

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 on the only available year (2005 by a)~~ from the global atmospheric chemical transport model (LMDz-INCA) as additional sources of external nutrient for a high resolution (1/12°) regional coupled dynamical–biogeochemical model of the Mediterranean Sea (NEMOMED12/PISCES). In general, dust is considered as 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 (PO₄, 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 $40.5 \cdot 10^{-6} \text{ mol PO}_4 \text{ m}^{-2} \text{ year}^{-1}$ over the entire Mediterranean Sea for the year 2005 and is the primary source over the northern part (e.g., $101 \cdot 10^{-6} \text{ mol PO}_4 \text{ m}^{-2} \text{ year}^{-1}$ from combustion deposited in 2005 over the North Adriatic against $12.4 \cdot 10^{-6}$ from dust). Lithogenic dust brings $17.2 \cdot 10^{-6} \text{ mol PO}_4 \text{ m}^{-2} \text{ year}^{-1}$ 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., $31.8 \cdot 10^{-6} \text{ mol PO}_4 \text{ m}^{-2} \text{ year}^{-1}$ from dust deposited in 2005 on average over the South Ionian basin against $12.4 \cdot 10^{-6}$ from combustion). We examine separately the ~~different soluble phosphorus (PO₄) 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. These different sources should therefore be accounted for in modeling studies.

25 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; Heimbürger 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, fungi 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 (Lejeune et al., 2010), ~~in part because~~. 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.](#) 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 $1/12^\circ$ over the Mediterranean (MED12). The $1/12^\circ$ 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 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.

105 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 110 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/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 115 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 reports 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~~ 120 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). The nutrient fluxes from the Atlantic are computed as the product of nutrient concentrations in the buffer zone by the water fluxes through the Strait of Gibraltar computed by the model.

The model is run in off–line mode like in the studies performed by Palmiéri et al. (2015), Guyenon 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 and not 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 the basin in spite of a too smooth nutricline, and the main productive zones that are the Alboran Sea, the Gulf of Lions and most coastal areas(~~see appendix~~) –. Richon et al. (2017) calculated the average and

standard deviation of chlorophyll 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 $0.290 \pm 0.177 \text{ } 10^{-3} \text{ g m}^{-3}$ and the average model value is $0.205 \pm 0.111 \text{ } 10^{-3} \text{ g m}^{-3}$. For PO_4 , the average measured value is $0.234 \pm 0.085 \text{ } 10^{-3} \text{ mol m}^{-3}$ and the modeled average is $0.167 \pm 0.179 \text{ } 10^{-3} \text{ mol m}^{-3}$. We report in appendix additional figures on the comparison of modeled and measured surface PO_4 concentrations.

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 in 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.

2.2 Atmospheric deposition of phosphate

~~We use atmospheric~~The objective of this study is to use atmospheric phosphate inputs from contrasted sources simulated by the same atmospheric model for consistency. 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). ~~This global model has a rather low spatial resolution of 0.94° in latitude $\times 1.28^\circ$ in longitude, but it is, to our knowledge, the only atmospheric model available including different phosphorus sources.~~ For the two sources, daily deposition fluxes are simulated globally ~~for 2005, only 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-desert~~ dust. 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 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 $\times 1.28^\circ$ in 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
170 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 2005 is close to the
multi-year deposition average (~~not shown~~).

The second source of atmospheric phosphorus is combustion. Here, the term combustion entails
anthropogenic combustion from energy production, ~~biofuels~~ and ~~biofuels~~, and wildfires emissions
175 (hereafter named *P_{comb}*). The *P_{comb}* deposition fields from LMDz-INCA used here have a coarser
resolution than for *P_{dust}* 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
180 for 2005 are close to the inter-annual average for the period 1997–2009. Wang et al. (2015a) esti-
mated global *P_{comb}* 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 (*P_{comb}*) (Longo et al., 2014).

The LMDz-INCA modeled atmospheric P deposition ~~rates~~ ~~fluxes~~ have been evaluated globally
185 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 (*P_{comb}*)
(Longo et al., 2014).~~
190

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.

195 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.) al-
though 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
200 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 par-
ticles (named PBAP). We chose to discard PBAP as a source of P since these 2 contrasted sources
205 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 name "total P" the sum of bioavailable phosphate from dust and combustion ($P_{dust} + P_{comb}$). The results presented in this study are based on the relative differences between the simulations. For instance, the impacts of P_{dust} 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 measured in the deposition 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 $10^{-6} \text{ g PO}_4 \text{ m}^{-2} \text{ month}^{-1}$ and compare it with our model average monthly fluxes in the grid cells corresponding to the stations. Figure 1 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.

240 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 ~~-We observe a~~ at several stations, in
spite of the low spatial variability in 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~~
245 ~~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 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)
250 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 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
255 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 Mediter-
ranean 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
260 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

265 The 2005 seasonal spatial distribution of *Pdust* deposition is shown in Figure 2. *Pdust* deposition is
highly variable in space and time. It is maximal in spring (MAM). In this season the main source is
the Sahara and 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
270 over the basin is $0.122 \cdot 10^9 \text{ g PO}_4 \text{ month}^{-1}$ with notable monthly variability (standard deviation =
 $0.102 \cdot 10^9 \text{ g PO}_4 \text{ month}^{-1}$). 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 3. Atmospheric depo-
275 sition of phosphate from combustion is on average $0.258 \cdot 10^9 \text{ g PO}_4 \text{ month}^{-1}$ over the entire basin. It

amounts twice the atmospheric deposition of phosphate from desert dust ($0.122 \cdot 10^9 \text{ g PO}_4 \text{ month}^{-1}$). The seasonal variability of *Pcomb* deposition is lower than that for *Pdust* (standard deviation of *Pcomb* = $0.046 \cdot 10^9 \text{ g PO}_4 \text{ month}^{-1}$). 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 $0.0529 \cdot 10^9 \text{ g PO}_4 \text{ month}^{-1}$ and $0.0118 \cdot 10^9 \text{ g PO}_4 \text{ month}^{-1}$ 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 result, which remain highly uncertain, the riverine inputs computed from the PISCES model would constitute the main phosphate source to the Mediterranean Basin ($3.16 \cdot 10^9 \text{ g PO}_4 \text{ month}^{-1}$ at the basin scale, see also). They account for over 85 % of the total (atmospheric + riverine input + Gibraltar Strait) as documented in Table 2).

We show the map for the month of June 2005 as an example of contrasted contribution of the respective fluxes in Figure 4. 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 North Adriatic, the South Adriatic and the South Ionian (See Figure 2). We took the definitions for these regions in Manca et al. (2004). These regions were selected because they highlight three contrasted conditions. The North Adriatic is under strong influence of both riverine inputs and atmospheric deposition of P from combustion (Figure 34), 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 2 shows the relative contribution of atmospheric ~~P-sources to other external fluxes~~ phosphate sources to total external phosphate supply to the Mediterranean basins given by our model. 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 P_{comb} is dominant over P_{dust} as a source of atmospheric phosphate at the basin scale~~ is P_{comb} for the year 2005, according to our model. This dominance is found in all regions of the Mediterranean, except in the Ionian Sea where P_{dust} and P_{comb} 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, P_{dust} and P_{comb} 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 P_{dust} 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 5 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 ~~-2005~~. The relative impacts of atmospheric deposition from different sources are ~~varying over time and depend~~ 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.

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 P_{comb} 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 ($90 \cdot 10^{-6} \text{ mol m}^{-2}$ 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.

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 2).

We focus here on the month of June that shows maximum impacts of deposition because of surface water stratification. Figure 6 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. These results may change in a non Redfieldian model.

Figure 7 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 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, *Pdust* deposition flux is minimal in summer. This explains 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 7 the area in the Adriatic influenced by riverine inputs. This zone

385 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).

390 *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~~

395 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 atmospheric phosphate deposition probably indicates that the region is co-limited in P and N.

400 As for the effects on PO₄ 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 depleted 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).

405 **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 PO₄ supply. In comparison, the average contribution of *Pdust* to PO₄ supply is 4 % (Table 2). 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 restricted availability of daily deposition fluxes to the only year 2005

415 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

420 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 Sandroni, 1999; Aghnatios et al., 2014). This
study 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
425 seems to be confirmed by the analysis of yearly model deposition fluxes of P_{dust} and P_{comb} over the
1997–2012 period (not shown). P_{comb} dominates P_{dust} contribution to PO_4 supply over the north-
ern basin (Adriatic and Aegean Seas in particular). For these regions in the vicinity of anthropogenic
sources, P_{comb} deposition has a low variability whereas P_{dust} deposition occurs during transient
430 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 contribu-
435 tion 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
 P_{dust} is very low. The high, nearly constant fluxes of P_{comb} 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
440 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 P_{comb} as a source of
bioavailable phosphate. 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 model must be
undertaken in order to make more precise assessments of the importance of atmospheric deposition
445 as a source of nutrients for the oligotrophic Mediterranean.

The LMDz-INCA model version used in this study integrates constant emissions of P_{comb} 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: P_{dust} deposition
450 forcing has 280x193 grid points globally and ~500 grid points covering the Mediterranean, and P_{comb} forcing has 144x143 grid points in total and ~200 grid points covering the Mediterranean. These forcings reproduce well the average forcing reproduce realistic deposition patterns at the basin
455 regional model Mediterranean model available that represents phosphorus deposition from both nat-

ural and anthropogenic sources. Investigating these atmospheric deposition fluxes from a higher resolution regional model is a perspective to consider.

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 ± 0.102 10⁹ g month⁻¹ over the Mediterranean in 2005 simulated with LMDz–INCA and 0.568 ± 0.322 10⁹ 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 1. 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. 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 Turquetly et al. (2014) indicates a ± 50 % annual variability, but it is impossible to give any inter–annual variability of *Pcomb* deposition at present. Simulating longer time periods of atmospheric deposition would give interesting perspectives on the evolution of anthropogenic 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 1). 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 2, 3). 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., 495 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 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 505 of organic phosphorus from atmospheric fluxes is a perspective to consider. Moreover, the composition of biogenic aerosols and its solubility is still poorly constrained.

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 515 the Mediterranean is highly oligotrophic, 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. There is up to date 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 520 this work that needs to be undertaken in order 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.

5 Conclusions

525 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 Mediter-

530 ranean 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 535 is strongly dominant in pelagic areas such as the Middle Ionian and Levantine basins. 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 540 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 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. 545

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 550 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 555 cover only very limited areas of the basin and short time periods. Plus, there is, to our knowledge, no experimental study addressing the source apportionment of phosphate deposition. 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 560 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.

Acknowledgements. The PhD grant of C.R. is funded by CEA. R.W. was funded by a Marie Curie IIF project from European Commission (FABIO, grant 628735). This study contributes to MERMEX (Marine Ecosystem Response Mediterranean Experiment; <https://mermex.mio.univ-amu.fr>) and ChArMEX (the Chemistry-aerosol Mediterranean Experiment; <http://charmex.lsce.ipsl.fr>) projects of the programme MISTRALS (Mediterranean Integrated Studies at Regional and Local Scales; <https://www.mistrals-home.org/>).

Appendix A: Evaluation of NEMOMED12/PISCES against available observations surface phosphate

570 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
575 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
580 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.

We evaluated the large scale vertical distribution of nutrients against observations from the BOUM
585 campaign (Moutin et al., 2012) (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
590 due to the anomalies in SSH in the Algero-Provençal basin (Hamon et al., 2015). Concentrations in
the eastern basin are correctly simulated.

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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 1. Dust deposition fluxes ($\text{g m}^{-2} \text{yr}^{-1}$) 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 1).

Basin	Total P	Pdust	Pcomb	Ref.
East	28			Krom <i>et al.</i> , 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 2. 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 2. [Values from Krom et al. \(2010\) also include river inputs](#).

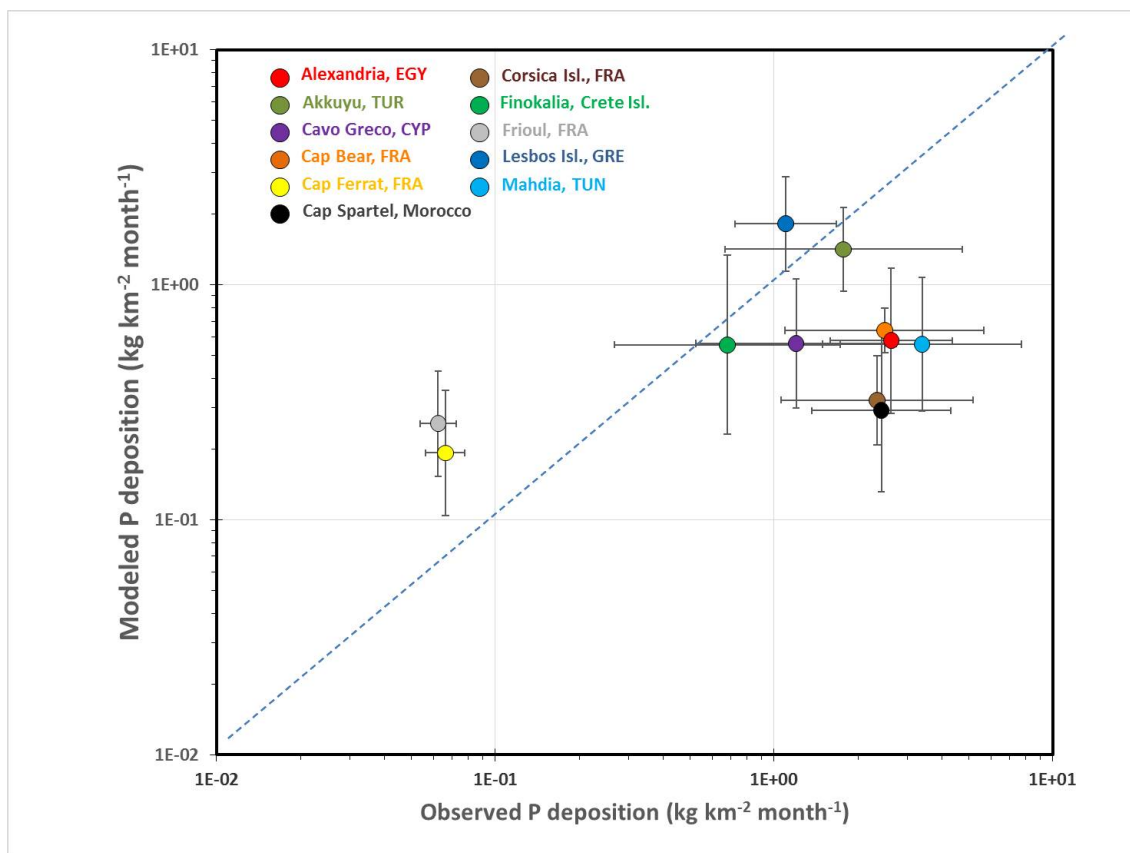


Figure 1. Comparison of modeled and observed monthly geometric mean of total P ($P_{dust} + P_{comb}$) deposition fluxes at the 9 ADIOS stations (Guieu et al., 2010) and soluble PO_4 at Frioul and Cap Ferrat stations (de Fommervault et al., 2015). Each point is the geometric mean of monthly observed 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).

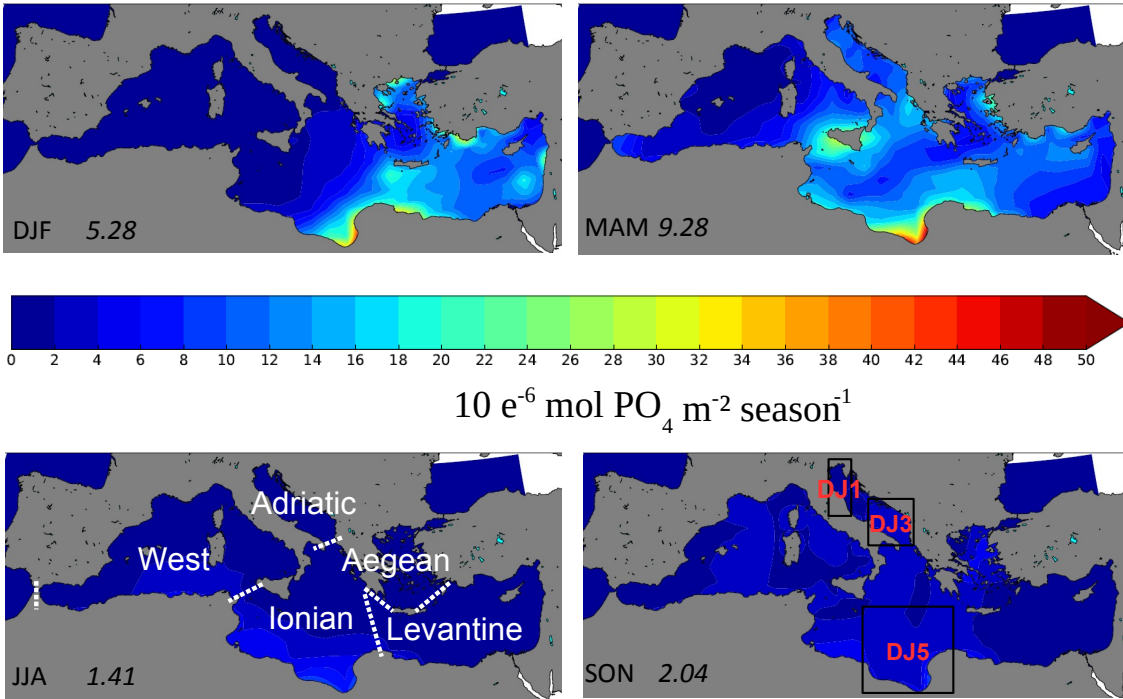


Figure 2. Total seasonal desert dust derived soluble phosphorus deposition (P_{dust} , in $10^{-6} \text{ molPO}_4 \text{ m}^{-2} \text{ season}^{-1}$) over each season of the year 2005 (molar flux is calculated as mass flux/phosphorus molar weight) [from the LMDz-INCA model](#). Numbers on the maps are the average seasonal deposition fluxes over the whole basin in $10^{-6} \text{ molPO}_4 \text{ m}^{-2} \text{ season}^{-1}$. In the Summer (JJA) deposition map, we display the different sub regions referred to in the text. In the Autumn (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.

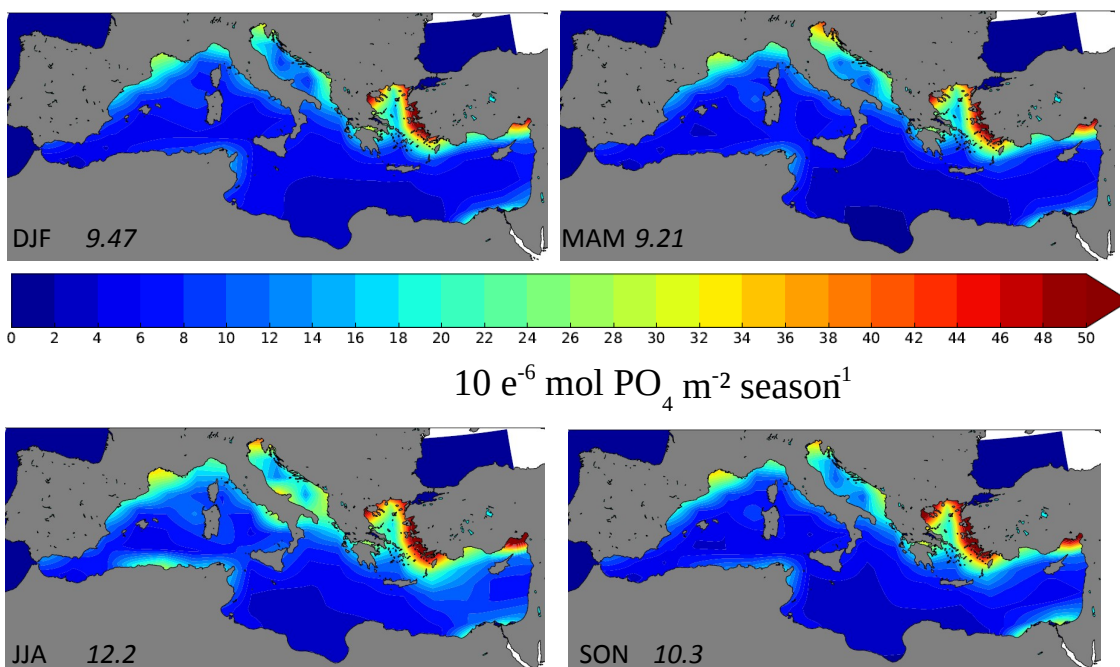


Figure 3. Total seasonal combustion-derived soluble phosphorus deposition (P_{comb} in $10^{-6} \text{ molPO}_4 \text{ m}^{-2} \text{ season}^{-1}$) over each season of the year 2005 (molar flux is calculated as mass flux/phosphorus molar weight) [from the LMDz-INCA model](#). Numbers on the maps are the average seasonal deposition fluxes over the basin in $10^{-6} \text{ molPO}_4 \text{ m}^{-2} \text{ season}^{-1}$.

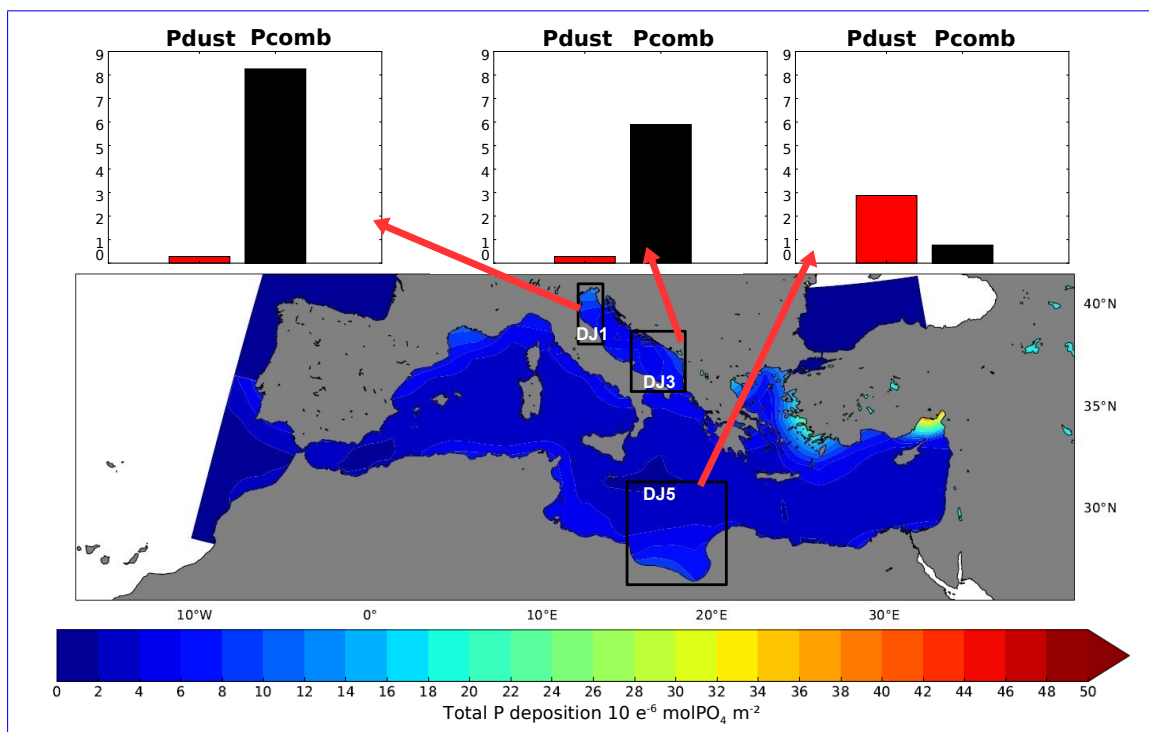


Figure 4. Map of total PO_4 deposition from both P_{dust} and P_{comb} ($10^{-6} \text{ molPO}_4 \text{ m}^{-2}$) for June 2005. **Bar plots** Red and black bars represent average PO_4 deposition (in $10^{-6} \text{ molPO}_4 \text{ m}^{-2}$) from the two sources in each framed area. The limits of the areas are described in Manca et al. (2004) and Figure 2. There is no atmospheric deposition modeled in the Marmara and Black Seas.

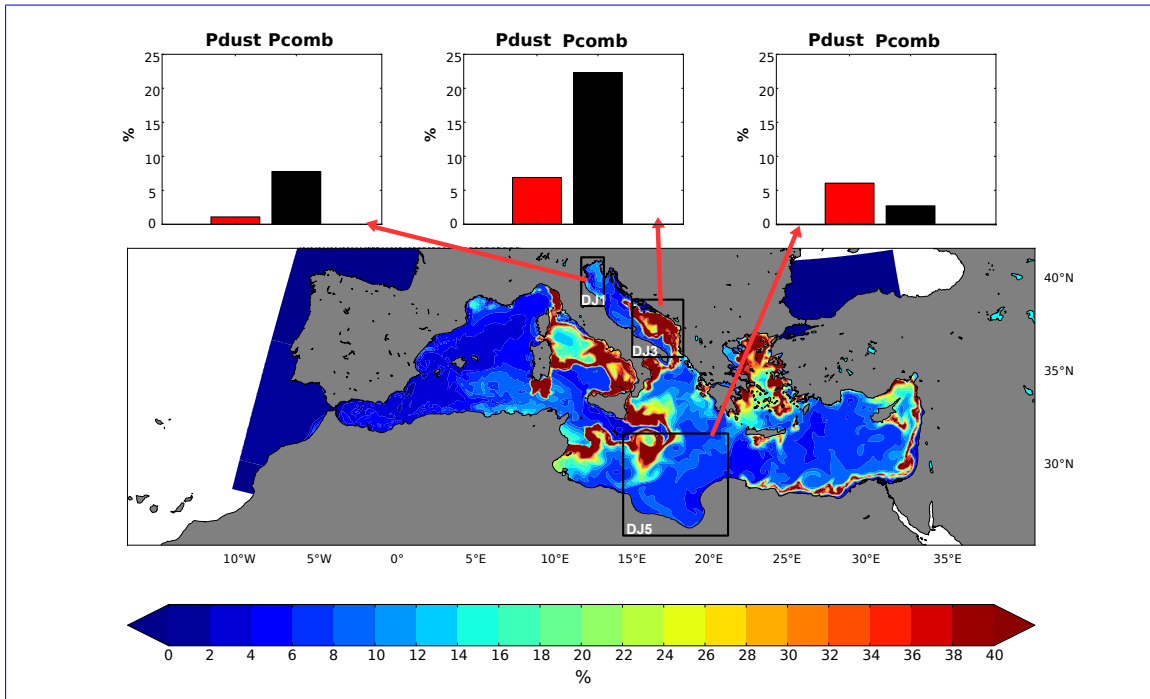


Figure 5. Map of maximal relative effects of total ($P_{dust}+P_{comb}$) deposition on PO_4 concentration in June 2005 (on a daily basis) on the surface Mediterranean phosphate concentration (0–10 m) in June 2005. Bar plots . The reference PO_4 concentration values are taken from the REF simulation without 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 2. There is no atmospheric deposition modeled in the Marmara and Black Seas.

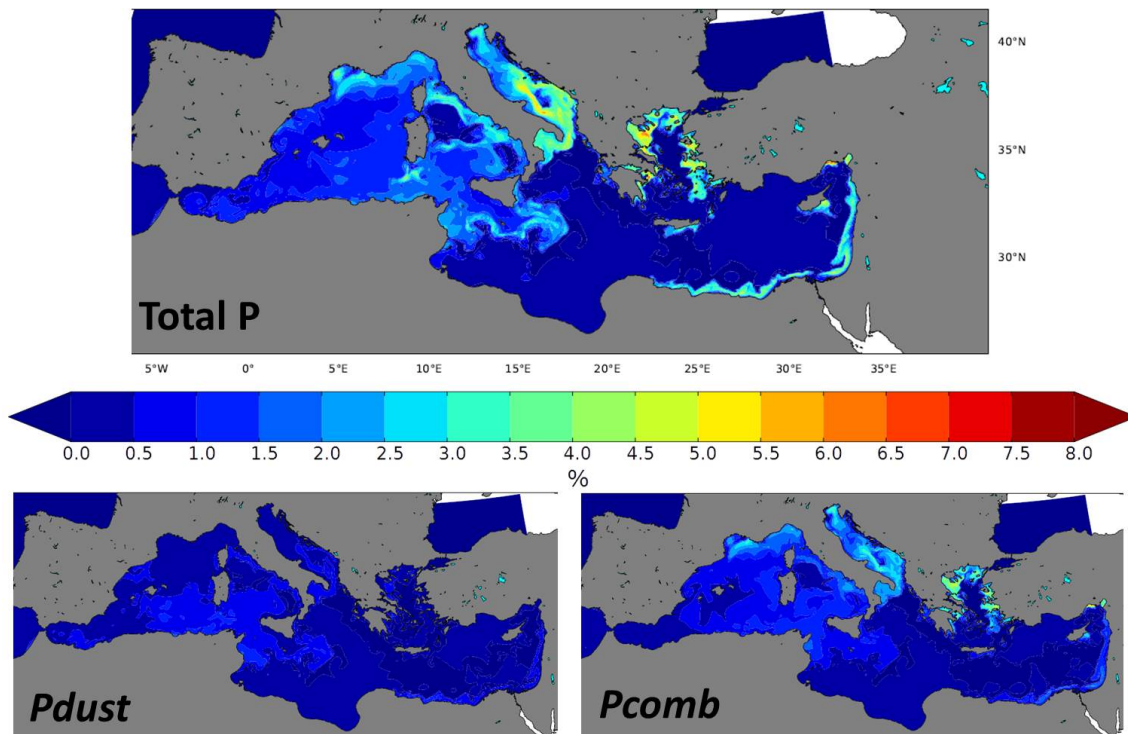


Figure 6. Average relative effects of total P, *Pdust* and *Pcomb* deposition on surface (0–10 m) chlorophyll *a* concentration for June 2005. There is no atmospheric deposition modeled in the Marmara and Black Seas.

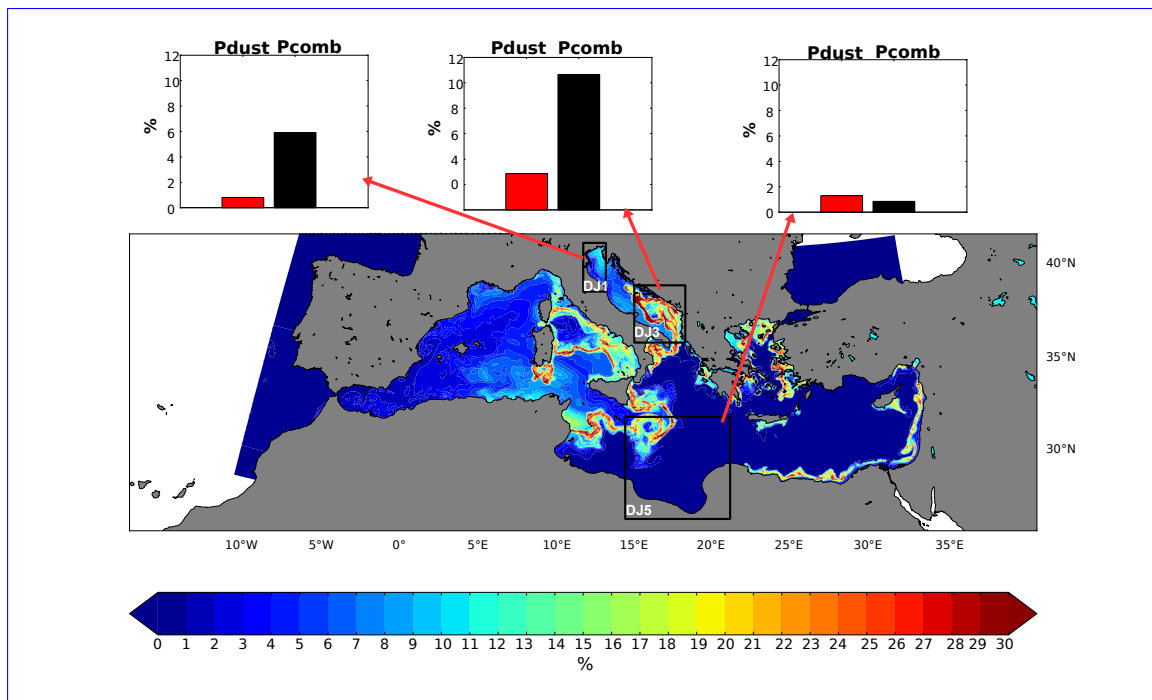


Figure 7. Map of maximal **daily**-relative effects of total ($P_{dust}+P_{comb}$) deposition on primary production in the surface Mediterranean (0–10 m) in June ~~2005–2005~~ (on a daily basis). The reference PO_4 concentration values are taken from the REF simulation without atmospheric phosphate deposition. 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.

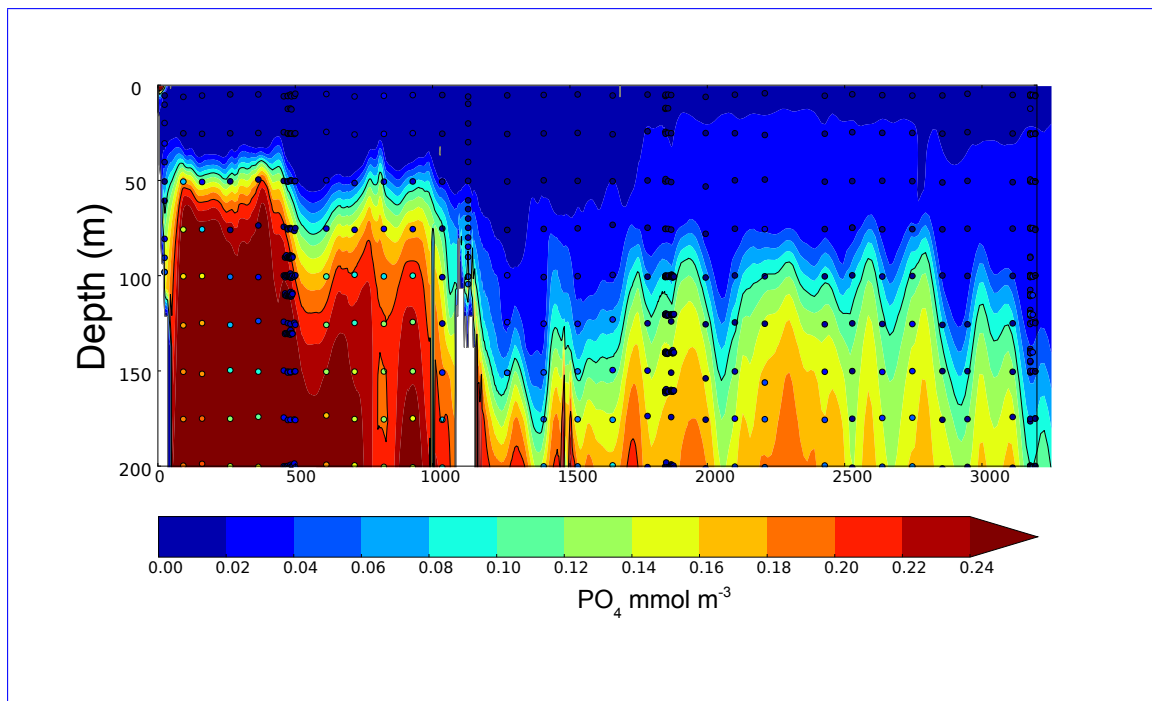


Figure A1. Evaluation of the model NEMOMED12/PISCES. Satellite (left) and modeled (right) surface chlorophyll a averaged over April (top) and June (bottom) 2005.

Nitrate (top) and phosphate (bottom) Phosphate concentrations computed by the NEMOMED12/PISCES model (background) and measured along the BOUM cruise (dots) (Moutin et al., 2012) between approximately Marseille and South of Cyprus.

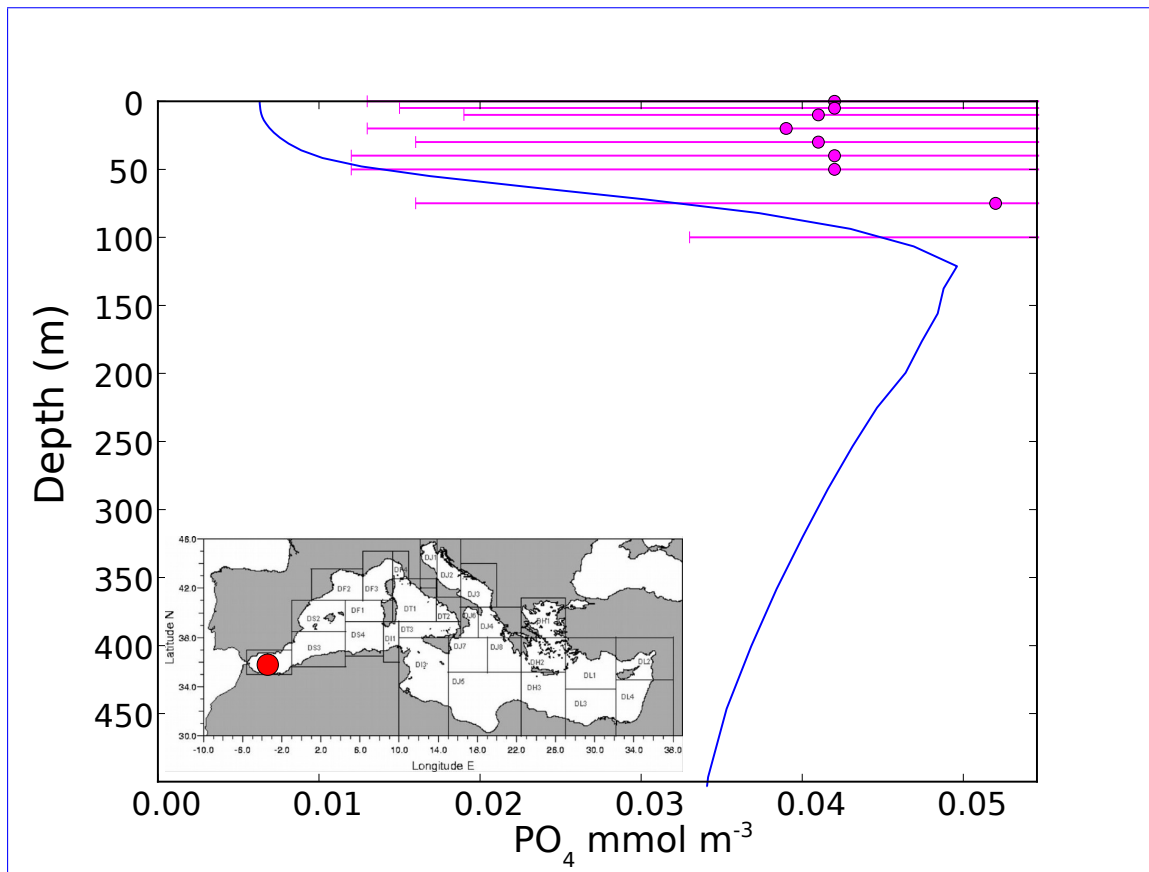


Figure A2. Vertical profile of average concentration in 2005 in the Algerian current region (see map).
Measurements and standard deviations are in pink, modeled values are represented by the blue line.

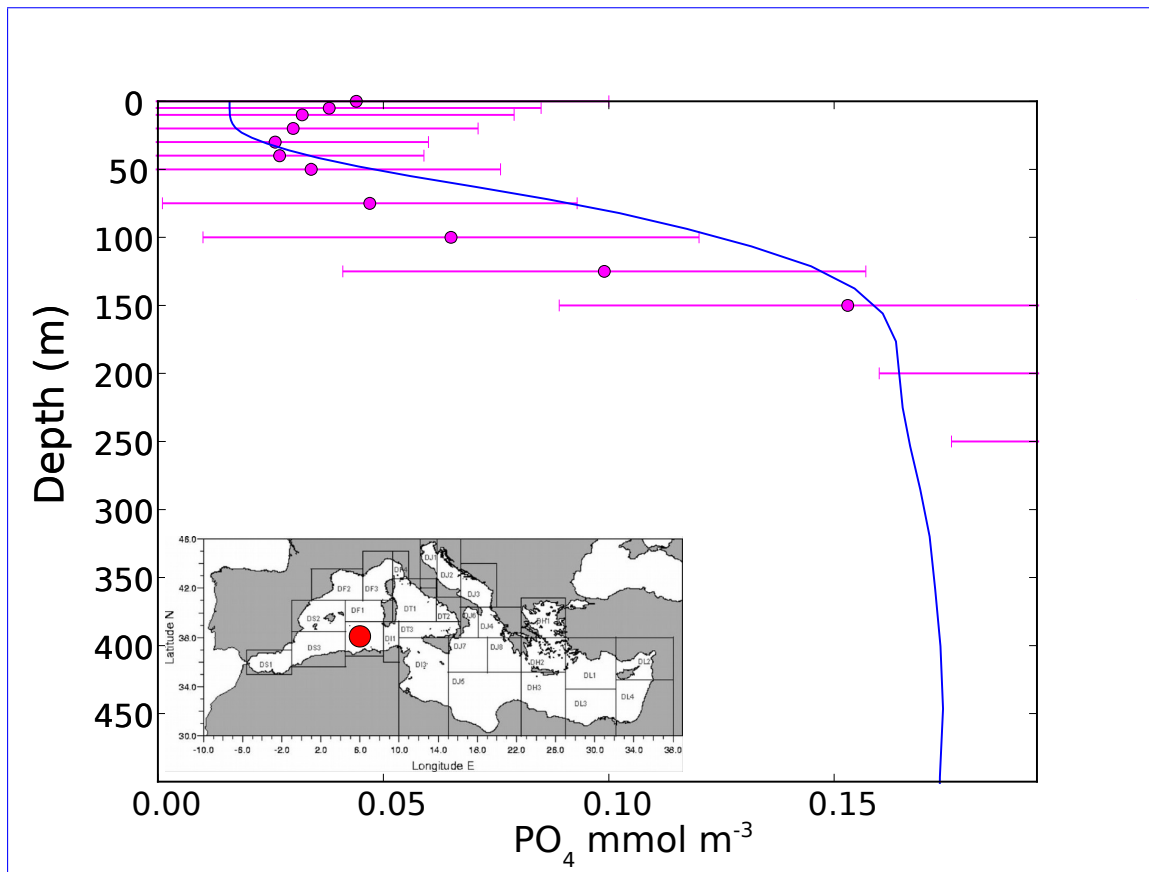


Figure A3. Vertical profile of average concentration in 2005 in the Alboran Sea region (see map). Measurements and standard deviations are in pink, modeled values are represented by the blue line.