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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 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 a high resolution regional coupled dynamical-–biogeochemical model of the Mediterranean Sea. 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 (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 (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 (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$_4$) sources and their respective fluxes variability and evaluate their impacts on marine surface biogeochemistry (phosphate concentrations, 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 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
25 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 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 nutri-
30 ent 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 ele-
ments 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 in many regions of the oceans. The main sources of atmospheric phos-
35 phorus 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
40 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
45 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
50 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
55 recipient of aerosols from a variety of different geographical sources. 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 in-
dustrialized 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
60 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–

65 –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

70 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

75 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

80 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).

85 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 west-

90 ern 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.

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. 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; microzoo-
plankton and mesozooplankton. PISCES is a Redfieldian model: the C/N/P ratio used for biology
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 from the estimation of Lud-
wig et al. (2009). Nutrient concentrations in the buffer zone are relaxed to the monthly climatology
of the World Ocean Atlas (WOA) (Locarnini et al., 2006).

The model is run in off–line mode like in the studies performed by Palmiéri et al. (2015), Guyen-
non et al. (2015), Ayache et al. (2015, 2016a, b) and Richon et al. (2017). PISCES passive biogeo-
chemical tracers are transported using an advection–diffusion scheme driven by dynamical variables
(velocities, pressure, mixing coefficients...) previously calculated by the oceanic model NEMO. Bio-
geochemical 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 and the main productive zones that are the Alboran Sea, the Gulf of Lions and most coastal
areas (see appendix).

2.2 Atmospheric deposition of phosphate

We use atmospheric deposition fields of total phosphorus from two different emission sources,
namely natural dust and combustion both simulated with the LMDz–INCA chemistry–climate global
model (Wang et al., 2015b). For the two sources, daily deposition fluxes are simulated globally for
2005. The form of deposited phosphate in the model is the same for all sources and is considered
to be soluble PO_4^{3-} which is the bioavailable form of phosphorus in PISCES (Aumont and Bopp,
2006).

The first source of phosphorus we include in this study is natural dust. In the atmospheric model,
natural 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 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 × 1.28° 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 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.

135   The second source of atmospheric phosphorus is combustion. Here, the term combustion en-
tails 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 emis-
sions from combustion due to anthropogenic activities are assumed constant throughout the year

140   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) es-
timated 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. The LMDz–INCA

145   modeled atmospheric P deposition rates 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

150   emitted P from combustion is bioavailable phosphate (*Pcomb*) (Longo et al., 2014).

Another important source of P aerosols in this region is 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.

155   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

160   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

165   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
170 (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
175 included. 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

180 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
185 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 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.,
190 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 1 shows the comparison between modeled and measured fluxes in terms of geometric means and standard deviations of monthly values of each
195 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

200 Table 1. The dust fluxes produced by the model are realistic. We observe a 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 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 is highly sensitive to model spatial resolution. The coarse resolution of LMDz may

205 significantly reduce the total dust emission in the model but also reduce surface winds and aerosol transport (see Discussion section hereafter).

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

210 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.

215 **3.2 Characterization of phosphate deposition from the different sources**

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

220 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).

225 The seasonal spatial distribution of *Pcomb* deposition is shown in Figure 3. 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 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

230 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 de-

235 position, 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

240 the dominant external source of phosphate for almost all Mediterranean regions (3.16 10^9 g PO$_4$ month^{-1} at the basin scale, see also 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 impacts from aerosol deposition in spite of the low fluxes, due to thermal stratification (Richon

245 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 riverine inputs and atmospheric deposition of P from combustion (Figure 3), the South

250 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

255 Myriokefalitakis et al. (2016).

By including the different sources of atmospheric phosphate separately, 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 to the Mediterranean basins. Our estimations of total aerosol contribution to PO$_4$ supply are slightly

260 lower than the literature values. This Table shows that the main atmospheric source of phosphate at the basin scale is *Pcomb*. 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 basin measurements from very few locations in Turkey and Greece. Vincent et al. (2016) report that recent desert dust deposition fluxes have

265 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

270 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 deposition on surface phosphate budgets

Atmospheric phosphorus deposition has different impacts 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

275 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 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 deposition from the two sources (combustion and dust) on surface PO_4 concentration for the month of June. The relative impacts of atmospheric

280 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.

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

285 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, espe-

290 cially 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 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 up-

295 welling (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.

300 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 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

305 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.

310   Figure 7 shows the relative impacts of P 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

315   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 deposition in this area.

320   *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

325   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).

330   *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.

335   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

340   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 2). *Pcomb* dominates *Pdust* contribution to PO$_4$ supply over the north-

345 ern 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

350 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

355 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.

360 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 ∼500 grid points covering the Mediterranean, and

365 *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. 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

370 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 overall average deposition estimation from the global model we use in this study appears much lower (0.122 ± 0.102 10^9 g month^{-1} over the Mediterranean in 2005 simulated with LMDz-–INCA and 0.568 ± 0.322 10^9 g month^{-1}

375 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
380   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 contri-
butions of *Pcomb* and *Pdust* may also vary. However, dust deposition fluxes available between 1997
385   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 Tur-
quety et al. (2014) indicates a $\pm$ 50 % annual variability, but it is impossible to give any inter–annual
390   variability of *Pcomb* deposition at present. Simulating longer time periods of atmospheric deposi-
tion 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–
395   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
400   combustion is, however, a satisfying first approach because it permits to highlight the high con-
centration 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
405   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.

410   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 com-
plex 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 vol-
canic sources respectively. However, most of the biogenic phosphorus is under the form of organic

415 phosphorus (DOP) that our model version does not include. Moreover, the composition of biogenic
aerosols and its solubility is still poorly constrained.

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, combus-

420 tion may be dominant over natural dust as an atmospheric source of phosphate for the Mediter-
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

425 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 relative
effects of each source are maximal in their areas of maximal deposition and can induce an enhance-

430 ment 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 emis-
sions, we showed that combustion-derived phosphorus deposition has effects on the biological pro-
ductivity. 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

435 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

440 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

445 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

450 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.

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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

The main characteristics of the surface chlorophyll *a* distribution are well simulated (Figure A1). We
460 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 observa-
465 tions. 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
470 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 campaign (Moutin et al., 2012) (Figure A2).

The NEMO/PISCES model reproduces the vertical structure of the nutrients with a gradient from
475 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.

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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 ($g\,m^{-2}\,yr^{-1}$) 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 1.

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 2. Relative atmospheric contribution (%) to total PO_4 supply in different sub–basins of atmospheric sources (atmospheric inputs/(atmospheric inputs + riverine inputs + Gibraltar inputs)). 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.

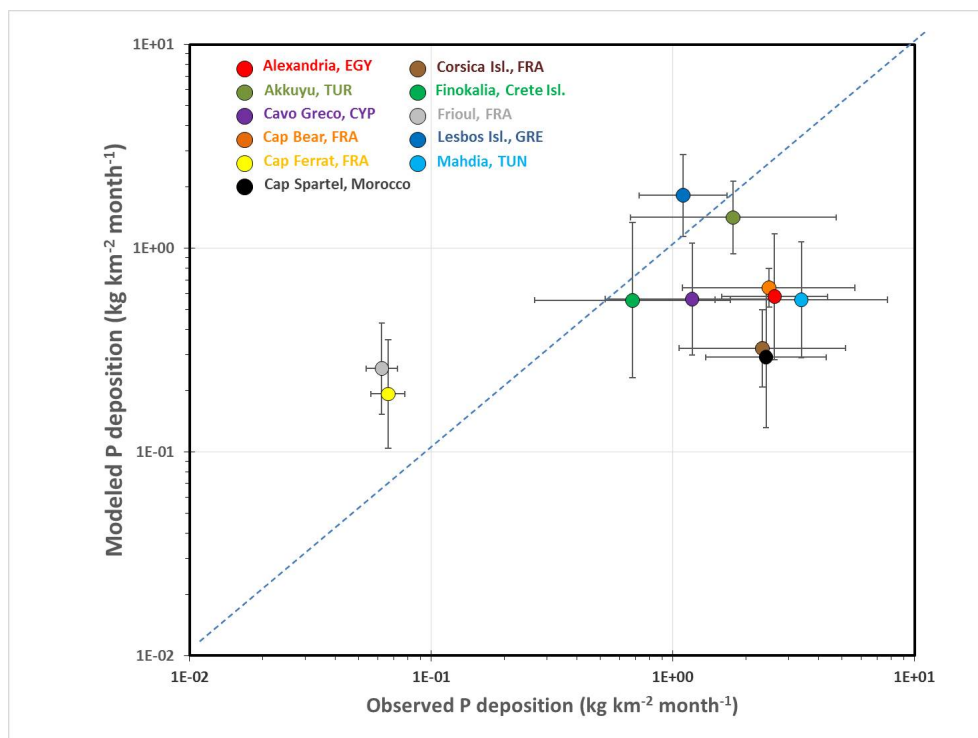



Figure 1. Comparison of modeled and observed monthly geometric mean of total P (*Pdust + Pcomb*) deposition fluxes at the 9 ADIOS stations (Guieu et al., 2010) and soluble PO_4 at Frioul and Cap Ferrat stations (de Fommervault et al., 2015). Each point is the geometric mean of monthly observed and modeled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

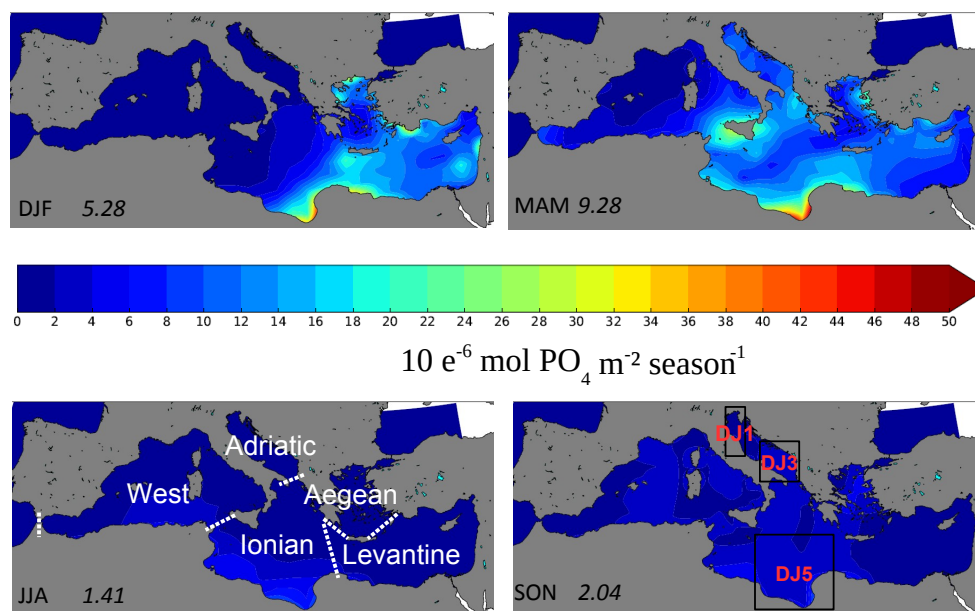



Figure 2. Total seasonal desert dust derived soluble phosphorus deposition (*Pdust*, in 10^{-6} molPO$_4$ m^{-2} season^{-1}) over each season of the year 2005 (molar flux is calculated as mass flux/phosphorus molar weight). Numbers on the maps are the average seasonal deposition fluxes over the whole 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.

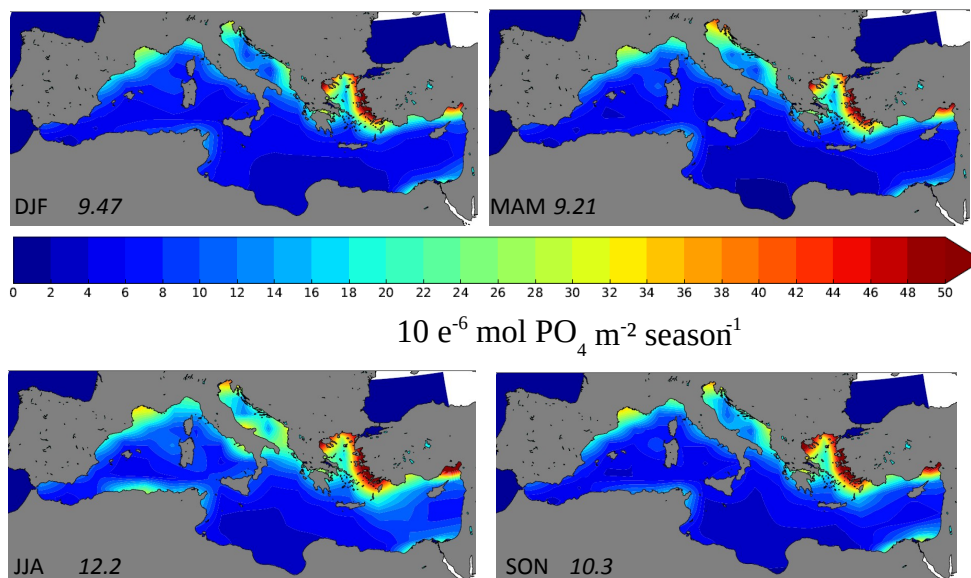



Figure 3. Total seasonal combustion–derived soluble phosphorus deposition (*Pcomb* in 10^{-6} molPO$_4$ m^{-2} season^{-1}) over each season of the year 2005 (molar flux is calculated as mass flux/phosphorus molar weight). Numbers on the maps are the average seasonal deposition fluxes over the basin in 10^{-6} molPO$_4$ m^{-2} season^{-1}.

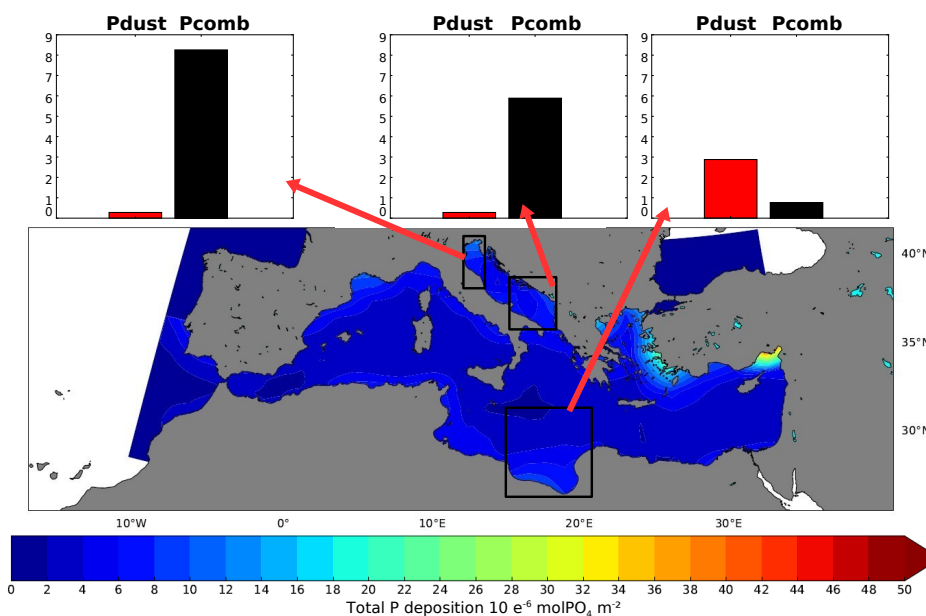



Figure 4. Map of total PO_4 deposition from both *Pdust* and *Pcomb* (10^{-6} molPO$_4$ m^{-2}) for June 2005. Bar plots represent average PO_4 deposition from the two sources in each framed area. The limits of the areas are described in Manca et al. (2004). There is no atmospheric deposition modeled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

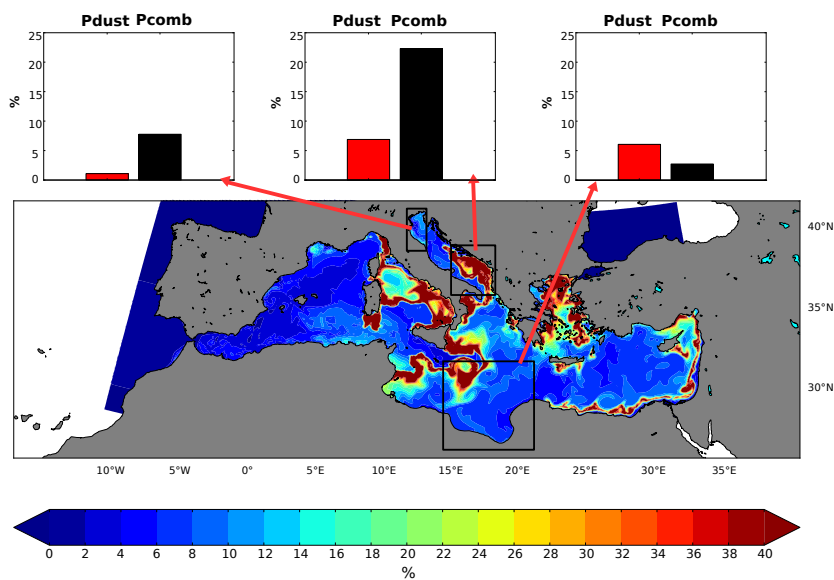



Figure 5. Map of maximal relative effects of total (*Pdust*+*Pcomb*) deposition on PO$_4$ concentration in the surface Mediterranean (0–10 m) in June 2005. Bar plots represent average maximal relative effects (%) within the framed areas of the two sources. The limits of the areas are described in Manca et al. (2004). There is no atmospheric deposition modeled in the Marmara and Black Seas.

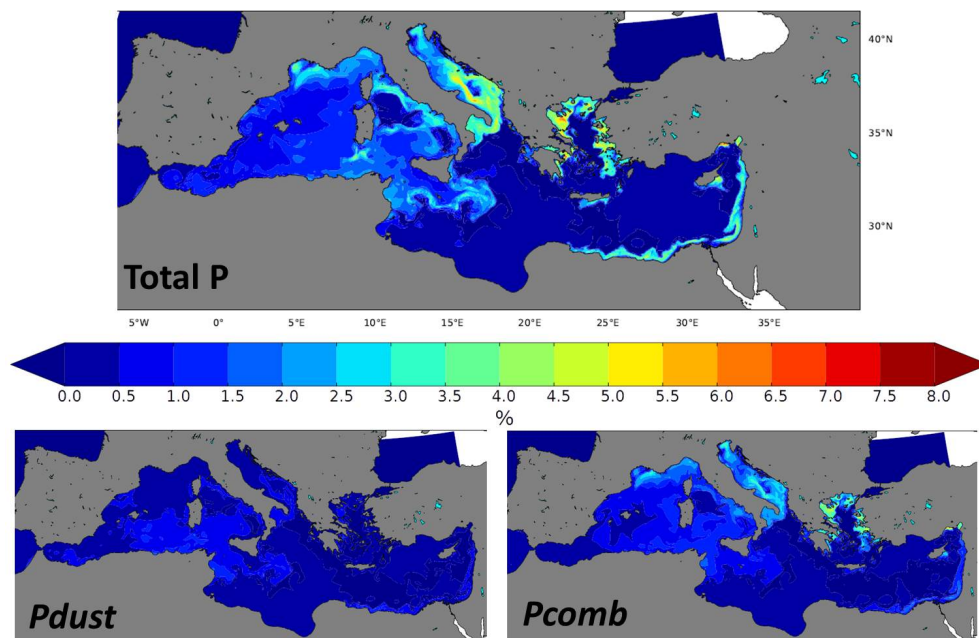



Figure 6. Average relative effects of total P, *Pdust* and *Pcomb* deposition on surface (0–10 m) chlorophyll *a* concentration for June 2005. There is no atmospheric deposition modeled in the Marmara and Black Seas.

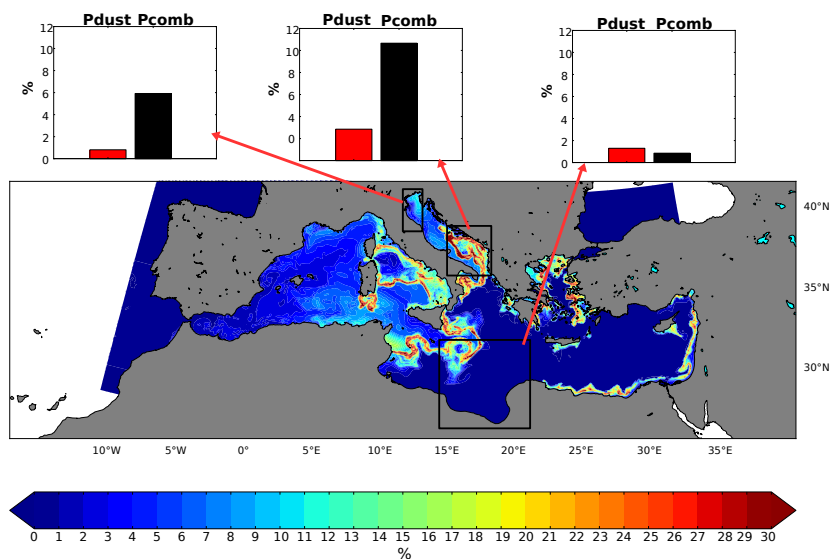



Figure 7. Map of maximal daily relative effects of total (*Pdust+Pcomb*) deposition on primary production in the surface Mediterranean (0–10 m) in June 2005. Barplots represent average maximal relative effects of each source (%) within the framed areas excluding land. The limits of the areas are described in Manca et al. (2004). There is no atmospheric deposition modeled in the Marmara and Black Seas.

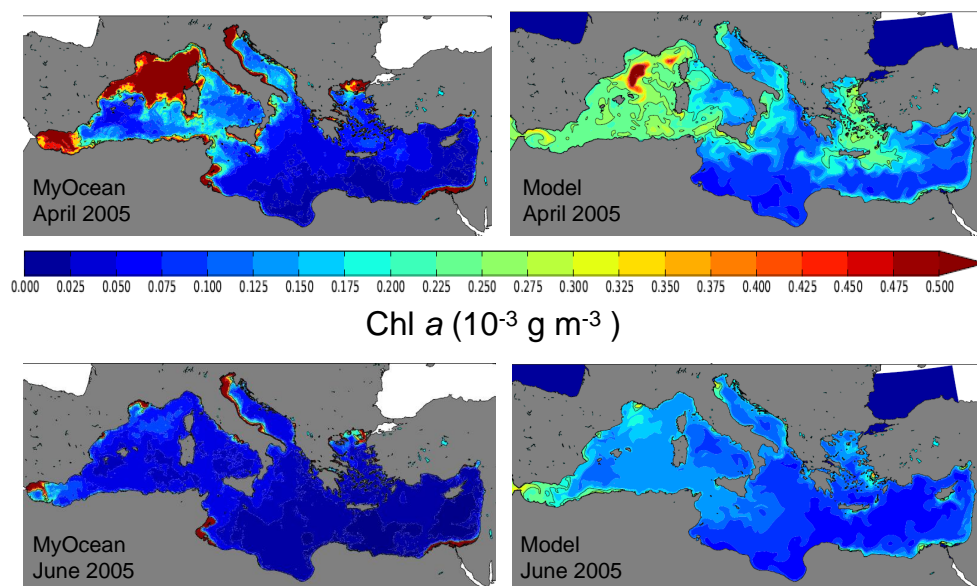
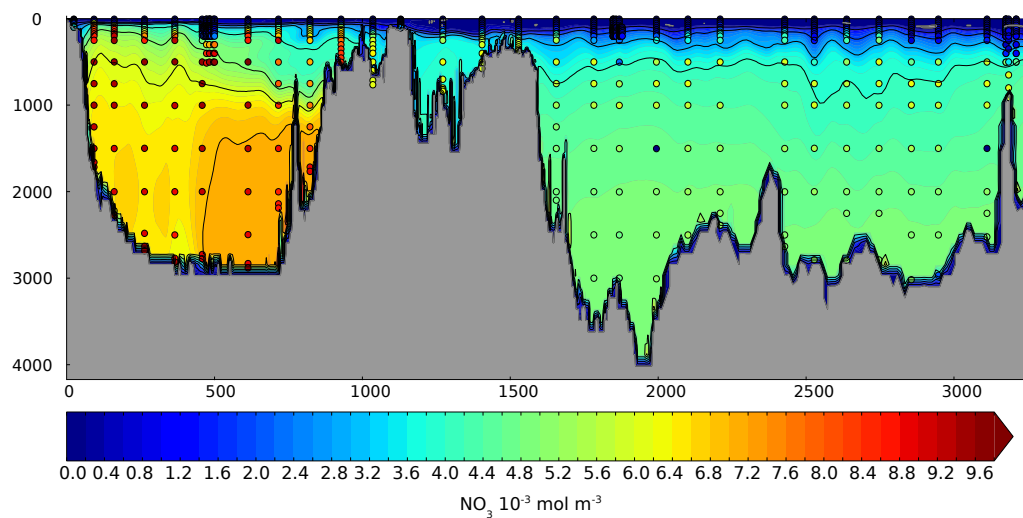



Figure A1. Evaluation of the model NEMOMED12/PISCES. Satellite (left) and modeled (right) surface chlorophyll *a* averaged over April (top) and June (bottom) 2005.

(a) NO$_3$ concentration alog the BOUM section

(b) PO$_4$ concentration along the BOUM section

Figure A2. Nitrate (top) and phosphate (bottom) 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.