consumption of different fuels (including wildfires) and the P content in all types of fuels for more than 222 countries and territories. We consider that 54 % of the total emitted P from combustion is bioavailable phosphate (*Pcomb*) (Longo et al., 2014). Up to now, there are at least two major data sets of *Pcomb* deposition. Mahowald et al. (2008) had published the first map of P deposition from combustion sources based on a bottom–up P emission inventory, which leads to a general agreement of modeled surface P concentrations with measurements with a large underestimation in the modeled P deposition. Wang et al. (2015a, 2017) revised the emission factors from all fuel types burned during combustion to come up with a new global inventory of phosphorus emitted from coal, biofuels and biomass burning. This inventory amounts to an emission of P from these 3 sources of 1.96 Tg P yr^{-1} for the year 1996 (Wang et al., 2015a), which is 28 times the inventory of 0.070 Tg P yr^{-1} compiled by Mahowald et al. (2008) for the same year. The same authors (Wang et al., 2015a) evaluated both P surface concentrations and P deposition and showed no systematic bias against measurements taken at the global scale.

The LMDz–INCA modeled atmospheric P deposition ~~rates~~ fluxes have been evaluated globally by comparing time series of deposition measurements, showing a significantly reduced model bias relative to observations when considering the contribution of P emissions from combustion than when considering only P from dust (Figure 4 in Wang et al., 2015a). However, it should be noted that there were only three sites with time series of P deposition over the Mediterranean region. ~~We consider that 54 % of the total emitted P from combustion is bioavailable phosphate (*Pcomb*) (Longo et al., 2014).~~

Another important ~~source~~ input of P aerosols in this region is from sea spray (Querol et al., 2009; Grythe et al., 2014; Schwier et al., 2015). Sea spray aerosols over the Mediterranean mainly come from the Mediterranean itself with little contribution from the Atlantic Ocean. Therefore, the net contribution of P from sea spray is considered negligible in our simulations.

Active volcanoes around the Mediterranean such as the Etna or the Stromboli are another potential source for aerosols. Phosphorus mass in volcanic aerosols is very low (P. Allard, pers. comm.) although it is considered to be almost entirely soluble (Mahowald et al., 2008). Finally, the 85 million hectares of forest around the Mediterranean are a potential source of biogenic particles such as pollen and vegetal debris (Minero et al., 1998) that contain phosphorus. The total mass flux of phosphorus from biogenic particles seems to be important on the global atmospheric phosphorus budget (Wang et al., 2015a). It is not included in our study, which can be seen as a potential limitation. However, biogenic particles have very low solubility in seawater (Mahowald et al., 2008). The LMDz–INCA model provides the summed bulk deposition of both phosphorus from volcanoes and biogenic particles (named PBAP). We chose to discard PBAP as a source of P since these 2 contrasted sources have very different solubilities but cannot be apportioned within PBAP.

2.3 Simulation set–up

We ran NEMOMED12/PISCES for one year with the 2005 physical and biogeochemical forcings. Initial conditions at the end of 2004 are taken from the 1997–2012 simulation described in Richon et al. (2017) including anthropogenic nitrogen deposition ("N" simulation). The reference simulation (REF) is a simulation performed with no atmospheric deposition of phosphate as described in Richon et al. (2017).

We investigate the impacts of each source of PO_4 by performing two different simulations: ”PDUST” and ”PCOMB”; they include, respectively, natural dust only and combustion–generated aerosol only as atmospheric sources of PO_4. We also performed a "Total P" simulation with the two sources included. From now on, we use "total P" to indicate the sum of bioavailable phosphate from dust and combustion ($Pdust + Pcomb$). The results presented in this study are based on the relative differences between the simulations. For instance, the impacts of *Pdust* are calculated as the difference between PDUST and REF simulations (PDUST-REF).

3 Results

3.1 Evaluation of P deposition fluxes

Very few measurements of atmospheric phosphorus deposition exist over the Mediterranean region. Moreover, it is difficult to apportion between different sources when analyzing bulk deposition samples. We did not find any available time series of total phosphorus deposition in the Mediterranean covering our simulation period. Therefore, we compare the monthly P deposition flux from LMDz–INCA with the non time–consistent monthly fluxes over years as close as possible to 2005. Estimates of Turquety et al. (2014) indicate that 2005 is not an exceptional year for fires and the time series of natural dust deposition modeled with LMDz–INCA indicate that the deposition flux of 2005 is close to the inter–annual average (not shown). We used the ~~times series of total P~~ time series of phosphorus measured at 9 different stations over the Mediterranean from the ADIOS campaign (Guieu et al., 2010) and the soluble ~~P measured~~ phosphate from deposition measurements at 2 stations in the South of France from the MOOSE campaign (de Fommervault et al., 2015). The ADIOS time series cover June 2001 to May 2002 and the sampling sites cover almost all regions of the basin. The MOOSE time series cover 2007 to 2012. We use the time series of average monthly flux in 10^{-6} g PO_4 m^{-2} month^{-1} and compare it with our model average monthly fluxes in the grid cells corresponding to the stations. Figure 2 shows the comparison between modeled and measured fluxes in terms of geometric means and standard deviations of monthly values of each time series. The fluxes are highly variable according to the station and the season (variability spans over several orders of magnitudes). Our comparison must be taken with caution since we compare different years in the model and the observations.

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 2. The dust fluxes produced by the model ~~are realistic. We observe a~~ at several stations are realistic, in spite of the low spatial variability ~~in~~ from the dust fluxes produced by the global model LMDz–INCA even though the geometric standard deviations of the fluxes can be regionally very high. ~~We relate this underestimation of dust fluxes and the low spatial variability to the low resolution of LMDz. Bouet et al. (2012) show that the total emission of dust~~ In Table 2, we present 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 ~~spatial~~ resolution. The coarse resolution of LMDz may significantly reduce the total dust emission in the model but also reduce surface winds and aerosol transport (see Discussion section hereafter). Therefore, the coarse resolution of the dust model used in Table 2 for 2001–2002 may explain the underestimation and the lack of spatial variability from the model. We also noted a better agreement (within a factor of 2) at the 4 stations of the eastern Mediterranean (Cyprus, Greek Islands and Turkey).

In order to assess properly the performance of the atmospheric model in reproducing deposition fluxes, we would need continuous times series of deposition in different stations over the Mediterranean and simulations covering the measurement periods. Our comparison is at the moment the most feasible evaluation with the existing data over the Mediterranean. This diagnostic reveals that the model probably tends to underestimate the P deposition from both dust and anthropogenic sources~~, but that the model seems to produce realistic fluxes~~. These results are consistent with Wang et al. (2015a). 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.

3.2 Characterization of phosphate deposition from the different sources

The 2005 seasonal spatial distribution of *Pdust* deposition is shown in Figure 3. *Pdust* deposition is highly variable in space and time. It is maximal in spring (MAM). In this season~~the main~~, the main dust source is the Sahara and it affects mostly the eastern basin (Moulin et al., 1998). In winter (DJF), the influence of dust from the Middle East is observed (Basart et al., 2012). In summer (JJA) and autumn (SON), the deposition is at its minimum and located close to the southern Ionian coasts. Average deposition flux over the basin is 0.122 10^9 g PO_4 month^{-1} with notable monthly variability (standard deviation = 0.102 10^9 g PO_4 month^{-1}). This seasonal cycle of dust deposition is similar to the one simulated by the regional model ALADIN–Climat (Nabat et al., 2012) used in Richon et al. (2017) but LMDz *Pdust* deposition flux is significantly lower than that from ALADIN (see Discussion section).

The seasonal spatial distribution of *Pcomb* deposition is shown in Figure 4. Atmospheric deposition of phosphate from combustion is on average 0.258 10^9 g PO_4 month^{-1} over the entire basin. It amounts twice the atmospheric deposition of phosphate from desert dust (0.122 10^9 g PO_4 month^{-1}). The seasonal variability of *Pcomb* deposition is lower ~~that~~ than for *Pdust* (standard deviation of *Pcomb* = 0.046 10^9 g PO_4 month^{-1}). This is linked to the anthropogenic nature of *Pcomb* emissions and the low contribution of atmospheric transport to seasonal variability. Maximal deposition occurs in summer, likely due to the forest fires around the Mediterranean. In particular, we observe higher deposition close to the Algerian, Spanish and Italian coasts in summer. These countries are particularly subject to dry and hot summer conditions that favor forest fires (Turquety et al., 2014). We observe a high spatial variability in the deposition field with a North–to–South decreasing gradient in deposition, the major part of total mass being deposited close to the coasts, especially in the Aegean Sea. The presence of many industrial areas around the Adriatic and Aegean explains the high deposition fluxes observed in these regions. In the Aegean Sea, *Pcomb* deposition constitutes a more than 4 times greater phosphate source than desert dust (respectively 0.0529 10^9 g PO_4 month^{-1} and 0.0118 10^9 g PO_4 month^{-1} for *Pcomb* and *Pdust* average deposition). ~~However, riverine inputs are the dominant external source of phosphate for almost all Mediterranean regions~~ According to our model forcings, for which uncertainties are still large, the riverine inputs would constitute the main phosphate source to the Mediterranean Basin (3.16 10^9 g PO_4 month^{-1} at the basin scale~~, see also~~). These inputs alone account for over 85 % of the total (atmospheric + riverine input + Gibraltar Strait) as documented in Table 3~~)~~.

~~We show the map for the month of June~~ Figure 5a illustrates that combustion is the dominant source of atmospheric phosphate to the Mediterranean basin for the year 2005. This map shows the average proportion of *Pdust* in total phosphorus deposition (*Pdust*+*Pcomb*). The results indicate that *Pdust* accounts for 30 % of phosphorus deposition on average in 2005 ~~as an example of contrasted contribution of the respective fluxes in~~ at the basin scale, and is only dominant along the southern Mediterranean coast, in the Gulf of Libya. This map highlights the contrasted areas influenced by the different atmospheric P sources. The North of the basin is primarily under the influence of combustion aerosol sources, and the South of the basin is under the influence of dust aerosol sources. Evaluation of deposition fluxes for the period 1997–2012 (available at a too coarse time resolution to perform oceanic simulations) shows a continuous dominance of *Pcomb* fluxes at the basin scale (see Figure 5b). These results agree with the ones of Desboeufs et al. (in prep) who noted that combustion aerosols are responsible for 85 % of P deposition in the northwestern coast of Corsica over a three years period (2008–2011, versus 15 % of P from dust).

Figure 6 ~~.~~ shows the contrasted contribution of the respective fluxes for the month of June 2005. Our previous study showed that June is the period of most ~~significant~~ important impacts from aerosol deposition on surface marine productivity in spite of the low fluxes, due to thermal stratification (Richon et al., 2017). The relative contribution of *Pdust* and *Pcomb* deposition fluxes are compared over three regions~~for the month of June, the~~: the North Adriatic, the South Adriatic and the South Ionian (See Figure 3). We ~~took the definitions for these regions~~ defined these regions as in Manca et al. (2004). ~~These regions were selected because~~ They were selected as they highlight three contrasted conditions. The North Adriatic is under strong influence of both riverine inputs and atmospheric deposition of P from combustion (Figure ~~4~~6), the South Adriatic encompasses atmospheric coastal deposition but is distant from major riverine inputs, and the South Ionian is a deep, highly oligotrophic area. The deposition flux of *Pcomb* is maximal in the northern Adriatic. In this basin, *Pcomb* flux is five times higher than *Pdust*. However, *Pdust* deposition flux increases towards the South to reach a value three times higher than *Pcomb* flux upon reaching the southern Ionian coasts. This spatial distribution of deposition is also found by Myriokefalitakis et al. (2016).

By including ~~the~~ different sources of atmospheric phosphate ~~separately~~from the same model, we can compare the relative contribution of each atmospheric source with the other external nutrient suppliers (rivers and Gibraltar). Table 3 shows the relative contribution ~~of atmospheric P sources to other external fluxes~~ simulated by the model of atmospheric phosphate sources in total external phosphate supply to the Mediterranean basins. Our estimations of total aerosol contribution to PO_4 supply are slightly lower than the literature values. This Table shows that~~the main atmospheric source of~~, in the model, *Pcomb* is dominant over *Pdust* as a source of atmospheric phosphate at the basin scale ~~is *Pcomb*.~~ for the year 2005. This dominance is found in all regions of the Mediterranean, except in the Ionian Sea where *Pdust* and *Pcomb* contributions are equivalent. We note that the estimates from Krom et al. (2010) were calculated by extrapolating to ~~a~~ the eastern basin measurements from very few locations in Turkey and Greece. Vincent et al. (2016) report that recent desert dust deposition fluxes have decreased in the 2010s by an order of magnitude compared to the 1980s that Krom et al. (2010) refer in part to. This may explain that we find combustion to be a more important source of atmospheric phosphate at the basin scale in 2005 in comparison to natural dust. In the pelagic Ionian basin, *Pdust* and *Pcomb* contributions are comparable on a yearly average (20 %). However, combustion represents at most a third of the contribution whereas dust–derived phosphate deposition is more seasonally variable and can be the major source of PO_4 for this basin during spring (contribution of *Pdust* to PO_4 supply up to 60 %).

3.3 Impacts of ~~P~~ atmospheric deposition on marine surface phosphate budgets

Atmospheric ~~phosphorus deposition~~ deposition of phosphate aerosol has different impacts in the model on PO_4 concentration depending on the source, the location, and the period of the year. The impacts of deposition depend on the flux and the underlying biogeochemical conditions in the water column. Even though the deposition fluxes are very low during the stratified season, ~~the relative impacts of deposition~~ their relative impacts are maximal because the major part of the Mediterranean is highly limited in nutrients (Richon et al., 2017).

Figure 7 shows the relative impacts of ~~phosphorus~~ phosphate deposition from the two sources (combustion and dust) on surface PO_4 concentration for the month of June ~~. The relative impacts of atmospheric deposition from different sources are varying over time and depend on both the underlying phosphate concentration and the bioavailable phosphate deposition flux. The relative biogeochemical impacts of PO_4 deposition are variable due to the biogeochemical state of the region.~~

2005. We can distinguish 3 different responses to nutrient deposition: two non responsive zones that are either not nutrient limited or limited in more than one nutrient and a responsive zone limited in the deposited nutrient. In the regions under riverine input influence such as the North Adriatic, relative impacts of atmospheric deposition are low even though the fluxes of *Pcomb* are maximal because the Pô river delivers high amounts of nutrients in this area. In very unproductive regions such as the South Ionian basin, we observe very low impacts of deposition on PO_4 concentrations (between 5 and 12 % enhancement close to the Libyan coast). This basin is highly depleted in nutrients, especially in summer. But the deposition fluxes are very low (90 10^{-6} mol m^{-2} of total PO_4). This low fluxes of nutrients are probably consumed very fast and do not yield a strong concentration enhancement. Finally, some areas respond strongly to ~~P~~ phosphate deposition. We observe PO_4 surface enhancement over 40 % in the South Adriatic, Tyrrhenian and North Aegean basins. These regions are under some nutrient sources influence; they are not fully pelagic and receive nutrients from coasts or upwelling (Sicily Strait front). The high response to phosphate deposition indicates that these regions are primarily P–depleted.

These contrasted results indicate that the relative impacts of atmospheric deposition from different sources are dependent on both the underlying phosphate concentration and the bioavailable phosphate deposition flux. The relative biogeochemical impacts of PO_4 deposition are variable due to the biogeochemical state of the region.

3.4 Biogeochemical impacts of P deposition

In the PISCES model, atmospheric deposition of nutrient is treated as an external forcing. The effects of the different aerosols on the Mediterranean biogeochemistry are considered simply additive. Fluxes of nutrients are added to the total pool of dissolved nutrients according to their deposition flux and chemical properties (fixed solubility and chemical composition). The effects of total ~~atmospheric phosphate~~ P deposition on marine biogeochemistry are a combination of effects of the two P sources in this model version (Table 3).

We focus here on the month of June that shows maximum impacts of deposition because of surface water stratification. Figure 8 shows the average relative effects of P deposition on surface chlorophyll *a*. The relative effects of total P deposition on surface chlorophyll *a* concentration are modest. The majority of *Pdust* effects on surface chlorophyll *a* are in the southwestern basin along the Algerian coasts. *Pcomb* has maximal impacts in the North of the basin, in areas of high deposition. However, *Pcomb* also affects the area influenced by *Pdust* in the South. In this Redfieldian version of PISCES, chlorophyll production is linked with nutrient uptake that is constrained by the Redfield ratio. Therefore, the addition of excess nutrient will enhance chlorophyll production as long as other nutrients are bioavailable in the Redfield proportions.

Figure 9 shows the relative impacts of ~~P~~ phosphate deposition from the two sources on surface total primary productivity for the month of June. We observe that combustion–~~derived~~-derived phosphate has the greatest impacts on surface biological production: averaged regionally over the framed areas, the enhancement in daily primary productivity is between 1 and 10 % but local maxima are up to 30 %.

The effects of atmospheric phosphate deposition are variable according to the source type. As for chlorophyll *a*, dust–derived phosphate deposition has maximal impacts in the southern part of the basin close to the Algerian and Tunisian coasts. The relative impacts of *Pdust* deposition in the South Ionian basin are very low (about 1.7 %). This region of the Mediterranean is highly oligotrophic and lacks all major nutrients, especially in summer. ~~Moreover,~~ Nutrient co–limitations associated to minimal *Pdust* deposition flux ~~is minimal in summer . This explains~~ in summer explain the weak impacts of ~~P~~ phosphate deposition in this area.

Pcomb deposition has maximal impacts in the North of the basin in the Adriatic and Aegean Seas. In the northern Adriatic, the relative impacts of *Pcomb* deposition are lower than in the southern Adriatic because the proximity of riverine inputs in the North reduces the relative importance of atmospheric deposition in nutrient supply. The North of the Adriatic is generally productive all year long. We can identify in Figure 9 the area in the Adriatic influenced by riverine inputs. This zone encompasses the North of the Adriatic and the western coast down to the region of Bari (40°N, 17°E). In this area, atmospheric deposition has low influence on primary productivity (below 15 %). This is in contrast with the southeastern part of the Adriatic under low riverine input influence. There, we observe high impacts of atmospheric deposition and especially of *Pcomb* on primary productivity (11 % on average but up to 50 % daily enhancement at some points).

Pcomb and *Pdust* have similar influences over the South of the basin. ~~We performed a Student's t–test on the grid matrix of relative impacts of *Pdust* and *Pcomb* over the three regions from Manca et al. (2004) and found that the mean values are statistically different (p–value < 0.01). This shows that even though the impacts of *Pdust* are close to the effects of *Pcomb* in the South Ionian, they are significantly dominant~~

In general, we can identify 3 different biogeochemical responses in the 3 areas indicated in Figure 9. Our hypothesis is that the different responses are linked to nutrient limitations. In the North Adriatic, the influence of coastal nutrient inputs leads to low nutrient limitation and high productivity. In the South Adriatic, the high impact of atmospheric phosphate deposition may be the sign of important phosphate limitation. Finally, the lack of response in South Ionian in spite of the atmospheric phosphate deposition probably indicates that the region is co–limited in P and N.

As for the effects on PO_4 concentration, we observe different impacts of P deposition on primary production according to the nutrient status of the region. We find very low deposition impacts in nutrient repleted areas (e.g. North Adriatic), very low to no response in highly nutrient limited areas such as the South Ionian, and high response in areas limited by phosphorus only (e.g. South Adriatic).

4 Discussion

In contrast to the global ocean, combustion appears as an important source of atmospheric bioavailable phosphorus to the surface waters of the Mediterranean Sea due to the proximity of populated and forested areas. Based on our large scale LMDz–INCA model, we estimate that combustion is responsible for 7 % on average of total PO_4 supply. In comparison, the average contribution of *Pdust* to PO$_4$ supply is 4 % (Table 3). These estimates are based on our modeling values and take into account only the sources of phosphate that are included in the simulations (namely rivers, Atlantic inputs, natural and combustion derived atmospheric phosphate). This provides an estimation of the relative importance of the 2 atmospheric sources under the specific conditions of the year 2005, but the restriction to only this particular year limits our conclusions. For this reason, 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. Saharan dust is a major source of particles in the Mediterranean (D'Almedia, 1986; Loÿe-Pilot et al., 1986) and does have an impact on the regional climate system (Nabat et al., 2012, 2015). The literature on Mediterranean aerosols is often centered on Saharan dust deposition, which is believed to have the largest impact on the basin's biogeochemistry (e.g. Bergametti et al., 1992; Migon and This study provides the first Mediterranean assessment of the contribution of another source of atmospheric phosphate than dust. It highlights that other sources, namely combustion, might be a dominant source of bioavailable phosphorus to Mediterranean surface waters.

The relative dominance of combustion over dust as a source of phosphate for the Mediterranean is confirmed by the analysis of monthly modeled deposition fluxes of *Pdust* and *Pcomb* over the 1997–2012 period (see Figure 5). *Pcomb* dominates *Pdust* contribution to PO$_4$ supply over the northern basin (Adriatic and Aegean Seas in particular). For these regions in the vicinity of anthropogenic sources, *Pcomb* deposition has a low variability whereas *Pdust* deposition occurs during transient events and is therefore highly variable on a monthly basis. This was already noticed by Bergametti et al. (1989) and Gkikas et al. (2016) who describe the majority of dust as occurring in a few episodic deposition events, whereas anthropogenic aerosols have a more constant flux. These results are also coherent with Rea et al. (2015) who estimate anthropogenic emissions to be the main component of PM$_{2.5}$ and dust to be the main component of PM$_{10}$ over the Mediterranean. The maximal contribution of atmospheric deposition to PO$_4$ budgets is observed in spring, when the deposition fluxes are maximal. In summer, the relative contribution in each sub–basin is very small because the flux of *Pdust* is very low. The high, nearly constant fluxes of *Pcomb* deposited close to the coasts, especially in semi—closed sub—basins such as the Adriatic and Aegean, constitute the major source of soluble atmospheric phosphate to the surface of the Mediterranean Sea. Although total mass deposition of phosphorus from desert dust exceeds that of combustion aerosols, the latter are much more soluble than lithogenic dust. This explains in our results the yearly predominance of *Pcomb* as a source of bioavailable phosphate. However, the underestimation of deposition fluxes indicated by Figure 2 limits the conclusions we can draw from our results on the relative contributions of the different phosphate external sources. More measurements and developments of the atmospheric model must be undertaken to make more precise assessments of the importance of atmospheric deposition as a source of nutrients for the oligotrophic Mediterranean.

The LMDz–INCA model version used in this study integrates constant emissions of *Pcomb* from anthropogenic sources. The variability of this deposition flux is only due to variability of atmospheric transport and deposition processes such as winds and rain or dry sedimentation. The atmospheric model LMDz–INCA has a low resolution given the regional Mediterranean scale: *Pdust* deposition forcing has 280x193 grid points globally and $\sim$500 grid points covering the Mediterranean, and *Pcomb* forcing has 144x143 grid points in total and $\sim$200 grid points covering the Mediterranean. These forcings reproduce ~~well the average~~ realistic deposition patterns at the ~~basin scale~~global scale, in spite of generally underestimating the measured fluxes (Wang et al., 2017) but may not be reliable when analyzing small scale deposition patterns. There is to our knowledge no regional model Mediterranean model available that represents phosphorus deposition from both natural and anthropogenic sources. Investigating these atmospheric deposition fluxes from a higher resolution regional model is a perspective to consider in order to strenghen our conclusions on the spatial distribution of *Pdust* and *Pcomb* influences.

Concerning the dust deposition component for which products from high resolution model exist (see the high resolution model ALADIN–Climat used in Richon et al., 2017), ~~tshe~~ the overall average deposition estimation from the global model we use in this study appears much lower ($0.122 \pm 0.102\ 10^9$ g month^{-1} over the Mediterranean in 2005 simulated with LMDz-–INCA and $0.568 \pm 0.322\ 10^9$ g month^{-1} simulated with ALADIN-–Climat). Table 3 in Richon et al. (2017) shows the same comparison between measured dust fluxes and the dust fluxes from the ALADIN–Climate regional model than in Table 2. The fluxes reproduced by this 1/12° resolution regional model are generally closer to the measurements. The coarse resolution of LMDz may lead to a global underestimation of the dust emission fluxes, as shown by Bouet et al. (2012). Moreover, the higher spatial resolution of ALADIN-–Climat allows one to better reproduce intense regional winds (Lebeaupin Brossier et al., 2011) that can favor transport of continental aerosols to the remote sea. 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. However, dust deposition fluxes available between 1997 and 2012 from the LMDz–INCA model indicate that 2005 is not an exceptional year (see also Figure 5). Similarly, the inter–annual time series of dust deposition analyzed in Richon et al. (2017) showed that 2005 is also not an exceptional year in the ALADIN–Climat model.

The recent estimate of burnt areas in the Euro–Mediterranean countries over 2003–2011 by Turquety et al. (2014) indicates a ± 50 % annual variability, but it is impossible to ~~give any inter–annual variability of~~ separate anthropogenic and wildfires in *Pcomb* deposition at present. Simulating ~~longer time periods of atmospheric deposition~~ separately atmospheric deposition from anthropogenic and wildfires would give interesting perspectives on ~~the evolution of anthropogenic~~ combustion aerosol deposition.

The reproduction of small scale atmospheric patterns such as coastal breezes that can transport aerosols far from the coasts above the marine atmospheric boundary layer is also limited at the low spatial resolution of LMDz (Ethé et al., 2002; Lebeaupin Brossier et al., 2011). This leads to low day–to–day variability in total *Pcomb* deposition flux together with much larger modeled fluxes in coastal areas. *Pcomb* deposition is limited in the model to coastal areas. However, our results indicate that *Pcomb* is dominant over *Pdust* in this instance as an atmospheric source of phosphate at the basin scale. Moreover, the atmospheric deposition model seems to underestimate phosphate deposition in most of the stations we found (see Figure 2). Constant emissions of phosphate from anthropogenic combustion is, however, a satisfying first approach because it permits to highlight the high concentration contributed from industries and major urban centers around the Mediterranean. However more refined emission scenarios would be interesting to consider in future modeling studies.

Some areas receive phosphate with different contributions from different sources (Figures 3, 4). In particular, islands in the Eastern basin such as the Greek Islands, Crete and Cyprus receive phosphate from the two sources, sometimes in a single deposition event (Koulouri et al., 2008; D'Alessandro et al., 2013). Atmospheric processing of different aerosols will alter the nutrient composition and solubility of this deposition (Migon and Sandroni, 1999; Desboeufs et al., 2001; Anderson et al., 2010; Nenes et al., 2011; D'Alessandro et al., 2013). However our study does not account for such mixing.

The atmospheric model used in our study does not provide biogenic and volcanic phosphorus deposition separately. The model of Myriokefalitakis et al. (2016) allows to represent a more complex atmospheric chemistry. This work showed that many different atmospheric P sources exist. In particular, they estimate 0.195 and 0.006 TgP year^{-1} of global emissions from biogenic and volcanic sources respectively. ~~However, most of the biogenic phosphorus is under~~ In the Mediterranean region that is surrounded by many forested areas, biogenic emissions may be an important source of atmospheric phosphorus in the form of organic ~~phosphorus (DOP) that our model version~~ matter. Moreover, Kanakidou et al. (2012) show that an important fraction of organic phosphorus can be emitted from combustion. In particular, the numerous forest fires occurring every summer in the Mediterranean region may constitute an important source of organic phosphorus. However, the PISCES version used in this study does not include ~~. Moreover, the composition of biogenic aerosols and its solubility is still poorly constrained.~~ organic phosphorus. In the ocean, organic phosphorus can be recycled by bacterial activity into inorganic phosphate that is bioavailable for plankton growth. Therefore, the inclusion of organic phosphorus in PISCES along with an estimation of organic phosphorus from atmospheric fluxes is a perspective to consider.

The PISCES version used in this study is based on the Redfield hypothesis that C:N:P ratios in organic cells are fixed. This fixed value determines the nutrient ratio for uptake and has the advantage of simplifying calculations in the 3–D high resolution coupled model and is supported by some observations (Pujo-Pay et al., 2011). However, because the Mediterranean is highly oligotrophic, this Refieldian hypothesis is questioned and the biogeochemical cycles may be determined by non–Redfieldian nutrient use. This non–Redfieldian behavior may imply complex nutrient limitations and co–limitations processes (Geider and La Roche, 2002) that can not be studied with the present PISCES version. As of today, there is no version of PISCES that includes the non–Redfieldian biogeochemistry in the Mediterranean. The development and use of such a version of PISCES is a perspective of this work that may help to fully understand nutrient dynamics and growth limitation process in the Mediterranean (Saito et al., 2008; Krom et al., 2010). However, this study provides interesting first results on the potential impacts of phosphate atmospheric deposition on the Mediterranean nutrient pool and potential implications on biological productivity. Moreover, the development and qualification of a non–Redfieldian version of the PISCES model may take several years. Plus, even if non–Redfieldian regional Mediterranean biogeochemical models such as ECO3M exist (Baklouti et al., 2006), their higher complexity leads also to a hard task, since the sensitivity of such models to parameter values is a delicate question that requires important computing time and data to solve before revisiting our conclusions.

5 Conclusions

This study is a first approach to quantify the effects of different atmospheric sources of phosphorus to the Mediterranean Sea surface. Our results indicate that contrary to the global ocean, combustion may be dominant over natural dust as an atmospheric source of phosphate for the Mediterranean Basin. This study is the first to examine separately the effects of atmospheric deposition of phosphate from different sources that have different seasonal cycles and deposition patterns over the Mediterranean Sea. According to our low resolution atmospheric model, phosphate deposition from combustion (which includes forest fires and anthropogenic activities) is mainly located close to the coasts and has low variability whereas phosphate deposition from dust is episodic and more widespread. The results indicate that combustion sources are dominant in the North of the basin close to the emission sources whereas natural dust deposition is dominant in the South of the basin and is strongly dominant in pelagic areas such as the Middle Ionian and Levantine basins. The study of atmospheric model low resolution deposition fluxes over the period 1997–2012 indicate that the dominance of *Pcomb* over *Pdust* in the Mediterranean basin is consistently observed over this time period. The yearly–averaged deposition patterns are constant over the period. The relative effects of each source are maximal in their areas of maximal deposition and can induce an enhancement of up to 30 % in biological productivity during the period of surface water stratification.

In the coastal Adriatic and Aegean Seas that are under strong influence of anthropogenic emissions, we showed that combustion-derived phosphorus deposition ~~has~~ may have effects on the biological productivity. It seems that only dust transported through large events can reach and fertilize pelagic waters. However, the pelagic zones far from coastal influence are often highly oligotrophic and co–limited in nutrients. Then, the deposition of one type of nutrient cannot relieve all the nutrient limitations to have strong fertilizing effect.

In spite of the limitations of our study linked to the availability of atmospheric P emission and the limited knowledge on atmospheric mixing processes impacts on bioavailability of deposited PO_4, we showed that atmospheric P deposition is an important source of bioavailable nutrients and has low but significant impacts on marine productivity. Combustion and soil dust sources display contrasted deposition patterns. Therefore, none should be neglected when accounting for atmospheric sources of nutrients in land and ocean biogeochemical models.

Our study highlights the difficulty to constrain atmospheric deposition in models because very few estimates of the deposition fluxes over the Mediterranean are available. The existing time series cover only very limited areas of the basin and short time periods. Plus, there is, to our knowledge, ~~no~~ only one experimental study addressing the source apportionment of phosphate deposition. Longo et al. (2014) measured the solubility of P aerosols coming from South and North regions of the Mediterranean and showed that aerosols from Europe deliver more soluble P. Also, Desboeufs et al. (in prep) showed that more than 85 % of of P deposition is brought by combustion aerosols in northern Corsica over the 2008–2011 period. We underline here the need for more deposition measurements in order to better constrain the modeling of such important nutrient sources for the Mediterranean.

Further development of atmospheric and oceanic models should be undertaken in order to account for the mixing and chemical processing of the different aerosol sources in the atmosphere and their effect on nutrient solubility in seawater, and for possible deviations from Redfield ratios in the marine biological compartments. Moreover, oceanic simulations taking into account daily atmospheric deposition of nutrients from dust and combustion over larger time periods would be necessary to assess the variability of the impacts of these sources on marine biogeochemistry.

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Appendix A: ~~Evaluation~~ Statistical evaluation of ~~NEMOMED12/~~PISCES ~~against available observations~~vertical profiles

The ~~main characteristics of the surface chlorophyll a distribution are well simulated (Figure ??). We analyze the results for the month of April typical of the spring bloom period (maximal productivity) and June as an example of the low productivity period. The west–to–east gradient and the main productive regions observed in the satellite estimation (http://www.myocean.eu) are simulated (Strait of Gibraltar, coastal zones and the Gulf of Lions). The spring bloom maximum is reproduced in the model but, the extension of the bloom zone in the Algero–Provençal basin is wider in the observations. The model produces a productive zone in the western basin but the chlorophyll concentrations are too low compared to the observations. We trace this discrepancy back to the mesoscale activity and circulation anomalies in the western basin (Hamon et al., 2015). In June, the Mediterranean is largely non productive, only coastal areas close to river mouth are productive. The model reproduces well chlorophyll a in all these zones, except in the Gulf of Gabes that is probably under the influence of coastal nutrient runoff that are not included in the model . We note an underestimation of about 50 % of chlorophyll estimations in the coastal areas, where satellite estimations are highly uncertain.~~ following Tables and Figures provide an evaluation of the model performances against measurements in different Mediterranean regions obtained from Manca et al. (2004). We provide observed and modeled annually averaged vertical profiles over the year 2005 of phosphate and nitrate and statistical indicators (annual vertical mean and standard deviation, RMSE, normalized and relative bias, Pearson's correlation coefficient R and associated p-value). We point out that the different indicators provide different information. For instance, bias evaluates how mathematically close are modeled and measured values, whereas Pearson's R indicates whether the model reproduces the evolution of PO_4 concentration with depth.

~~We evaluated the large scale~~ These figures show that the model reproduces on average the vertical distribution of ~~nutrients against observations from the BOUM campaign (Moutin et al., 2012)~~ phosphate. In some regions such as the Algerian basin, surface concentration is closer to measurements than deep concentrations (Figure ~~??).~~

~~The NEMO/PISCES model reproduces the vertical structure of the nutrients with a gradient from west to east and a subsurface maximum of concentrations due to Levantine Intermediate Water (LIW). The model, however, underestimates the concentrations in the western basin producing a too smooth nutricline (depth of rapid nutrient change) compared to the observations. This is probably due to the anomalies in SSH in the Algero–Provençal basin (Hamon et al., 2015). Concentrations in the eastern basin are correctly simulated~~A1). In the South Adriatic and in the Gulf of Lions, phosphate concentration below 200 m is close to the measurements (Figures A4 and A5 show normalized bias of -0.09 and 0.0 respectively). On the other hand, very low bias in the South Adriatic region is paired with low Pearson's R (0.30 in the deep layer) whereas PO_4 concentration evolution with depth is well reproduced in the Algerian basin (R=0.89 and R=0.97 in the intermediate and deep layers) in spite of a mismatch between measured and modeled values.

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Basin	**Model mean (σ)**	**Data mean (σ)**	**RMSE**	**normalized bias**	**% bias**
Whole Med.	0.137 (0.04)	0.140 (0.1)	0.06	-0.01	-0.4
West	0.175 (0.03)	0.215 (0.1)	0.08	-0.10	-23
Adriatic	0.155 (0.007)	0.205 (0.04)	0.06	-0.13	-31
Aegean	0.141 (0.02)	0.145 (0.1)	0.10	-0.01	-3
Ionian	0.108 (0.03)	0.094 (0.04)	0.03	0.07	13
Levantine	0.108 (0.2)	0.07 (0.02)	0.04	0.23	38

Table 1. Average chlorophyll *a* concentration (spatial standard deviation in brackets) and statistical indicators (spatial RMSE, normalized and relative bias) for different Mediterranean sub–basins (see Figure 3 for the sub–basins limits). Values are calculated from Figure 1. Coastal areas are filtered out as in Figure 1.

Station	**ADIOS**	**LMDz–INCA (ADIOS period)**	**LMDz–INCA (2005)**
Cap Spartel, Morocco	6.8 (2.7)	2.7 (1.5)	6.3 (4.4)
Cap Béar, France	11 (3.1)	3.4 (4.8)	2.1 (3.8)
Corsica, France	28 (4.6)	3.6 (4.4)	3.1 (3.9)
Mahdia, Tunisia	24 (2.8)	3.7 (1.8)	11.6 (3.3)
Lesbos, Greece	6.0 (2.3)	3.7 (4.1)	18.8 (5.2)
Crete, Greece	9.0 (3.2)	3.3 (2.3)	8.9 (4.1)
Akkuyu, Turkey	10 (3.2)	3.7 (4.0)	14.1 (4.9)
Cavo Greco, Cyprus	4.1 (1.8)	3.6 (3.1)	8.6 (4.3)
Alexandria, Egypt	21 (3.3)	3.4 (2.5)	8.2 (4.1)

Table 2. 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 LMDz–INCA model on the ADIOS period (June 2001 - May 2002) and the simulation period (2005). Values in brackets indicate the geometric standard deviations of monthly fluxes (same restrictions on the number of values as in Figure 2.

Basin	**Total P**	**Pdust**	**Pcomb**	**Ref.**
East	28			Krom *et al.*, 2010
Whole Med.	11 (9-21)	3.6 (1-10)	7.5 (5-11)	This work
West	9 (6-15)	1.7 (0-5)	7.3 (5-11)	This work
Adriatic	6 (4-16)	0.97 (0-5)	5.1	This work
Aegean	11	2.0 (0-5)	9.0 (6-11)	This work
Ionian	40 (27-71)	20 (5-60)	20 (10-33)	This work
Levantine	11 (7-18)	4.3 (1-14)	7 (4-10)	This work

Table 3. Relative atmospheric contribution (%) to total PO_4 supply in different sub–basins of atmospheric sources (atmospheric inputs/(atmospheric inputs + riverine inputs + Gibraltar inputs)) according to the model. The values in parentheses show the minimum and maximum monthly contributions over the year when variability is more than 3 %. The sub basins are described in Figure 3. Values from Krom et al. (2010) also include river inputs.

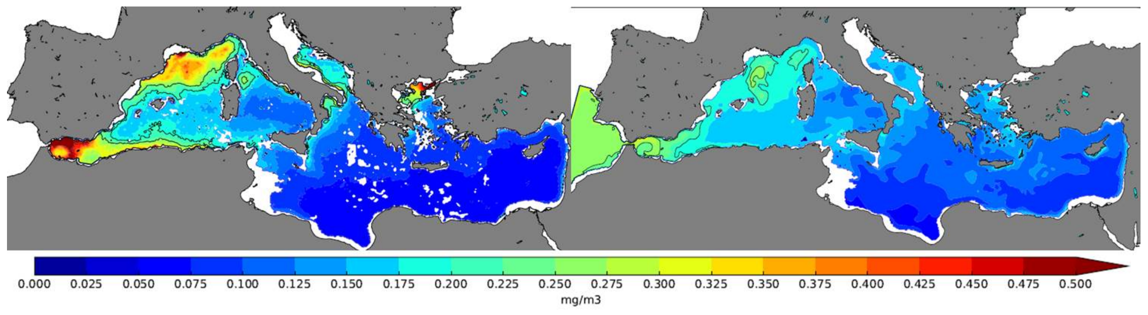


Figure 1. Satellite map of average surface chlorophyll *a* concentration from Bosc et al. (2004) (1997–2004, left) and modeled average surface chlorophyll *a* concentration (right). Model and satellite data are filtered for coastal waters (white areas). Additional white areas on the satellite maps are lack of data.

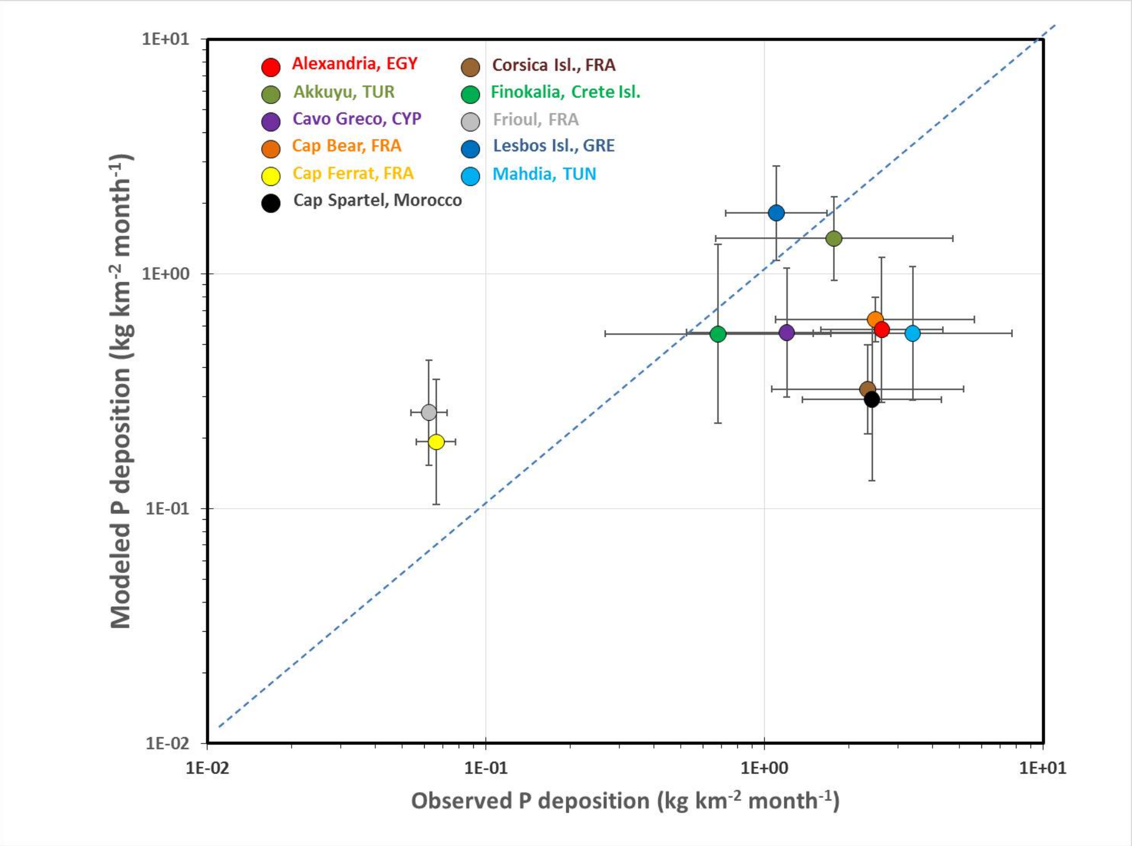


Figure 2. Comparison of modeled and observed monthly geometric mean of total P (*Pdust* + *Pcomb*) deposition fluxes at the 9 ADIOS stations (Guieu et al., 2010) and soluble PO_4 at Frioul and Cap Ferrat stations (de Fommervault et al., 2015). Each point is the geometric mean of monthly observed and modeled values at the given station over 1 year, namely 2005 for the model and June 2001–May 2002 for the ADIOS observations (only 6 values are available at Alexandria to compute the observed mean and standard deviation, 10 at Mahdia, and 11 at Finokalia) and between 2007 and 2012 for the observations at Frioul and Cap Ferrat stations. Error bars represent the geometric standard deviation on model (y—axis) and measurements (x—axis).

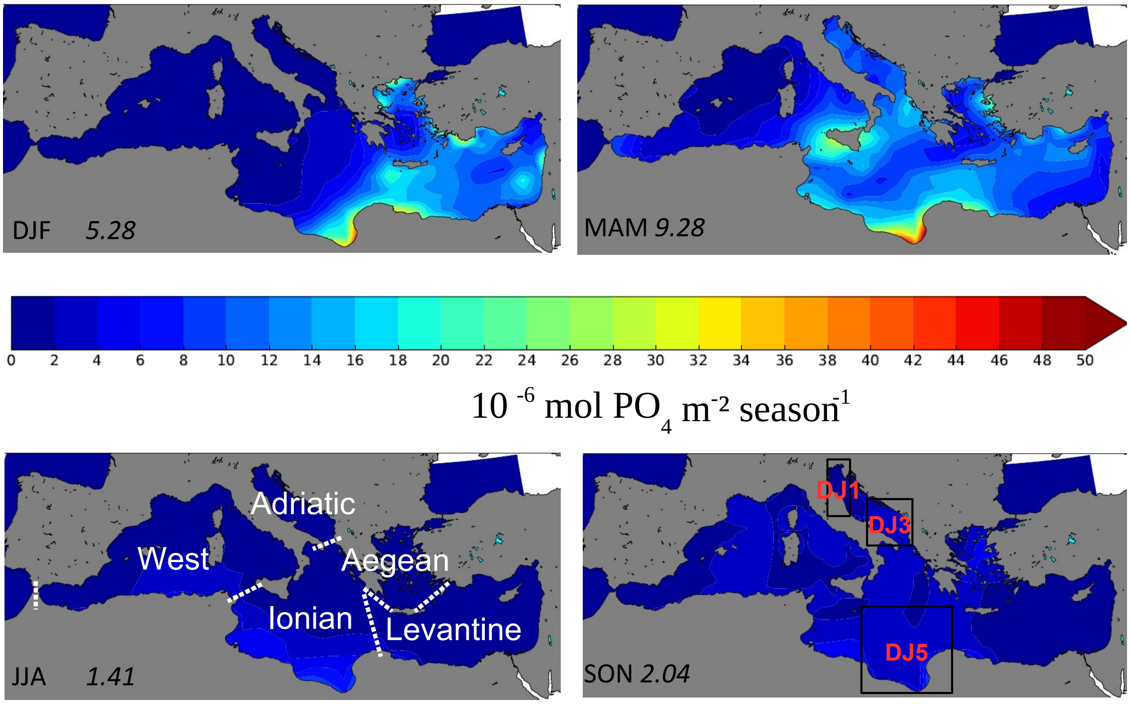


Figure 3. Total seasonal desert dust derived soluble phosphorus deposition (*Pdust*, in 10^{-6} molPO$_4$ m^{-2} season^{-1}) over each season of the year 2005 (molar flux is calculated as mass flux/phosphorus molar weight) from the LMDz–INCA model. Numbers on the maps are the average seasonal deposition fluxes over the whole basin in 10^{-6} molPO$_4$ m^{-2} season^{-1}. In the Summer (JJA) deposition map, we display the different sub regions referred to in the text. In the Automn (SON) map, we display sub regions as defined in Manca et al. (2004): DJ1 is the North Adriatic region, DJ3 is the South Adriatic region and DJ5 is the South Ionian region.

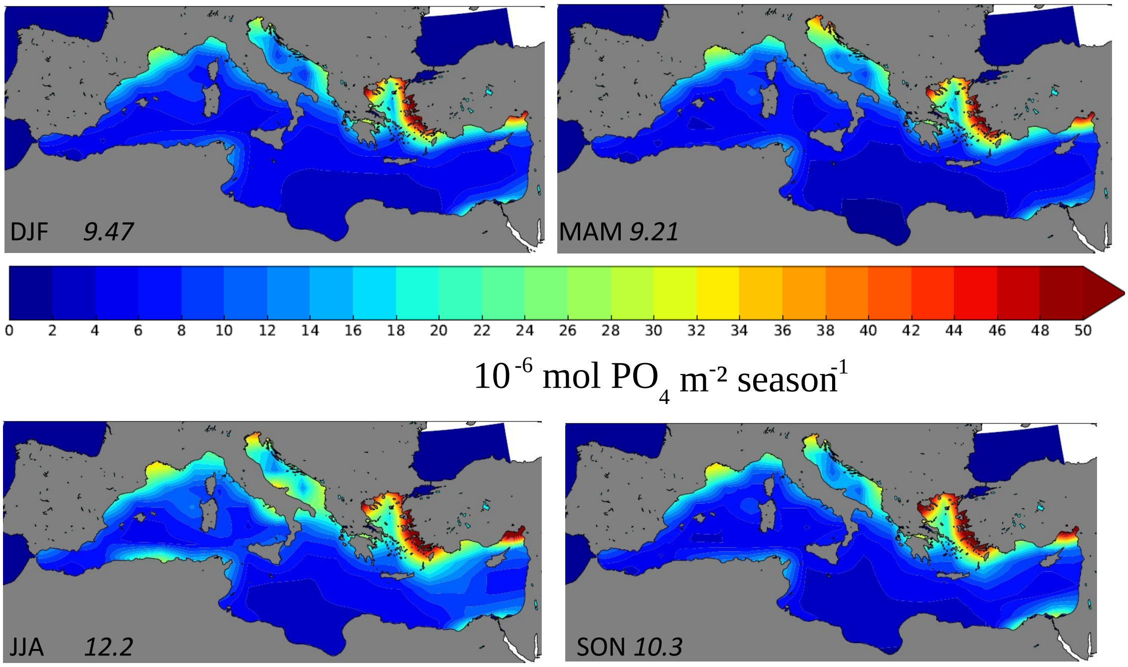
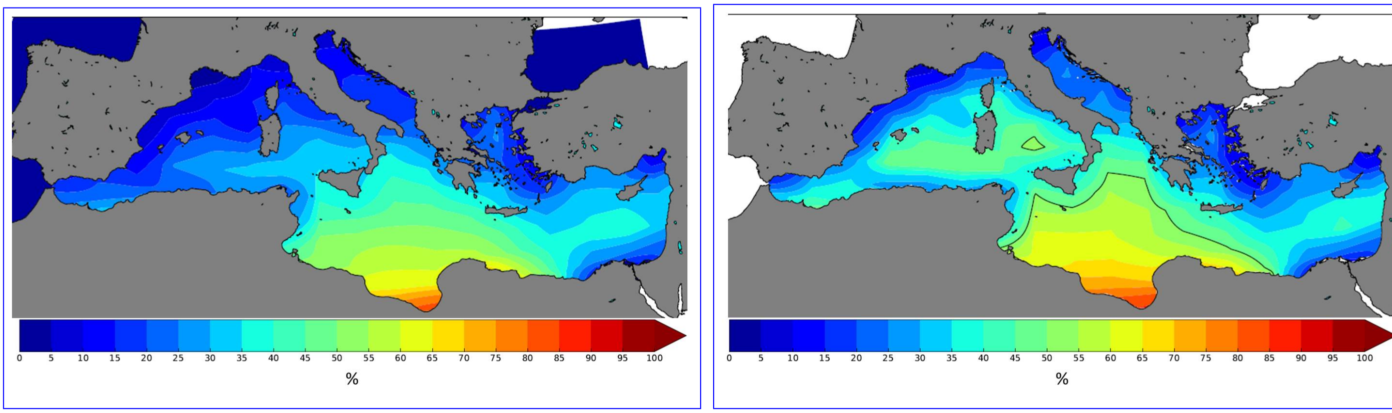


Figure 4. Total seasonal combustion–derived soluble phosphorus deposition (*Pcomb* in 10^{-6} molPO$_4$ m^{-2} season^{-1}) over each season of the year 2005 (molar flux is calculated as mass flux/phosphorus molar weight) from the LMDz–INCA model. Numbers on the maps are the average seasonal deposition fluxes over the basin in 10^{-6} molPO$_4$ m^{-2} season^{-1}.



(a) 2005 average

(b) 1997–2012 average

Figure 5. Map of average *Pdust* proportion in total P deposition for 2005 (left) and 1997–2012 (right). The black line on the right map represents the 50 % *Pdust* proportion limit.

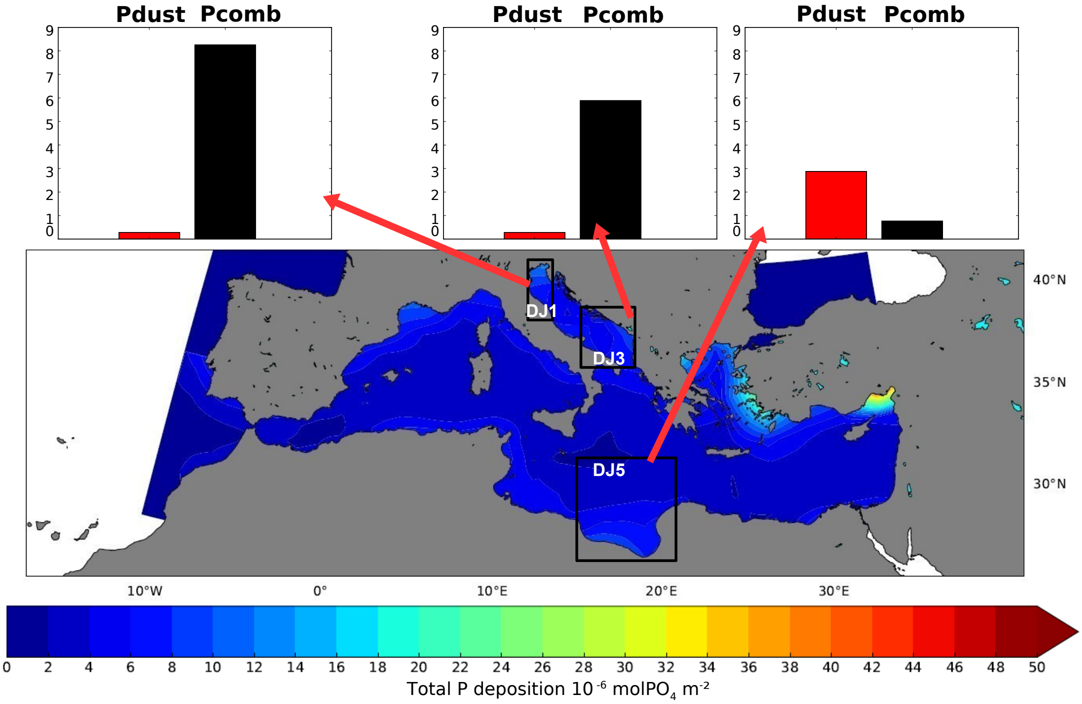


Figure 6. Map of total PO_4 deposition from both *Pdust* and *Pcomb* (10^{-6} molPO$_4$ m^{-2}) for June 2005. ~~Bar plots~~ Red and black bars represent average PO_4 deposition (in 10^{-6} molPO$_4$ m^{-2}) from the two sources in each framed area. The limits of the areas are described in Manca et al. (2004) and Figure 3. There is no atmospheric deposition modeled in the Marmara and Black Seas.

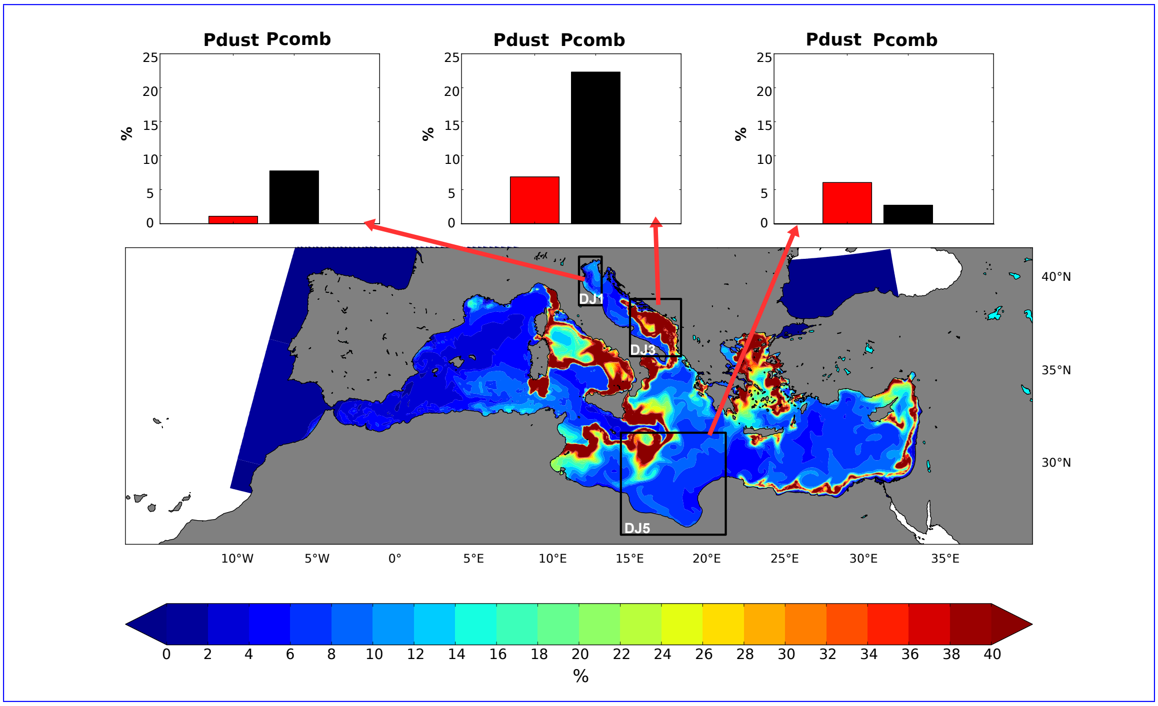


Figure 7. Map of maximal relative effects of total (*Pdust*+*Pcomb*) deposition ~~on PO$_4$ concentration~~ in June 2005 (on a daily basis) on the surface ~~Mediterranean~~ phosphate concentration (0–10 m)~~in June 2005. Bar plots~~. The reference PO$_4$ concentration values are taken from the REF simulation without atmospheric phosphate deposition. Red and black bars represent average ~~maximal~~ relative effects (%) within the framed areas ~~of the two sources~~for each P source. The limits of the areas are described in Manca et al. (2004) and Figure 3. There is no atmospheric deposition modeled in the Marmara and Black Seas.

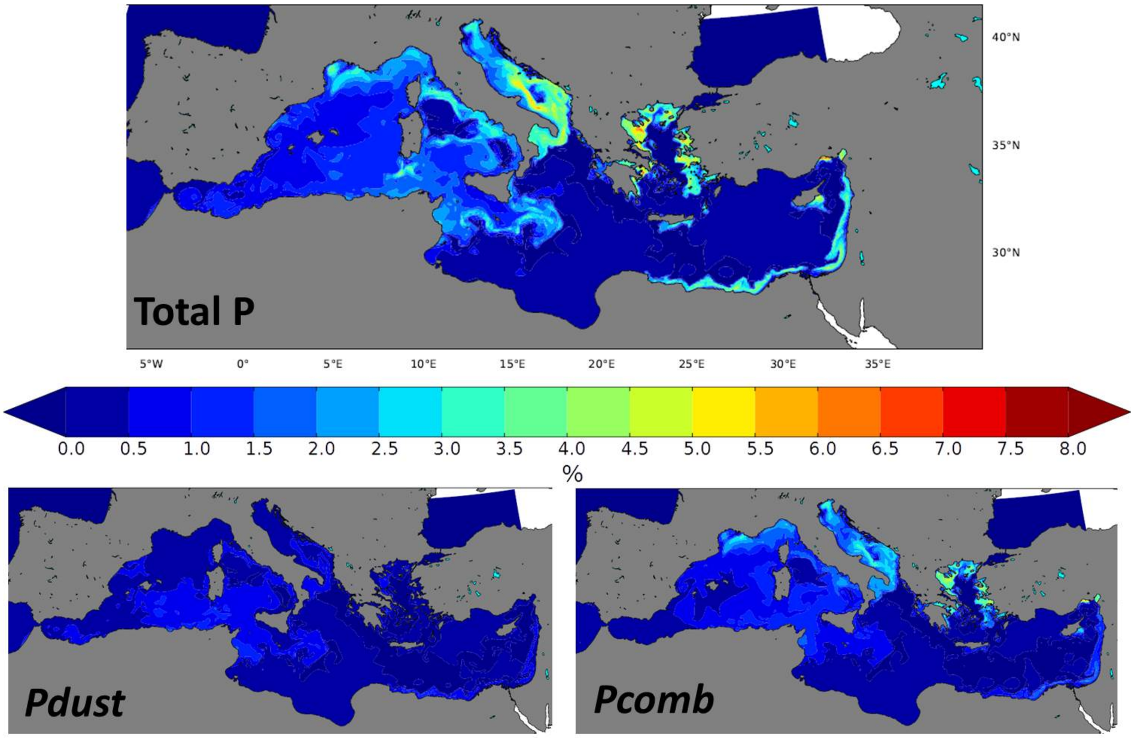


Figure 8. Average relative effects of total P, *Pdust* and *Pcomb* deposition on surface (0–10 m) chlorophyll *a* concentration for June 2005. There is no atmospheric deposition modeled in the Marmara and Black Seas.

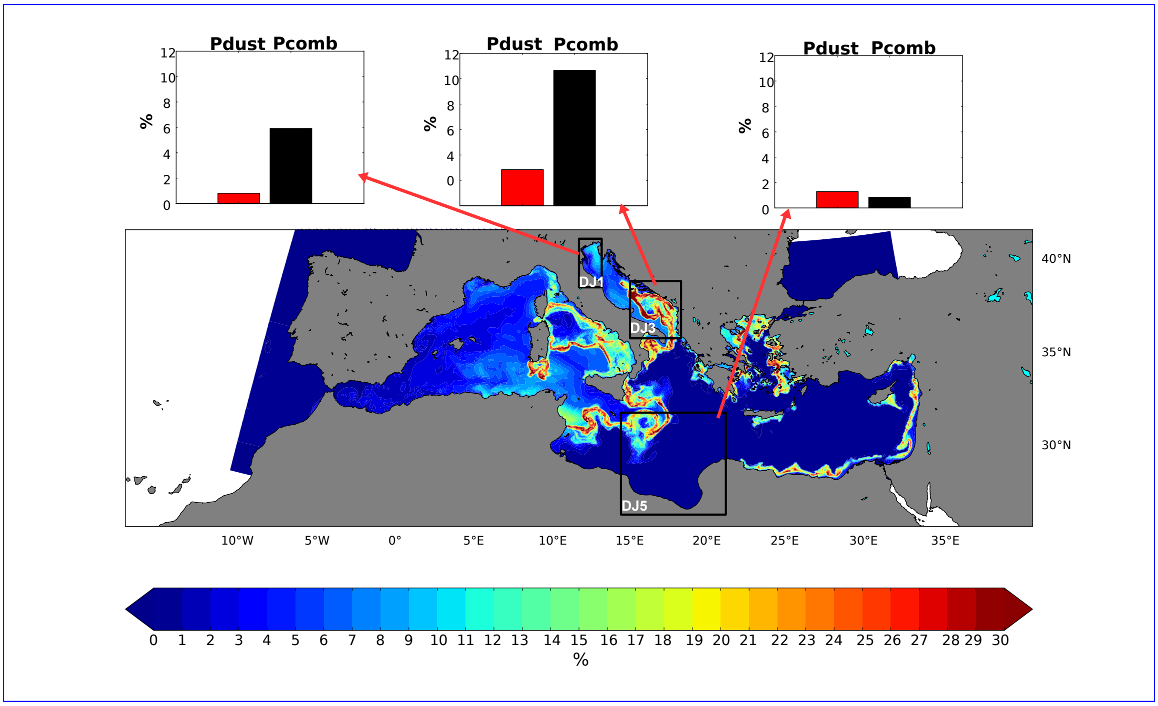
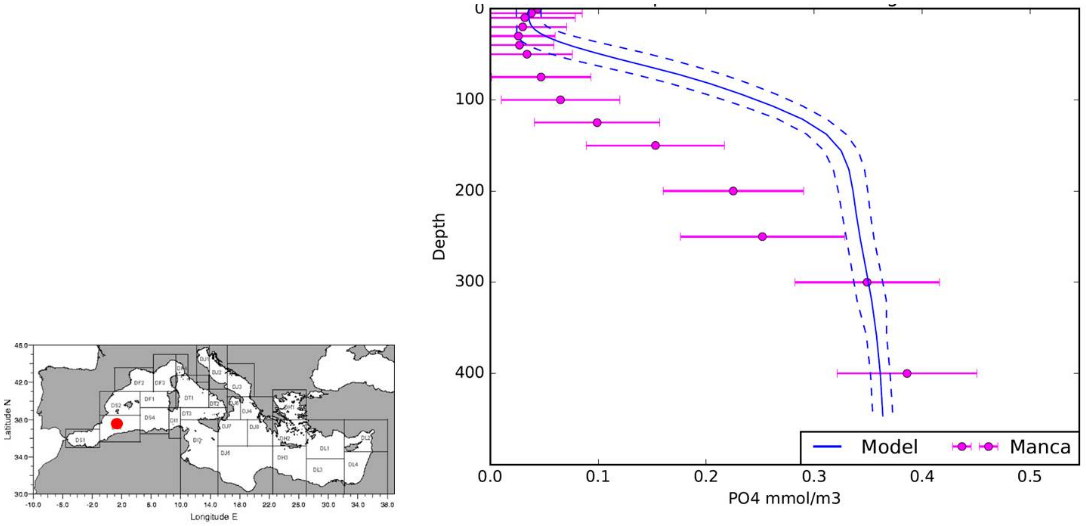
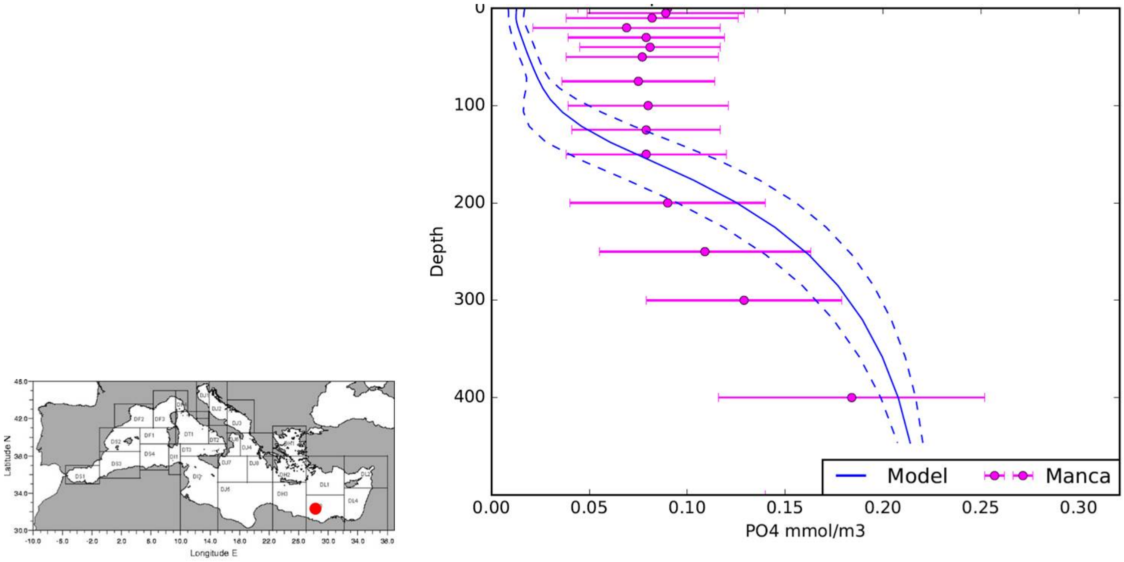


Figure 9. Map of maximal ~~daily~~ relative effects of total (*Pdust*+*Pcomb*) deposition on primary production in the surface Mediterranean (0–10 m) in June ~~2005.~~ 2005 (on a daily basis). The reference PO_4 concentration values are taken from the REF simulation without atmospheric phosphate deposition. Barplots represent average ~~maximal~~ relative effects of each source (%) within the framed areas excluding land. The limits of the areas are described in Manca et al. (2004). There is no atmospheric deposition modeled in the Marmara and Black Seas.



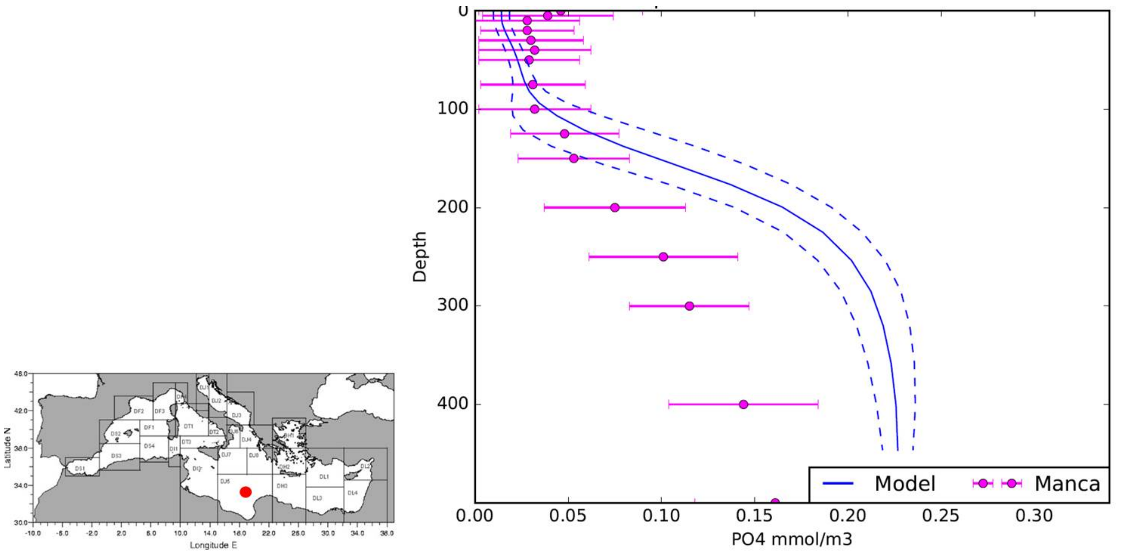
Depth	Model mean	Data mean	Model std	Data std	RMSE	normalized bias	% bias	Pearson's R	P-value
0-50	0.045	0.033	0.014	0.006	0.02	0.16	27.02	-0.64	0.17
50-200	0.23	0.080	0.079	0.043	0.16	0.48	65.27	0.89	0.04
200-500	0.35	0.30	0.009	0.067	0.07	0.07	12.78	0.97	0.03

Figure A1. ~~Evaluation of~~ Annually averaged PO_4 vertical profile in the ~~model NEMOMED12/PISCES. Satellite~~ Algerian sub–basin (~~left)~~ see map and ~~modeled (right~~Manca et al. (2004)) ~~surface chlorophyll *a* averaged~~ and statistical indicators over ~~April (top)~~ different depths. Model values are the blue line (2005 average, Ntot simulation from Richon et al., 2017) and ~~June (bottom) 2005.~~measured values from Manca et al. (2004) are in pink. Horizontal bars and dashed lines indicate spatial standard deviation of observations and model results respectively.



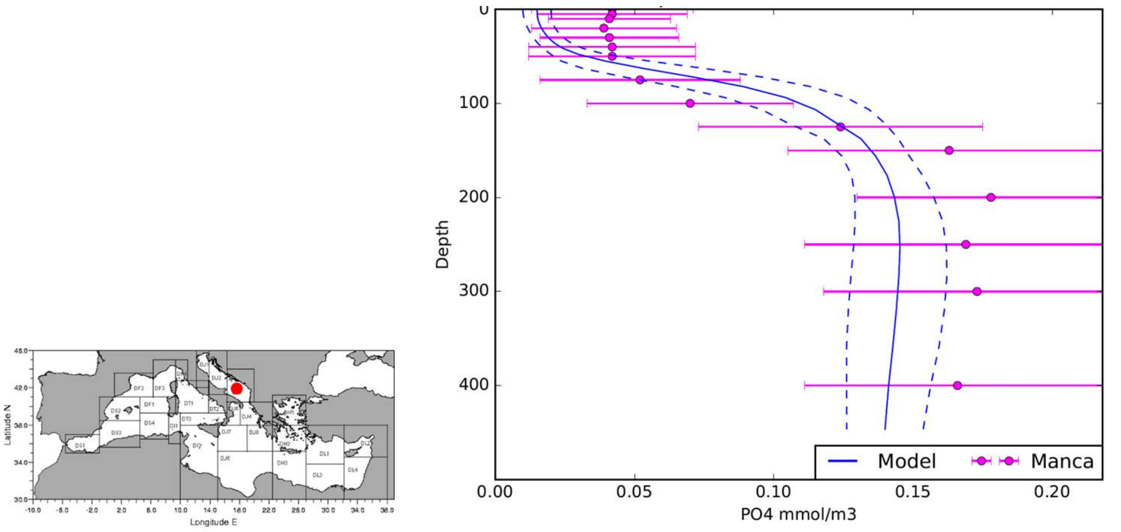
Depth	Model mean	Data mean	Model std	Data std	RMSE	normalized bias	% bias	Pearson's R	P-value
0-50	0.014	0.082	0.0017	0.007	0.07	-0.71	-483.57	-0.31	0.55
50-200	0.040	0.078	0.020	0.002	0.04	-0.32	-95.09	0.55	0.34
200-500	0.17	0.13	0.030	0.035	0.04	0.14	24.20	0.94	0.06

Figure A2. Annually averaged PO_4 vertical profile in the South Levantine sub–basin (see map and Manca et al. (2004)) and statistical indicators over different depths. Model values are the blue line (2005 average, Ntot simulation from Richon et al., 2017) and measured values from Manca et al. (2004) are in pink. Horizontal bars and dashed lines indicate spatial standard deviation of observations and model results respectively.



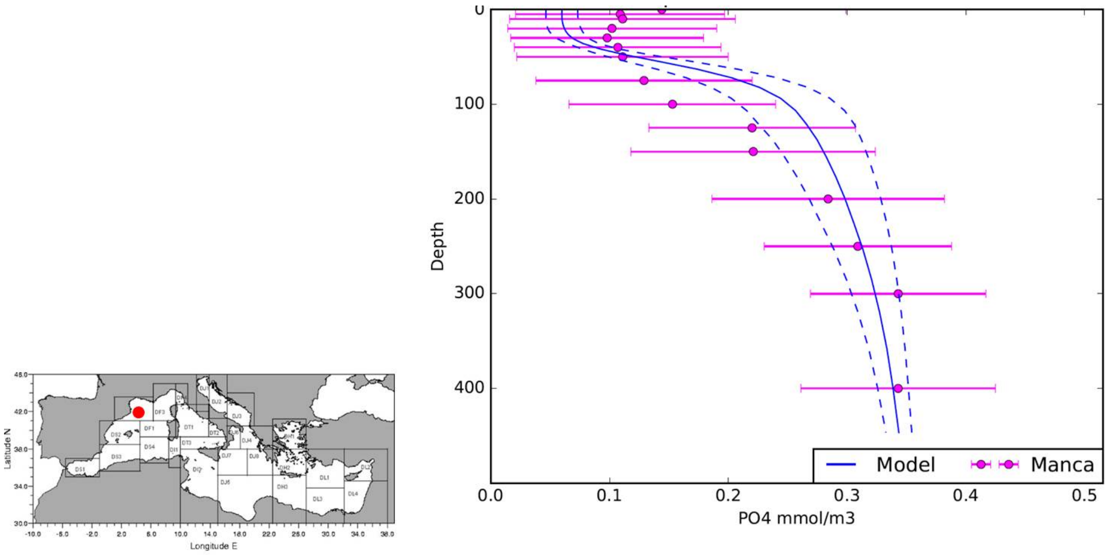
Depth	Model mean	Data mean	Model std	Data std	RMSE	normalized bias	% bias	Pearson's R	P-value
0-50	0.016	0.034	0.003	0.007	0.02	-0.35	-107.36	-0.40	0.43
50-200	0.050	0.039	0.028	0.010	0.02	0.13	22.86	0.96	0.01
200-500	0.20	0.11	0.023	0.025	0.09	0.30	46.03	0.96	0.04

Figure A3. ~~Nitrate~~ Annually averaged PO_4 vertical profile in the South Ionian sub–basin (~~top)~~ see map and ~~phosphate (bottom~~Manca et al. (2004)) ~~concentrations computed by~~ and statistical indicators over different depths. Model values are the ~~NEMOMED12/PISCES model (background)~~ blue line (2005 average, Ntot simulation from Richon et al., 2017) and measured ~~along the BOUM cruise (dots) (Moutin et al., 2012) between approximately Marseille~~ values from Manca et al. (2004) are in pink. Horizontal bars and ~~South~~ dashed lines indicate spatial standard deviation of ~~Cyprus~~observations and model results respectively.



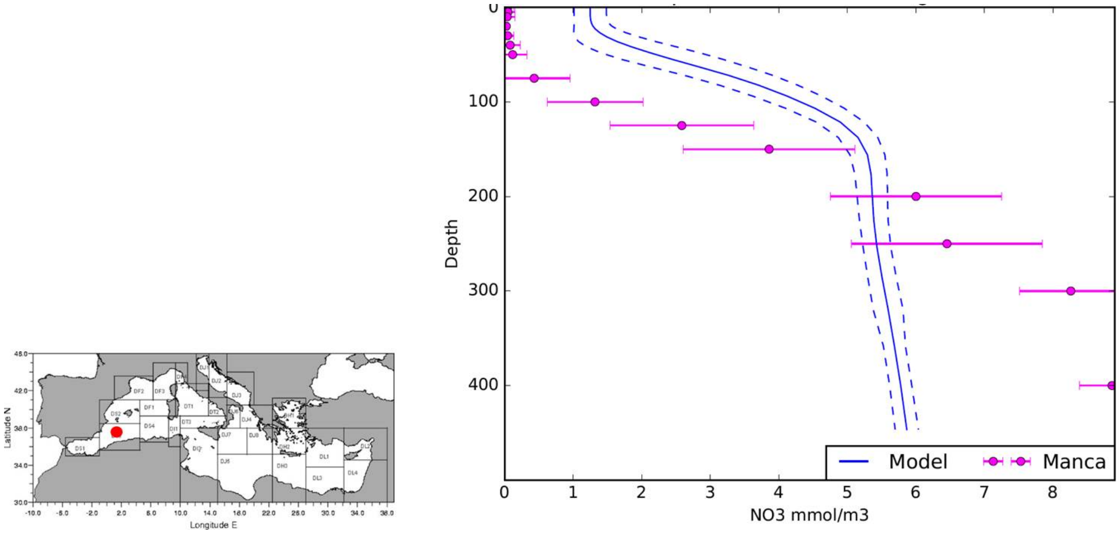
Depth	Model mean	Data mean	Model std	Data std	RMSE	normalized bias	% bias	Pearson's R	P-value
0-50	0.018	0.041	0.003	0.001	0.02	-0.40	-135.84	0.15	0.78
50-200	0.096	0.090	0.037	0.046	0.02	0.03	5.66	0.86	0.06
200-500	0.14	0.17	0.002	0.005	0.03	-0.09	-19.43	0.30	0.70

Figure A4. Annually averaged PO_4 vertical profile in the South Adriatic sub–basin (see map and Manca et al. (2004)) and statistical indicators over different depth. Model values are the blue line (2005 average, Ntot simulation from Richon et al., 2017) and measured values from Manca et al. (2004) are in pink. Horizontal bars and dashed lines indicate spatial standard deviation of observations and model results respectively.



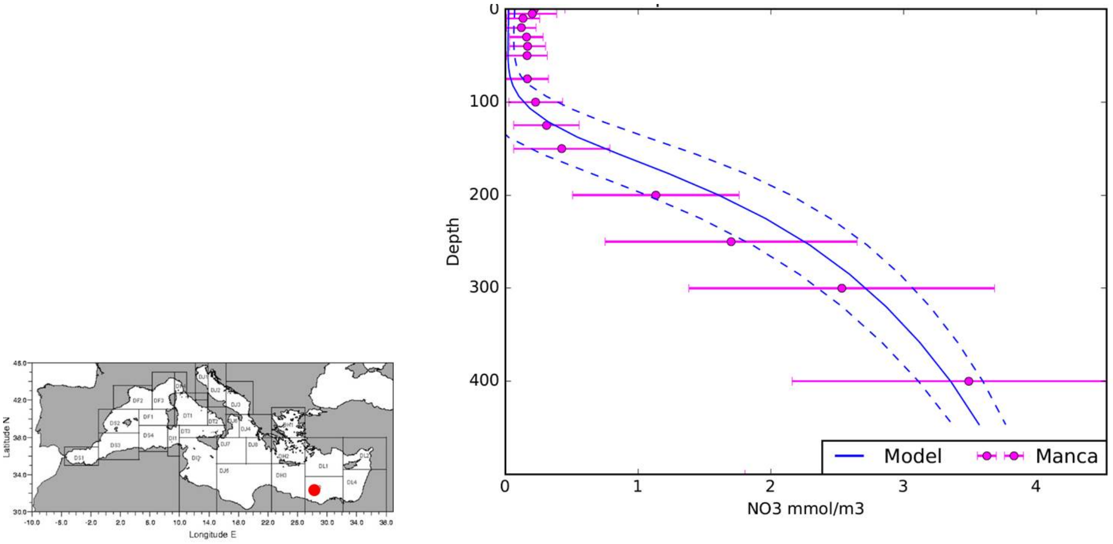
Depth	Model mean	Data mean	Model std	Data std	RMSE	normalized bias	% bias	Pearson's R	P-value
0-50	0.066	0.11	0.010	0.015	0.05	-0.26	-68.59	-0.31	0.56
50-200	0.23	0.17	0.057	0.046	0.07	0.15	26.15	0.86	0.06
200-500	0.32	0.32	0.015	0.025	0.01	0.00	-0.48	0.93	0.07

Figure A5. Annually averaged PO$_4$ vertical profile in the Gulf of Lions sub–basin (see map and Manca et al. (2004)) and statistical indicators over different depth. Model values are the blue line (2005 average, Ntot simulation from Richon et al., 2017) and measured values from Manca et al. (2004) are in pink. Horizontal bars and dashed lines indicate spatial standard deviation of observations and model results respectively.



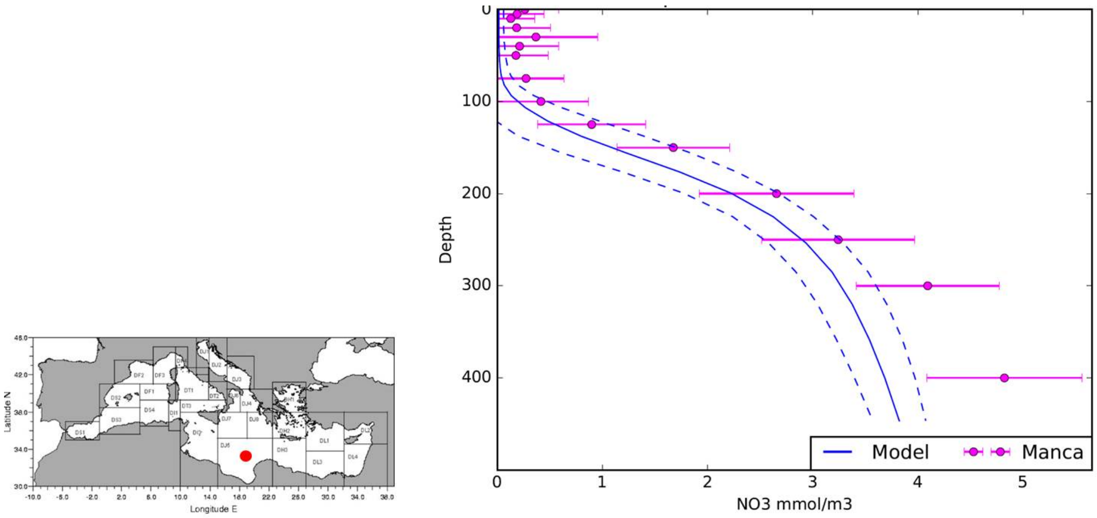
Depth	Model mean	Data mean	Model std	Data std	RMSE	normalized bias	% bias	Pearson's R	P-value
0-50	1.38	0.05	0.20	0.02	1.35	0.94	97	0.83	0.04
50-200	4.02	1.66	1.11	1.39	2.44	0.42	59	0.91	0.03
200-500	5.53	7.39	0.16	1.20	2.14	-0.14	-34	0.93	0.07

Figure A6. Annually averaged NO$_3$ vertical profile in the Algerian sub–basin (see map and Manca et al. (2004)) and statistical indicators over different depths. Model values are the blue line (2005 average, Ntot simulation from Richon et al., 2017) and measured values from Manca et al. (2004) are in pink. Horizontal bars and dashed lines indicate spatial standard deviation of observations and model results respectively.



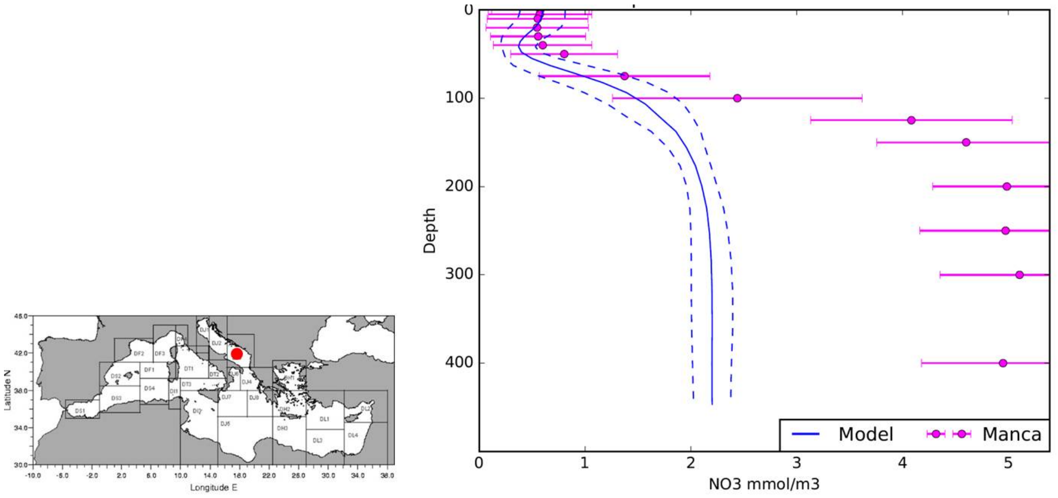
Depth	Model mean	Data mean	Model std	Data std	RMSE	normalized bias	% bias	Pearson's R	P-value
0-50	0.02	0.17	0.00	0.03	0.15	-0.75	-606	0.49	0.33
50-200	0.27	0.26	0.27	0.10	0.18	0.01	3	0.99	0.00
200-500	2.48	2.22	0.64	0.89	0.38	0.06	11	0.99	0.01

Figure A7. Annually averaged NO_3 vertical profile in the South Levantine sub–basin (see map and Manca et al. (2004)) and statistical indicators over different depths. Model values are the blue line (2005 average, Ntot simulation from Richon et al., 2017) and measured values from Manca et al. (2004) are in pink. Horizontal bars and dashed lines indicate spatial standard deviation of observations and model results respectively.



Depth	Model mean	Data mean	Model std	Data std	RMSE	normalized bias	% bias	Pearson's R	P-value
0-50	0.015	0.224	0.001	0.075	0.22	-0.87	-1394	0.14	0.78
50-200	0.381	0.687	0.402	0.552	0.34	-0.29	-81	1.00	0.00
200-500	3.020	3.704	0.534	0.824	0.76	-0.10	-23	0.98	0.02

Figure A8. Annually averaged NO_3 vertical profile in the South Ionian sub–basin (see map and Manca et al. (2004)) and statistical indicators over different depths. Model values are the blue line (2005 average, Ntot simulation from Richon et al., 2017) and measured values from Manca et al. (2004) are in pink. Horizontal bars and dashed lines indicate spatial standard deviation of observations and model results respectively.



Depth	Model mean	Data mean	Model std	Data std	RMSE	normalized bias	% bias	Pearson's R	P-value
0-50	0.52	0.57	0.09	0.02	0.11	-0.05	-10	-0.40	0.43
50-200	1.31	2.66	0.54	1.48	1.66	-0.34	-103	0.95	0.01
200-500	2.17	5.00	0.04	0.06	2.84	-0.40	-131	0.23	0.77

Figure A9. Annually averaged NO_3 vertical profile in the South Adriatic sub–basin (see map and Manca et al. (2004)) and statistical indicators over different depth. Model values are the blue line (2005 average, Ntot simulation from Richon et al., 2017) and measured values from Manca et al. (2004) are in pink. Horizontal bars and dashed lines indicate spatial standard deviation of observations and model results respectively.

Reviewer 1

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:
Referee#1 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 ± 0.177 10^{-3} g m^{-3} and the average model value is 0.205 ± 0.111 10^{-3} g m^{-3}. For PO4, the average measured value is 0.234 ± 0.085 mmol m^{-3} and the modeled average is 0.167 ± 0.179 mmol m^{-3}. This has been added in the manuscript lines 129-133.

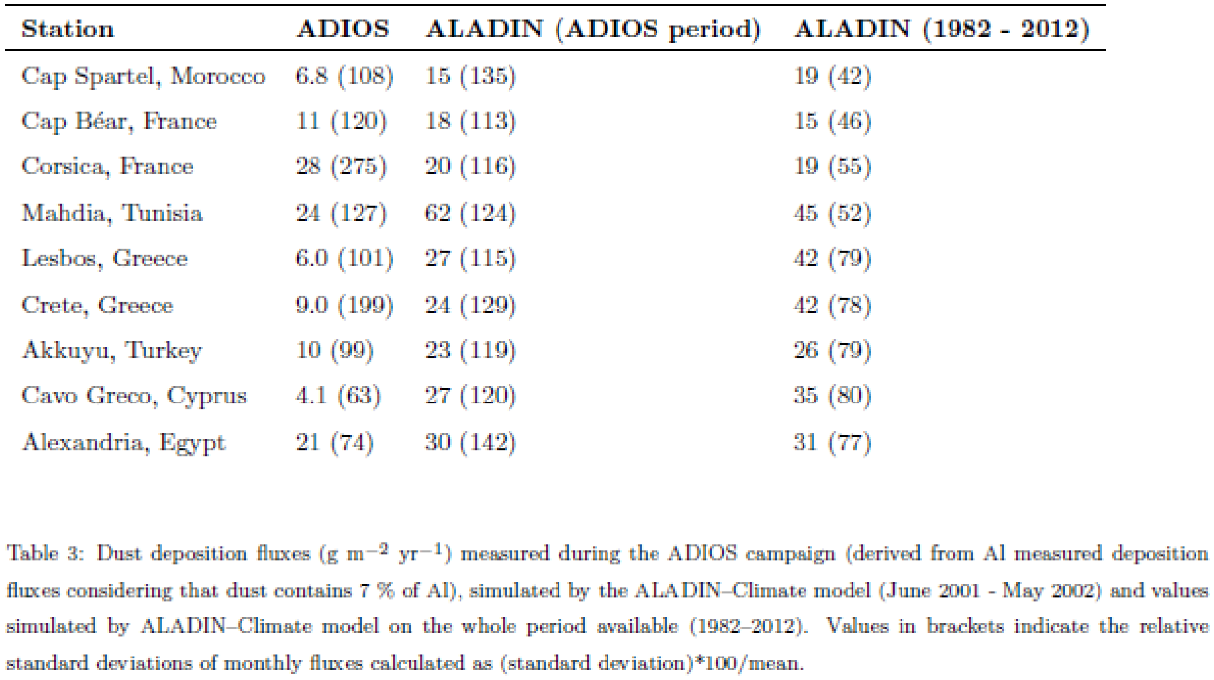
It should be noted that as suggested by Reviewer 2, we withdrew the appendix because the model evaluation is already shown in Richon et al. (2017, Prog. Ocean.) and shows the same figures and results.

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.



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.

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.

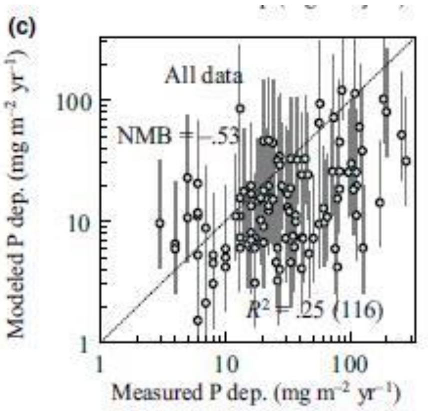
Moreover, the extrapolation of the deposition fluxes measured in a few localities to a basin scale average deposition flux as in Powley *et al.* (see their supplementary material) may lead to high uncertainties in the estimates. In particular, this method may not represent the important gradients in 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 % of the total (atmospheric + riverine input + Gibraltar Strait) as documented in Table 2."

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.



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

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).”

Total deposition of soluble phosphate from combustion (blue line) and natural dust (green line) over the entire Mediterranean basin for the 1997-20012 period.

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, phosphate inputs from desert dust outbreaks may be dominant on regional and daily or weekly time scales.

Reviewer 2

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 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 Pcomb and Pdust 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).

We thank the reviewer for these comments. It is true that the Mediterranean is likely to be a non-Redfieldian Sea and modeling this behavior would give interesting insights. However, the present version of the biogeochemical model PISCES we use is in a redfieldian configuration. A new version of the PISCES model is being developed with non-redfieldian ratios (Aumont, in prep). This non-redfieldian version of PISCES has been developed to treat the global ocean, and qualifying it for the Mediterranean Sea will likely take a few years more to come up with satisfactory results for the region.

Specific comments

In some cases, the manuscript methodology could benefit from further explanation. For example: I was very confused about how PO4 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 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.*

We changed the following sentence that brought confusion about the way PISCES treats biogeochemical tracers: "*PISCES passive biogeochemical tracers are transported using an advection–diffusion scheme.*" Into "*PISCES biogeochemical tracers are transported using an advection–diffusion scheme* "… In PISCES, PO_4 is one of the nutrients necessary for plankton growth; it is not a passive tracer. Phosphate concentration, as well as the 4 other nutrients represented in PISCES (NO_3, NH_4, Si and Fe), are used to calculate the nutrient limitation terms (that have a Michaelis-Menten formulation). These limitation terms allow calculating the productivity terms based on the use of each nutrient. The phytoplankton biomass, which is linked to chlorophyll production, is then derived from the productivity terms. All equations are in Aumont *et al. (*2015).

Offline models, in contrast to online or coupled models are run thanks to the use of climatological values of physical and biogeochemical boundary fluxes. In our case, the physics of the ocean is described by the model NEMO, and the biogeochemical cycles are represented by the PISCES model. NEMO allows calculating the movements of water masses using climatological values (forcings) of atmospheric and physical conditions such as winds, runoff or precipitations. The biogeochemical model PISCES calculates the biogeochemical state of the Mediterranean (nutrient and tracers concentrations) using the physical state from NEMO and biogeochemical conditions from climatologies such as nutrient inputs from rivers. The biogeochemistry in PISCES is calculated in the same way as in Guyennon et al. To clarify, we add the sentence "Biogeochemical variables are prognostically calculated and not read from forcing files."

On a related note, how exactly was surface PO4 related to Chl a in the model? I did not see this discussed, or any of the associated uncertainties.

To clarify, we added in section 2.1 "The concentration of nutrients is linked with phytoplankton productivity and chlorophyll-a production according to the equations described in Aumont *et al.* (2015). Phytoplankton growth rate is dependent on nutrient concentrations via the growth limiting factors"

Section 3.3 and figure 5: Where does the referred-to surface PO4 data come from? From the model or from observations?

We changed the Figure 5 legend "Map of daily maximal relative effects of total (*Pdust* + *Pcomb*) deposition in June 2005 on the surface phosphate concentration (0—10 m) compared to the reference simulation without atmospheric P deposition."

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

In the LMDz-INCA model, an explicit deposition scheme is implemented. It represents 3 physical processes of deposition including sedimentation, turbulent dry deposition and wet deposition (in-cloud and below-cloud scavenging). These schemes allow accounting for more complex physical processes than the simple hypothesis of a constant deposition speed.

The observations we use for model evaluation are direct measurements of phosphorus bulk deposition (see Guieu *et al.* 2010 mar. chem. for protocol details). No estimations from atmospheric concentrations or optical properties such as AOD are 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 PO4 by performing two different simulations: "PDUST" and "PCOMB"; they include, respectively, natural dust only and combustion–generated aerosol only as atmospheric sources of PO4. 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 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 PO4. 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.

We agree with the reviewer that the use of different terms is quite confusing. We modified some sentences in the text to clarify this point:

Section 2.3: "From now on, we name "total P" the sum of bioavailable phosphate from dust and combustion (*Pdust* + *Pcomb*)."

Section 3.1 "We used the times series of total phosphorus measured at 9 different stations over the Mediterranean from the ADIOS campaign (Guieu et al 2010) and the soluble phosphate measured in the deposition at 2 stations in the South of France from the MOOSE campaign"

Section 3.3 We renamed the section "Impacts of atmospheric deposition on marine surface phosphate budgets ", and line 298: "Atmospheric deposition of phosphate aerosols has different impacts"

"Figure 5 shows the relative impacts of phosphate deposition from the two sources (combustion and dust) on surface PO4 concentration for the month of June 2005. The relative impacts of atmospheric deposition from different sources are dependent on both the underlying phosphate concentration and the bioavailable phosphate deposition flux."

We use the term "phosphate deposition" in the text because our focus is on the deposition of this bioavailable nutrient.

On a similar vein, P1 l.15: "We examine separately the different soluble phosphorus (PO4) sources. . ." Please keep in mind again that soluble phosphorus and PO4 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.*

We thank the reviewer for this remark. We are conscious that soluble P can also describe organic P. In this manuscript, we refer to the only soluble phosphorus form in PISCES which is PO4. We replace soluble phosphorus in this sentence by "phosphate".

The authors talk about other sources of surface PO4 (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.

As described in section 2.1, riverine fluxes of nutrients are prescribed from Ludwig et al. 2009. This study groups the nutrient fluxes from 239 rivers around the Mediterranean and Black Sea obtained from measurements and model data. Unfortunately, the estimations of riverine fluxes are not available after 2000. This is why we used the riverine fluxes from the year 2000 in our study. The nutrient fluxes from the Atlantic are computed as the product of the buffer zone concentrations constructed from the World Ocean Atlas (2005) and the water fluxes through the Strait of Gibraltar computed by the model. We added these precisions to the section 2.1.

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 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.

We thank the reviewer for this comment and refer to the general comments at the beginning of this section.

We added a paragraph on the implications of the use of a redfieldian model in the discussion section.

"The PISCES version used in this study is based on the Redfield hypothesis that C/N/P ratios in organic cells are fixed. This fixed value determines the nutrient ratio for uptake and has the advantage of simplifying calculations in the 3-D high resolution coupled model. However, because the Mediterranean is highly oligotrophic, the biogeochemical cycles may be determined by non-Redfieldian dynamics (see Ribera d'Alcala *et al.* 2003, JGR). This non-Redfieldian behavior may imply complex nutrient limitations and co-limitations processes that can not be studied with the present PISCES version. To this day, there is no version of PISCES available that includes the non-Redfieldian biogeochemistry in the Mediterranean. The development and use of such a version of PISCES is a perspective of this work that needs to be undertaken in order to fully understand nutrient dynamics and growth limitation process in the Mediterranean. This study provides first results on the potential impacts of phosphate atmospheric deposition on the Mediterranean nutrient pool and potential implications on biological productivity assuming the Redfield hypothesis."

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 PO4 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.

We agree with the reviewer that including organic phosphorus is an important step for the community. Our hypotheses concerning phosphorus combustion in this study are only based on Mahowald *et al* (2008). However, Myriokefalitakis et al (2016) consider that organic phosphorus (DOP) can be deposited in the Mediterranean with combustion and biogenic aerosol. DOP is not included in the version of PISCES used in this study. However, if we consider the hypothesis of Kanakidou *et al* and Myriokefalitakis *et al*, and given that dissolved organic matter is recycled into inorganic nutrients in the sea, we may be able to consider the inclusion organic phopshorus as a source of atmospheric phosphate. We add some elements in the discussion section: "In the Mediterranean region that is surrounded by many forested areas, biogenic emissions may be an important source of atmospheric phosphorus in the form of organic matter. Moreover, Kanakidou *et al.* (2012) show that an important fraction of organic phosphorus can be emitted from combustion. In particular, the numerous forest fires occurring every summer in the Mediterranean region may constitute an important source of organic phosphorus. However, the PISCES version used in this study does not include organic phosphorus. In the ocean, organic phosphorus can be recycled by bacterial activity into inorganic phosphate that is bioavailable for plankton growth. Therefore, the inclusion of organic phosphorus in PISCES along with an estimation of organic phosphorus from atmospheric fluxes is a perspective to consider."

3) Uncertainties with the model assumptions themselves require further discussion. For example: The majority of the results focus and rely on modeled ocean surface PO4 concentrations. However, the majority of the model evaluation focuses on subsurface ocean PO4 trends, or surface Chl a trends. There was no in-depth discussion of how well the model compared to surface PO4 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 PO4, either as parameterized in the model, or in actual observations.

The following Figure displays the PO4 concentration on the BOUM section in the top 200 m (zoom from the A1 Figure from the manuscript). We have included in appendix a couple of figures to evaluate surface PO_4.

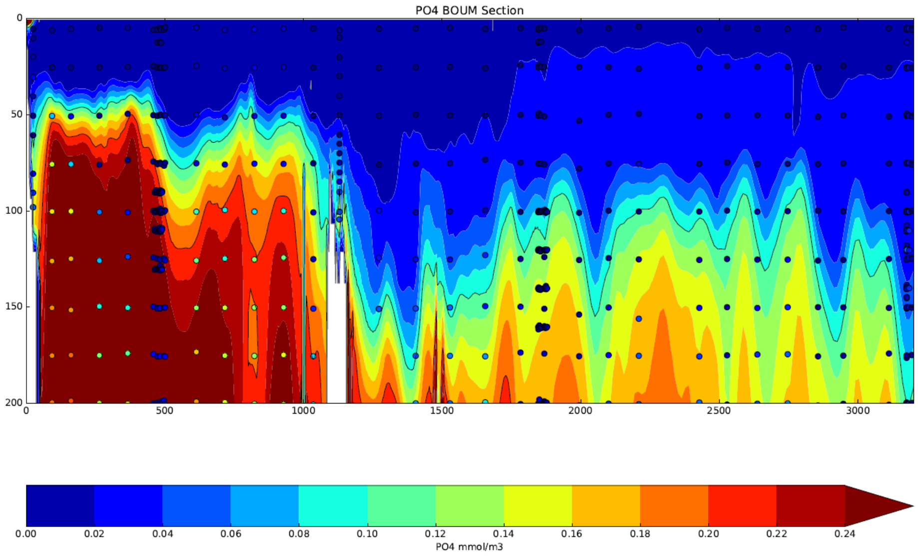


Figure 1 PO4 concentration along the BOUM section (Moutin et al 2012). Zoom from the top 200m.

BOUM is the most complete dataset available for the Mediterranean because it covers a full, recent west-to-east transect. There are very few estimations of nutrient concentration (and especially phosphate) in the surface layer of the Mediterranean (first 5-100 m) because the concentrations are so low that measures are often below the detection limit of sensors. In this figure, we can see that the model reproduces the increase in concentration below 50 m observed in the western basin but that the increase in concentration modeled in the eastern basin (below 100-150 m) is not observed.

For further comparison, we show in the figures below the average phosphate profiles in different regions of the Mediterranean compared with data from Manca *et al.* (2004). These figures show that the model can reproduce the phosphate vertical distribution and that the model values are generally in the range of data standard deviations in surface waters.

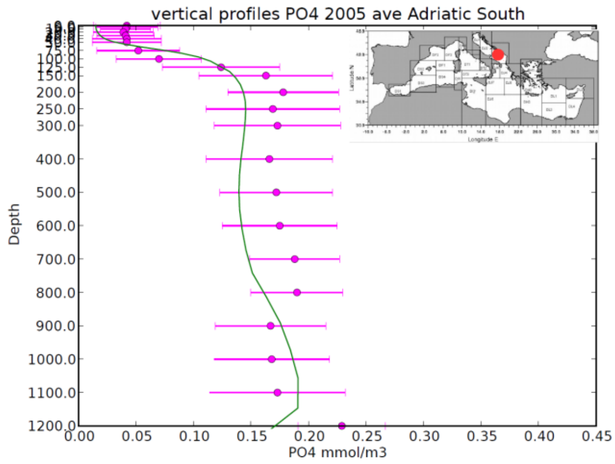


Figure 2 Average concentration in 2005 in the South Aegean region (see map). Measurements and standard deviations are in pink, modeled values are represented by the green line.

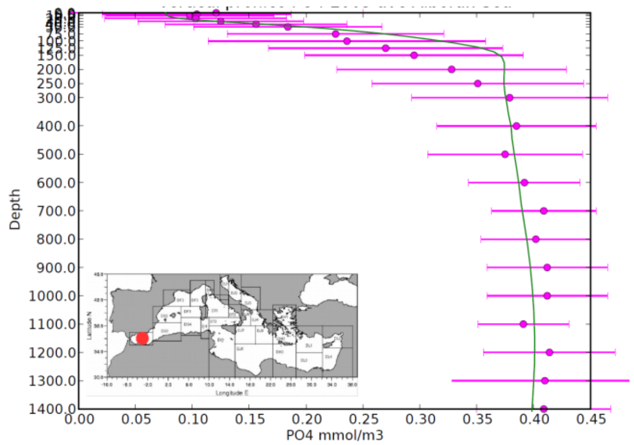


Figure 3 Average concentration in 2005 in the Alboran Sea region (see map). Measurements and standard deviations are in pink, modeled values are represented by the green line.

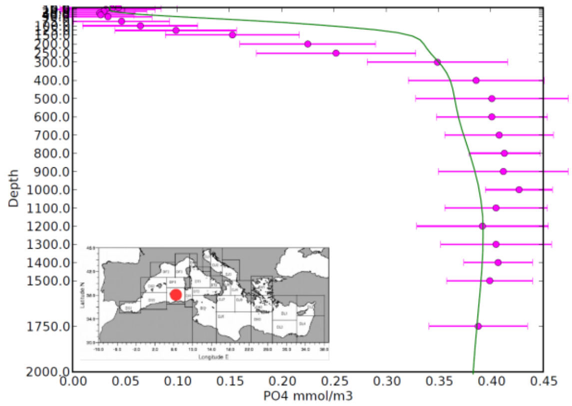


Figure 4 Average concentration in 2005 in the Algerian current region (see map). Measurements and standard deviations are in pink, modeled values are represented by the green line.

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 PO4 supply. In comparison, the average contribution of Pdust to PO4 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.

The contribution values given in p.11, l342 are the values from Table 2. They are based on our modeling values and take into account only the sources of phosphate that are included in the simulations (namely rivers, Atlantic inputs, desert dust and combustion derived atmospheric phosphate). These are estimates for our present simulation that do not represent the absolute truth on the contribution of atmospheric phosphate deposition, but give light on the relative importance of the 2 atmospheric sources under the specific conditions of the year 2005, according to the LMDz-INCA model outputs. The purpose of this Table (and of this study in general) 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. The Table aims at shading new light on the other sources and their potential role. Acknowledging that model limitations makes those number highly uncertain, they suggest that Saharan dust might not be as dominant as it was previously believed as a source of bioavailable nutrients. We added these precisions in the discussion section. (lines 371-390).

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.

Reviewer is right to point out that nutrient co-limitation is a key question in the study of marine biogeochemical cycles and in particular in oligotrophic areas such as the Mediterranean. PISCES is a Monod type model in which nutrient limitations are calculated in a Michaëlis-Menten formulation. This means that growth rates of phytoplankton increase linearly with nutrient concentrations when these concentrations are below a threshold. In an oligotrophic region such as the Mediterranean the concentrations are low enough for the growth rate to increase linearly with concentration. As a consequence, having no increase in productivity (that is linked to nutrient limitations) after phosphate deposition is a sign that growth rates are limited by at least one other nutrient (most probably N).

We added a paragraph on section 3.4. "In general, we can identify 3 different biogeochemical responses in the 3 framed areas of Figure 7. Our hypothesis is that the different responses are linked to nutrient limitations. In the North Adriatic, the influence of coastal nutrient inputs leads to low nutrient limitation and high productivity. In the South Adriatic, the high impact of atmospheric phosphate deposition may be the sign of important phosphate limitation. Finally, the lack of response in South Ionian in spite of the relatively high atmospheric phosphate deposition probably indicates that the region is co-limited in P and N."

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.

Largest by mass.

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

We added a reference to Moore et al (2013).

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.

We agree with the reviewer and removed sea spray.

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.

We rephrased this part : "The Mediterranean Sea is also a hot-spot for climate change impacts. Moreover, it is the recipient of aerosols from a variety of different geographical sources. The impacts of aerosol deposition on the Mediterranean region are not fully understood and they may change in the future as a result of climate change impacts on land and sea."

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

Changed to "These evaluations showed that the NEMO model is able to produce satisfying results when studying characteristics such as age-tracer of water masses of passive tracer transport."

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 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.

The model setup in this paper is the same as the one in Richon et al (2017). In the present study, we compare the model outputs with data for the year 2005. We decided to follow the advice of the reviewer and removed the appendix. We added in section 2.1 "The model NEMOMED12/PISCES is run in the same configuration than in Richon *et al.* (2017) who provide an evaluation of the model. In particular, the authors show that NEMOMED12/PISCES reproduces a correct west-to-east gradient of productivity when compared to satellite chlorophyll estimates in spite of some underestimation in the areas of high productivity such as the Gulf of Lions that they trace back to the circulation anomalies of the western basin. The vertical distribution of nutrients is satisfyingly reproduced by the model in spite of underestimations in the Levantine Intermediate Waters (LIW) because of the too smooth nutricline."

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.

Changed.

P7, l213: "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.

We agree with the reviewer that calculating deposition fluxes from aerosol concentration can lead to high uncertainties, in particular when extrapolating the fluxes to an entire region based on average concentrations. This is the reason we tried to evaluate directly the modeled deposition fluxes, taking into account dry and wet deposition processes and daily variability.

Deposition time series are only available at a few stations, and observations are not available in 2005 (our model year). This leads to a high uncertainty in our comparison data. But we believe that these measurements are more reliable to compare deposition fluxes than basin scale estimations that do not account for large deposition gradients. Our approach is more process-based and should lead to less uncertainty than basin scale extrapolation from velocity fluxes. However, the exclusion of soluble organic phosphorus, PBAB and sea salt inevitably leads to some additional uncertainties.

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

Done

Table 2: Please mention in the Table or the caption that these estimates are modelderived. 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 PO4. Does the Krom et al estimate include rivers? Please specify

Precisions added in the figure caption

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.

Caption changed.

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?

Changed to "more important impacts on surface marine productivity"

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?

We changed the reference

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.

We did not follow this reviewer suggestion. However, as previously stated, we do not believe that AOD is a good proxy for deposition. High deposition is generally related to rain, which means very cloudy conditions unfavorable to AOD measurements.

P9, l. 273: "Atmospheric phosphorus deposition has different impacts on PO4 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 PO4 concentration depending on the source, the location, and the period of the year."

Done

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 PO4 data? From the model or from observations? If in the model, how well does the model reproduce observations?

We rephrased the caption.

P9 l. 278: "Figure 5 shows the relative impacts of phosphorus deposition from the two sources (combustion and dust) on surface PO4 concentration for the month of June. The relative impacts of atmospheric deposition from different sources are varying over 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.

As stated in section 3.2, we focus on June because it is the month of the year when maximal effects of deposition on surface productivity are observed. In Richon et al. 2017, we link this result to the vertical stratification and high surface nutrient limitations associated with sufficient deposition fluxes. We removed the term "varying over time" from the sentence because it was confusing.

Fig. 6: again, what does Total P represent in this instance? Pdust + Pcomb? 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 PO4 and Chl a, Redfieldian assumptions, etc.

We added the following information: "In this Redfieldian version of PISCES, chlorophyll production is linked with nutrient uptake that is constrained by the Redfield ratio. Therefore, the addition of excess nutrient will enhance chlorophyll production as long as other nutrients are bioavailable in the Redfield proportions. These results may change in a non Redfieldian model."

P10 l.330: "We performed a Student's t–test on the grid matrix of relative impacts of Pdust and Pcomb 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 Pdust are close to the effects of Pcomb 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).

Reviewer is right. Also, given the high uncertainty on deposition fluxes, we chose to remove this sentence.

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.*

We thank the reviewer for this suggestion. We modified the sentence and added to this paragraph "In general, results from this idealized study suggest that the impacts of atmospheric deposition of phosphate are likely to be fairly small, even though atmospheric sources of phosphate seem to be important contributors to the total nutrient pool in some regions of the basin."