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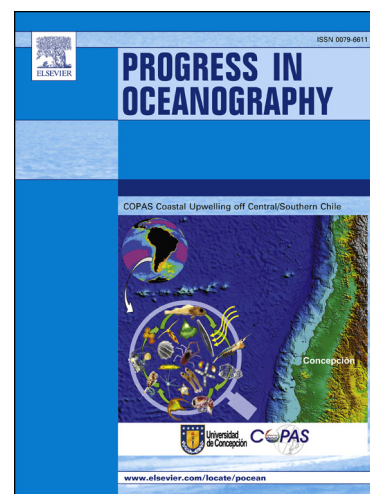
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# Modeling the impacts of atmospheric deposition of nitrogen and desert dust-derived phosphorus on nutrients and biological budgets of the Mediterranean Sea.

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## Abstract

Atmospheric deposition represents a significant source of nutrients at the Mediterranean basin scale. We apply aerosol deposition fields simulated from atmospheric models into the high resolution oceanic biogeochemical model NEMOMED12/PISCES with nutrient ratios used for plankton growth set to Redfield ratio. We perform 3 simulations to determine the impact of nutrients on productivity over the period 1997–2012: (i) without atmospheric deposition, (ii) with nitrogen deposition from anthropogenic and natural sources, and (iii) with deposition of both nitrogen (from anthropogenic and natural sources) and phosphate from desert dust. Time series of modeled deposition fluxes are compared to available measurements. This comparison with measurements shows that both variability and intensity ranges are realistic enough for our main purpose of estimating the atmospheric deposition impact on Mediterranean biogeochemical tracers such as surface nutrient concentrations, chlorophyll *a* and plankton concentrations. Our results show that atmospheric deposition is one of the major sources of nitrogen and phosphorus for some regions of the oligotrophic Mediterranean Sea. More than  $18 \text{ } 10^9 \text{ gN month}^{-1}$  are deposited to the whole Mediterranean Sea. This deposition is responsible for an average increase of 30 to 50 % in primary production over vast regions. Natural dust-derived deposition of phosphorus is sparser in space and time ( $0.5 \text{ } 10^9 \text{ g month}^{-1}$  on average over the entire basin). However, dust deposition events can significantly affect biological production. We calculate fertilizing effects of phosphate from dust to be low on average (6 to 10 %) but up to 30% increase in primary productivity can be observed during the months when surface water stratification occurs. Finally, these fertilizing effects are shown to be transmitted along the biological chain (primary production, Chl *a*,

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phytoplankton, zooplankton, grazing). We also perform a preliminary study on the maximal biological response of the Mediterranean by simulating extreme deposition events throughout the basin over a full year period. We show that nitrogen deposition effects observed in our long-term simulations (1997–2012) are close to maximal effects (i.e. those produced by high intensity deposition events) whereas dust-derived phosphate effects are substantially weaker than the effect on productivity reached when an extreme deposition event occurs.

*Keywords:* Mediterranean, biogeochemistry, aerosol, nutrient, deposition

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## 1. Introduction

The Mediterranean Sea is an oligotrophic, low-nutrient low-chlorophyll (LNLC) semi enclosed basin displaying strong spatial and seasonal variations in nutrient concentrations and chlorophyll production (Béthoux et al., 1992; Moutin et al., 2012; Mignot et al., 2014). Two deep basins are separated by the shallow Strait of Sicily (~200 m depth). Nutrient concentrations (P, N and Si) are higher over the western part, sustaining more primary productivity than the eastern part (Béthoux et al., 1998). In the western part, one of the main sources of nutrients is the Atlantic through the Strait of Gibraltar but major rivers such as the Rhône and the Ebro deliver nutrients to the Algero-Provençal basin. In the eastern part, coastal regions benefit from runoff from the major rivers such as the Pô and the Nile and from the Dardanelles Strait (Ludwig et al., 2009). During summer, the Mediterranean is highly stratified vertically and the extreme reduction of vertical exchanges that upwell nutrient-rich deep water to the surface layer causes the surface layer to be strongly depleted in nutrients (D’Ortenzio et al., 2005). During this period ranging from the end of May to the end of August, the main nutrient source for the remote parts of this LNLC region is atmospheric deposition of natural and anthropogenic aerosols (Herut et al., 2001; Carbo et al., 2005). Atmospheric deposition of aerosols from continental sources brings bioavailable nutrients to the surface Mediterranean Sea such as nitrate and phosphate that are limiting biological production over the major part of surface waters in the basin (Tanaka et al., 2011; Palmiéri, 2014) and also carbon in the form of sugars and carbohydrates (Theodosi et al., this issue). The main aerosol source to the pelagic Mediterranean Sea is Saharan soil dust which is a significant source of phosphorus (Ridame and Guieu, 2002). But important anthropogenic aerosols deposition fluxes have been measured over northern and eastern parts of the basin (Violaki et al., 2010; Izquierdo et al., 2012). External nutrient inputs into the Mediterranean Sea are still not fully constrained. River inputs have been quantified using integrated modeling approach (Ludwig et al., 2009), the input from the Gibraltar Strait and the Dardanelles Strait have been constrained by *in situ* observation (Béthoux et al., 2002), remote sensing and modeling approaches (Huertas et al., 2012; Kopasakis et al., 2012) while atmospheric contribution is still poorly quantified. Net exchange of  $\text{NO}_3$  and  $\text{PO}_4$  at the Gibraltar Strait are negative, which means there is an outgoing flux of these nutrients from the Mediterranean to the Atlantic. The different measurements and models to constrain nutrient inputs from coastal inputs and Gibraltar allowed to reproduce average seasonal climatologies of nutrient inputs. The nutrient fluxes estimates from rivers, Dardanelles and Gibraltar are

based on water discharge estimates which display more than 50% variability according to the season, the  
30 region and the estimation method (see Béthoux et al., 1998; Ludwig et al., 2009; Huertas et al., 2012). The  
few existing estimations of atmospheric nutrient deposition are based on local observations that are not nec-  
essarily representative of the whole basin. Guerzoni et al. (1997) estimated that from 2 to 46 g m<sup>-2</sup> year<sup>-1</sup>  
of Saharan dust are deposited over the Mediterranean Sea. The major part of this deposition is through wet  
deposition whereas dry deposition has a lesser contribution (see also Bergametti et al., 1989; Loÿe-Pilot and  
35 Martin, 1996; Guerzoni et al., 1999). Mass deposition of nutrients from atmosphere is equivalent or even  
dominant in some places such as the Ionian basin and other offshore areas like Corsica over riverine nutrient  
fluxes reaching the pelagic zone (Loÿe-Pilot et al., 1986).

Many studies have shown that dust deposition from the Sahara is an important source of dissolved trace  
elements to the upper layers of the sea, in particular of trace metals (e.g., Dulac et al., 1989; Guerzoni et al.,  
40 1999; Guieu et al., 2002). Unlike river inputs and other external sources of nutrients (e.g. Gibraltar and  
the Dardanelles) for which climatological nutrient fluxes were measured, atmospheric deposition is poorly  
constrained: measurements and monitoring studies of deposition fluxes are sparse. Monitoring studies reveal  
that dust deposition is highly variable in space and time and that most of the yearly amount of deposition  
occurs from a few episodic events (e.g., Bergametti et al., 1992; Loÿe-Pilot and Martin, 1996; Vincent et al.,  
45 2016). In order to constrain the seasonal cycle of dust deposition, measurements of deposition fluxes in  
different regions (several stations covering the East and West basins) over several years would be necessary.  
Measurements of natural dust deposition on coastal regions indicate that several grams per square meters  
can be deposited within hours under a dust storm (Ganor and Mamane, 1982; Loÿe-Pilot and Martin, 1996;  
Bonnet and Guieu, 2006; Guieu et al., 2010; Ternon et al., 2010). Rare monitoring network datasets are  
50 available in the Mediterranean region to constrain regional atmospheric deposition simulations (Guieu et al.,  
2010). The recent MOOSE (<http://mistrals.sedoo.fr/ChArMEx/>) and ChArMEx/CARAGA (Vincent et al.,  
2016) deposition sampling networks give new constrains on deposition rates in the northwestern and western  
part of the coastal Mediterranean. In addition to the uncertainties on the deposition rate, the chemical com-  
position of the aerosols deposited and their transformations in seawater is not yet fully understood. Once  
55 lifted by winds, aerosols experience various conditions in the air column (UV, irradiation, processing by cloud  
droplets, mixing). These conditions can modify the composition and size distribution of soil dust particles  
(Desboeufs et al., 2001).

To date, few studies have investigated the consequences of dust deposition on the nutrient budgets and the  
biological activity of the Mediterranean Sea in realistic conditions (mesocosm or *in situ* studies based on  
60 autochthonous water masses and species). Experimental work such as the DUNE project aimed at study-  
ing the response of mesocosms to the addition of artificially aged Saharan dust, reproducing the conditions  
of natural dust deposition events (Guieu et al., 2010, 2014a). Results showed an increase in chlorophyll *a*  
production within a few hours after dust deposition. The results also showed that primary producers and  
primary consumers (small phytoplankton, diatoms and zooplankton) displayed a rather low increase in abun-

65 dance. Small phytoplankton ( $< 3 \mu\text{m}$ ) and bacteria were more impacted by dust deposition than primary producers resulting in a more heterotrophic natural assemblage. Another conclusion was that the sinking of POC through DOM/dust ballast effect and aggregation processes was favored under dust deposition event decreasing the residence time of nutrients in the euphotic layer by several days (Guieu et al., 2014b).

In the context of climate change, models provide a useful tool to investigate the effects of changing environmental conditions on the biogeochemical cycles of the oceans (Steinacher et al., 2010; Bopp et al., 2013). The Mediterranean basin is thought to be highly sensitive to climate change impacts due to the short residence time of the waters (less than 100 years) and the high impact of anthropogenic activities. The Mediterranean Sea has been described as a miniature ocean and an ideal laboratory for impact studies (The Mermex Group, 2011). Coupled physical–biogeochemical models allow us to represent the evolutions of the different physical and biological tracers on multidecadal time scales. They give the opportunity to disentangle the influences of various external conditions on processes such as primary production or carbon export. Guieu et al. (2014b) performed numerical biogeochemical experiments in the global ocean by simulating intense episodic atmospheric deposition events. Their results suggest that using high temporal resolution atmospheric deposition forcing induces a more important biological response from the models, compared to previously used large scale and long time averaged deposition inputs. To our knowledge, the present work is the first to use high-resolution long-term (i.e. multi-year) modeling to study the impacts of Saharan dust and anthropogenic nitrogen deposition on the Mediterranean basin biogeochemistry. The explicit representation of atmospheric nutrient source when modeling the Mediterranean Sea provides a valuable approach to apprehend the effect of the natural and anthropogenic aerosol deposition on biogeochemical cycles.

85 In this study, we aim at simulating the impacts of atmospheric aerosol deposition on the Mediterranean Sea, quantifying its contribution on marine nutrient budgets of the basin and investigating its consequences for biological production at the regional and seasonal scale. To this end, we used the 3-D bio-geochemical model PISCES (Aumont et al., 2015) with a high resolution ( $1/12^\circ$ ), with physical oceanic forcing from simulations made with NEMO-MED12 (Hamon et al., 2016), atmospheric deposition forcing of nitrogen (nitrate and ammonium) from both natural and anthropogenic sources modeled with LMDz-INCA (Hauglustaine et al., 2014) and phosphate inputs from Saharan dust modeled with ALADIN-Climate (Nabat et al., 2015). Our methodology is presented in section 2. Section 3 presents results in terms of seasonal variability of the deposition, impact on nutrient budgets, and spatial variability of the biological response. In section 4, we discuss our new basin scale assessment of nutrient budgets including atmospheric deposition, the variability of responses to this deposition, the transmission of impacts along the biological chain and the maximal potential response of the Mediterranean surface waters to idealized very intense aerosol deposition events occurring monthly over a full year time.

## 2. Methods

### 2.1. Ocean model

100 We use a regional configuration of the oceanic model NEMO (Madec, 2008) over the Mediterranean Sea (MED12 configuration). The MED12 grid is the standard irregular ORCA grid with a  $1/12^\circ$  horizontal resolution stretched in latitude, equivalent to a resolution ranging between 6 km and 8 km at respectively  $46^\circ\text{N}$  and  $30^\circ\text{N}$ . This resolution is so-called "eddy resolving" because it resolves important features of the Mediterranean circulation that are fine-scale structures and small eddies. In the vertical, 75 unevenly spaced  
105 levels are used ranging from 1 m at the surface, to 134 m at the bottom. The physical parameters are fully described in Hamon et al. (2016).

This physical model is coupled to the biogeochemical model PISCES (Aumont and Bopp, 2006). PISCES is constructed on the assumption that phytoplankton growth is directly limited by the external availability of nutrients (Monod, 1958). The model has 24 compartments including 2 phytoplankton (nanophytoplankton  
110 and diatoms) and 2 zooplankton classes (microzooplankton and mesozooplankton). Phytoplankton growth can be limited by five different forms of nutrients: nitrate, ammonium, phosphate, silicate and iron. A full description of PISCES together with its last developments can be found in Aumont et al. (2015). The regional Mediterranean configuration of the coupled model NEMOMED12/PISCES was developed by Palmiéri (2014). In this model, nutrient ratios in the plankton cells are fixed following the revised Redfield ratio for  
115 C:N:P of 122:16:1 (Takahashi et al., 1985). Bacterial concentration is only indirectly derived from zooplankton concentrations and is not explicitly modeled.

Initial nutrient concentrations in the Mediterranean come from the SeaDataNet database (Schaap and Lowry, 2010) and nutrient concentrations in the buffer zone (between  $11^\circ\text{W}$  and the Strait of Gibraltar) are prescribed from World Ocean Atlas (WOA) (Locarnini et al., 2006). Coastal and riverine nutrient and freshwater  
120 fluxes are derived from Ludwig et al. (2009), the Dardanelles' Strait is included in these forcings and treated as a river. From now on, the term 'rivers' refer to both river mouth and the Dardanelles Strait. Salinity and temperature are initialized from the MEDATLAS II climatology (Fichaut et al., 2003). Temperature, salinity and nutrients are relaxed to seasonal climatologies with a time constant of one month in the buffer zone. This short relaxation time is necessary to keep the values in the buffer zone close to reality. Because  
125 of the small size of the buffer zone, water circulation in this area would otherwise induce highly unrealistic fluxes.

The biogeochemical model is ran in its offline mode, similar to what was done in the studies by Palmiéri et al. (2015) ; Ayache et al. (2015a) ; Ayache et al. (2015b) ; Guyennon et al. (2015) and Ayache et al. (2016). Biogeochemical quantities being passive tracers, they are transported during the simulation with PISCES  
130 from an advection-diffusion equation with dynamical fields (velocities, mixing coefficients...) beforehand calculated by NEMO dynamical model.

## 2.2. Atmospheric deposition forcings

In this study, we focus on the deposition of nitrogen and phosphorus, which are well-known to be limiting elements in Mediterranean waters (Tanaka et al., 2011). Using available model deposition gridded fields, we consider atmospheric deposition of N from both natural and anthropogenic sources and of P from desert dust. We make no distinction in terms of solubility between particle size or deposition type (dry vs. wet). The 16-year period over which the deposition fields were available (1997–2012) is our study period. Spatial resolution of the dust deposition fields is  $0.5^\circ$  ( $135 \times 189$  total grid points, 1007 grid points over the Mediterranean Sea). Spatial resolution of the nitrogen deposition fields is  $1.9^\circ$  in latitude and  $3.75^\circ$  in longitude ( $144 \times 143$  grid points in total,  $\sim 120$  grid points over the Mediterranean). The deposition fields are spatially interpolated to the  $1/12^\circ$  grid of the NEMOMED12 model ( $567 \times 264$  grid points in total,  $> 55000$  grid points over the surface Mediterranean Sea).

### 2.2.1. Nitrogen deposition

The nitrogen deposition forcing is derived from a long-term simulation performed with the global chemistry-climate model LMDz-INCA (Hauglustaine et al., 2014). Meteorological data for nudging come from the European Centre for Medium-Range Weather Forecasts (ECMWF) reanalysis. We use monthly files ranging from 1997 to 2012 of wet and dry deposition of inorganic nitrate and ammonium. These species come from different natural and anthropogenic sources described in Wang et al. (2015): e.g., agricultural emissions, fossil fuels, biofuels, wildfires, natural soil and oceans. The nitrogen deposition fields read in PISCES consist of the sum of nitrogen from  $\text{NO}_3$  and  $\text{NH}_4$  deposition coming from LMDz-INCA. These species are not directly emitted in the atmosphere but are secondary products from chemical transformations of  $\text{NO}_{(X)}$  and  $\text{NH}_3$  emitted from soil, combustion, ocean and other natural sources. Deposition of  $\text{NO}_{(X)}$  is not included in our simulations. The chemical reactions in the atmosphere producing  $\text{NH}_4$  and  $\text{NO}_3$  are dependent on temperature and relative humidity (see Hauglustaine et al. (2014) for detailed chemical reactions). A limitation of our approach is that we do not take into account  $\text{NO}$ ,  $\text{NO}_2$ ,  $\text{N}_2\text{O}$ ,  $\text{NH}_3$  deposition because these species are not explicitly represented in the PISCES model. For the rest of the study, we refer to "N deposition" as the sum of  $\text{NO}_3 + \text{NH}_4$  which are both considered 100% soluble independently of atmospheric conditions and chemical origin.

### 2.2.2. Phosphorus deposition from desert dust

The desert dust deposition fields used in this study represent desert dust particles coming from the Sahara and Middle East. They have been lifted by winds and sandstorms, then transported and deposited over the Mediterranean basin. This dataset has been obtained from a high resolution ( $50 \text{ km}$ ) atmospheric simulation, carried out with the regional climate model ALADIN-Climate. This model (Nabat et al., 2015) has an interactive aerosol scheme, including the representation of dust particles in three size bins ( $0.01\text{--}1.0$ ,  $1.0\text{--}2.5$  and  $2.5\text{--}20.0 \mu\text{m}$ ) as well as the associated processes (emission, transport, sedimentation, dry deposition, and wet deposition in and below the clouds). Deposition output at a 3-hour frequency have been saved from



a multi-annual simulation (1980–2012) for use in the present study. Note that in this ALADIN–Climate simulation based on a regional domain including the Mediterranean basin and the surrounding dust–source regions, lateral boundary conditions come from the ERA–Interim reanalysis (Dee et al., 2011).

170 We implemented these natural dust forcings in PISCES for the period 1997 – 2012 with the hypothesis that mineral soluble fraction of dust in P, Si and Fe is fixed regardless of the environmental conditions. We chose to focus on P deposition from dust with the hypothesis that it contains 735 ppm of phosphorus among which 10% is in soluble phosphate form (Anderson et al., 2010).

Desert dust is not the only atmospheric source of bioavailable phosphorus for the Mediterranean region.

175 Combustion processes related to energy consumption and biomass burning are also an important source of soluble aerosol material (Mahowald et al., 2008; Myriokefalitakis et al., 2016). These sources are estimated to account on the global scale to 5 to 15% of total  $\text{PO}_4$  deposition. But Mahowald et al. (2008) estimated that anthropogenic phosphorus may account for more than 50% of the deposition over oligotrophic areas. We focus here on phosphate from natural dust and do not account for anthropogenic deposition.

### 180 2.3. Simulation set up

Figure 1 sketches our simulation set-up. We compare 3 different simulations of the period 1997-2012 in order to investigate separately the impacts of phosphate and nitrogen atmospheric deposition. The simulations are summarized in Table 1. The reference (“No dep”) simulation is run without any nutrient deposition, only with physical forcing and with external nutrient inputs (biogeochemical forcing) from rivers and the 185 Gibraltar strait. One simulation (“N”) considers only atmospheric deposition of nitrogen, and the last one (“N+ $\text{PO}_4$ ”) considers both nitrogen and dust–derived phosphate deposition. We stored monthly averages of the biogeochemical variables during the whole historical runs. The results we show are the separate effects of nitrogen and dust–derived phosphorus deposition by difference between the simulations.

190 The initialization state was obtained from a prior 15–year simulation over 1982–1996 without atmospheric deposition as well, itself initialized by a 30–year spin–up performed in order to set all biogeochemical variables to equilibrium. This spin–up was done by running six 5–year loops using physical and biogeochemical forcings from 1982 to 1986.

195 We did an additional one year simulation with dust–derived phosphate deposition only (“ $\text{PO}_4$  only”). This simulation was designed as a test whether the effects of nitrogen and phosphate depositions were independent. We observe the same effects of phosphate deposition in the “ $\text{PO}_4$  only” simulation than in the “N+ $\text{PO}_4$ ” simulation (not shown). This finding indicates that the effects of phosphate deposition can be derived by difference between the “N+ $\text{PO}_4$ ” and “N” simulations.

200 *2.4. Sensitivity test: Maximum biological response to intense deposition events*

In order to quantify the maximum potential biological response to atmospheric deposition, we performed additional simulations following Guieu et al. (2014b) which consist in simulating spatially uniform intense deposition of N and PO<sub>4</sub> at different periods of the year. We conducted one-year simulations with spatially uniform intense deposition fluxes occurring the first day of every month during 24 hours. After the first  
205 day, aerosol deposition was kept at zero until the end of the month. Based on recorded fluxes during high deposition events around the Mediterranean, we chose to apply uniform deposition of 40 10<sup>-3</sup> g m<sup>-2</sup> day<sup>-1</sup> of NO<sub>3</sub> and the same amount of NH<sub>4</sub> for "N" and the phosphate from 10 g m<sup>-2</sup> day<sup>-1</sup> of desert dust in the "N+PO<sub>4</sub>" test. In the ALADIN Climate model, deposition fluxes of dust over 24 h can be locally as large as 10 to 12 g m<sup>-2</sup> day<sup>-1</sup> in the 1997–2012 period.

210 The fluxes applied represent large scale extreme deposition events that have been observed in some Mediterranean regions. The likelihood of such events varies according to the region and the season and event covering the entire basin at once are highly unlikely. These idealized fluxes aim at determining the most sensitive regions and season to atmospheric deposition. Moreover, simulating these episodic uniform events monthly allows us to study the potential maximum response of all regions of the Mediterranean under different phys-  
215 ical and biological conditions. We assume that the maximum relative change in surface biological tracer concentrations (nutrients, chlorophyll *a*, plankton biomass) we observe is the maximum potential biological response to an intense atmospheric deposition event.

### 3. Results

#### *3.1. Evaluation of the modeled marine nutrients and surface chlorophyll *a**

220 The NEMO–MED simulation used in this study to force the PISCES model is described and validated in Hamon et al. (2016) (simulation named NM12–FREE). The simulation reproduces well the general circulation of the water masses and its variability, despite some shortcomings pointed out by the authors, such as too slow transports through the Strait of Gibraltar (by about 0.1 Sv), a slightly too weak circulation and mesoscale activity in the Western basin (Algerian Current, Northern Current), and a temporal drift in the  
225 heat and salt contents in the intermediate layer all over the Mediterranean Sea, leading to too warm (about +0.25°C) and too salty (about +0.1 psu) intermediate waters at the end of the period. The ability of the model to reproduce the general circulation of the water masses was also evaluated in a similar configuration by Ayache et al. (2016) and the representation of biological parameters in a Mediterranean configuration of PISCES was evaluated by Palmiéri (2014).

230 We evaluate the outputs from our reference simulation using measurements from different campaigns. Our reference simulation reproduces reasonably well observations at the basin scale. The model captures the main characteristics of the nutrients distributions observed along the BOUM cruise section (Moutin et al., 2012), producing a realistic gradient from West to East and a vertical structure showing subsurface maximum of concentrations due to Levantine Intermediate Water (LIW) (Figure 2). However we can identify

235 some shortcomings. The model underestimates the concentrations in the western basin and produces a nutri-  
cline too smooth compared with observations (See inserts in Figure 2). The model also simulates a realistic  
biogeochemical production, as diagnosed from the surface Chl *a* distribution (Figure 3). The model produces  
the salient West-to-East gradient in Chl *a* seen by satellite, with the main productive regions located in the  
western basin, in the Alboran Sea influenced by the nutrient rich water masses flowing through Gibraltar, and  
240 in the Gulf of Lions where winter vertical mixing brings nutrient rich water masses to the surface. We note  
however a severe underestimation of about 50% in the coastal areas. Satellite estimations in coastal turbid  
waters are known to be highly uncertain. We compared the average chlorophyll concentration measured at  
the DYFAMED station for the period 1997–2005 (Marty et al., 2002; Faugeras et al., 2003) with our model  
outputs. We found that the average measured chlorophyll *a* in the top 200 meters is  $0.290 \pm 0.177 \text{ } 10^{-3} \text{ g m}^{-3}$   
245 and the average model value is  $0.205 \pm 0.111 \text{ } 10^{-3} \text{ g m}^{-3}$ . The model also reproduces well the chlorophyll  
vertical distribution observed in summer at the DYFAMED station with a deep chlorophyll maximum around  
50 to 80 m depth. However, the vertical stratification is too weak before the end of May in the model. As a  
consequence, we do not produce the deep chlorophyll maximum in spring as in the DYFAMED observations.

250  
Nutrient inputs at Gibraltar represent an important source for the surface Alboran Sea (see Figure 3),  
but net exchanges of  $\text{NO}_3$  and  $\text{PO}_4$  are negative (outflow of nutrients to the Atlantic) and mass inputs of ni-  
trogen and phosphorus through the Strait of Gibraltar are orders of magnitude lower than inputs from coastal  
and atmospheric sources. We computed the average concentrations in the inflowing and outflowing water  
255 masses at Gibraltar in our 3 simulations and compare them to values from Huertas et al. (2012) (Table 2).  
The model reproduces the net outflows of nutrients. However the concentrations of nitrate and phosphate  
in the outflowing waters are too low. We trace back this discrepancy to the general underestimation of the  
concentrations in the western basin (Figure 2).

260 In conclusion, this model reproduces well the main structures, in spite of some shortcomings that we tried  
to point out. We consider this model suitable for the scope of the biogeochemical study which is the main  
purpose of this work.

### 3.2. Evaluation of the modeled deposition fluxes

In the global ocean, atmospheric deposition occurs mainly through wet deposition. Dry deposition ac-  
265 counts for less than 20% of total deposition in the majority of Mediterranean areas (Migon et al., 2001;  
Izquierdo et al., 2012; Violaki et al., this issue). Measurements of atmospheric deposition over the Mediter-  
ranean basin are sparse and mainly from coastal areas. We evaluated nitrogen and desert dust deposition  
fluxes with the few time series available in several stations around the Mediterranean in the frame of the  
CARAGA (see Vincent et al. (2016) for measurements analysis and network presentation) and MOOSE  
270 projects (Figure 5). Figure 4 shows the comparison of N deposition fluxes at the Frioul and Cap Ferrat

stations of the MOOSE network and modeled data. The MOOSE (Mediterranean Ocean Observing System for the Environment) network is a French sampling network designed to assess the spatial and temporal distribution of atmospheric nutrient inputs in the northwestern Mediterranean (see de Fommervault et al., 2015). The variability of measured fluxes is high (filters for measurements are collected weekly for dust; 275 for nitrogen, dry deposition was sampled every two weeks and wet deposition was collected after each rain event), moreover, there are some missing measurement periods in 2007 and 2008 at Cap Ferrat (Figure 5d). We also observe that the peaks of deposition recorded in the observations do not coincide to the simulated peaks and are more intense in the measurements than in the model. The modeled fluxes represent the average deposition over a month in the grid cell corresponding to the station location. Hence, measurements that 280 represent the fluxes at a station display a higher temporal variability. Overall, both the annual and monthly modeled fluxes are in good agreement with the measurements.

Figure 5 shows that the simulated desert dust deposition fluxes at Cape Corsica are very low and only one significant peak ( $> 0.2 \text{ g m}^{-2} \text{ week}^{-1}$ ) of dust deposition was recorded during the sampling period. 285 This event is well reproduced by the model in spite of a large overestimation of the flux. Dust deposition events at the Frioul, Lampedusa and Majorque stations appear more frequent both on the observations and the model. The amplitude of the deposition fluxes is correctly reproduced by the ALADIN–Climate model. We also compared the spatial variability of our dust deposition model to the measures made during the ADIOS campaign from June 2001 to May 2002 (Guieu et al. (2010) and Table 3). Modeled fluxes generally 290 overestimate the observations and the modeled variation coefficients vary less from station to station than the measured variation coefficients. The gap between model and observations is larger in the eastern part of the basin with up to 5 times overestimation of deposition flux at the Cyprus station whereas fluxes in the western part, especially in France and Morocco, are well estimated (Table 3). However, Ridame et al. (1999) showed that the bulk sampling method used during this campaign tends to underestimate the deposition fluxes. The 295 model deposition estimates fall in the range of observations in terms of both magnitude and variability, so we consider them as a realistic forcing for our modeling study. Indeed, we do not intend to reproduce the exact evolution in space and time of atmospheric deposition (we would need a coupled ocean–atmosphere model for this purpose), but we rather attempt to use a realistic estimation of the magnitude and variability of deposition to estimate its consequences on the Mediterranean biogeochemistry.

### 3.3. Seasonal variability of modeled atmospheric inputs

305 Figures 6 and 7 show the seasonal variations of both N and P atmospheric deposition forcings. Deposition fluxes of  $\text{NO}_3$  and  $\text{NH}_4$  display similar seasonal cycles and spatial distributions. Figure 6 shows the seasonal

mass flux of  $\text{NO}_3 + \text{NH}_4$  averaged over 1997 to 2012. Inter-seasonal spatial variability of this deposition is low, most of the mass being deposited in the northern part of the basin, mainly in the Adriatic and Tyrrhenian Seas. We observe a significant seasonal cycle in deposition with a maximum during autumn and winter when combustion sources are maximal and a minimum in summer (de Fommervault et al., 2015). We focused our results on the impacts of N deposition on  $\text{NO}_3$  concentration because  $\text{NH}_4$  external inputs (from rivers and from the Atlantic) are not available.

Figure 7 shows the average seasonal  $\text{PO}_4$  deposition from dust. Mass deposition of dust-derived phosphate is low and highly variable in space and time. Model outputs reveal a strong decreasing gradient from South to North. Most of the Saharan deposition occurs during spring, with the exception of the far eastern part of the basin where the maximum occurs in winter with possible deposition from the Near and Middle East, in Cyprus and southern Turkey.

### 3.4. Basin scale impacts of aerosol deposition on marine nutrient budgets

In the "N" simulation, there is more than  $18 \cdot 10^9 \text{ g N month}^{-1}$  deposited on average ( $\pm 2.5 \cdot 10^9 \text{ g N month}^{-1}$ ) over the Mediterranean in the form of  $\text{NH}_4$  and  $\text{NO}_3$ . This deposition is about a third lower during summer (JJA). In the "N+ $\text{PO}_4$ " simulation, there is on average  $2.42 \pm 0.23 \cdot 10^9 \text{ g PO}_4 \text{ year}^{-1}$  deposited in addition to the nitrogen deposition,  $0.80 \cdot 10^9 \text{ g}$  of which is deposited during the summer period.

#### 3.4.1. Quantification of atmospheric contribution to the nutrient budgets

The newly computed deposition fluxes allow us to propose a new quantification of the atmospheric external supply of nitrate and phosphate to the different basins of the Mediterranean Sea (Table 4). In addition, we describe their spatial and temporal structure. A few previous studies assess the atmospheric contribution to external nutrient inputs in the Mediterranean. According to Krom et al. (2010), atmospheric inputs account for 61 to 70% of  $\text{NO}_3$  inputs to the eastern part when compared with riverine inputs, inputs from the Black Sea and inputs from the rest of the basin, as indicated in Table 4. From localized measurement, they also extrapolated  $\text{PO}_4$  deposition to account for 28% in the eastern basin. Our modeling approach allowed us to estimate the relative contribution of all external sources of  $\text{NO}_3$  and  $\text{PO}_4$  for the Mediterranean basin (namely river and coastal inputs, net Gibraltar exchange and atmospheric deposition). According to our simulations, atmospheric deposition accounts on average for respectively 10% of total  $\text{NO}_3$  and for 5 to 30% of  $\text{PO}_4$  external inputs to the entire water column at the basin scale over the simulation period (1997–2012). These estimates of phosphate inputs agree with the assessments made by Krom et al. (2010). However, our estimate of the nitrate atmospheric supply is well below the one from Krom et al. (2010). Estimations made by these authors are derived from measurements sparse in time at few localized stations and this extrapolation to the basin scale is very uncertain. We can also infer from Figure 4 that our model largely underestimates

the measured peak deposition fluxes. The maximal atmospheric contribution to nutrient inputs is observed in the Ionian basin. This area exhibits the highest variability in the atmospheric contribution (Table 4). The Ionian basin is the deepest and most pelagic area of the Mediterranean Sea, high oligotrophy and the weak influence of coastal inputs in this region lead to an important contribution from atmospheric nutrient inputs (29% for  $\text{NO}_3$  and 36% for  $\text{PO}_4$ ).

### 3.4.2. Spatial and seasonal variability of the impacts

Maximum of N deposition occurs in the northern parts of the basin, especially in the Adriatic and Aegean Seas (Figure 6). However, the main relative increase of annually averaged  $\text{NO}_3$  concentration occurs in the remote Ionian and Levantine basins and to a lesser extent in the eastern half of the western basin (Tyrrhenian Sea) (Figure 8a). Average surface  $\text{NO}_3$  enhancement in the Adriatic is about  $300 \text{ } 10^{-6} \text{ g m}^{-3}$ . This represents a relative enhancement of 13%. Whereas in the Ionian and Levantine basins, yearly surface nitrate enhancement consists of on average 23% and locally over 100%. The influence of P deposition is more visible in the southern part of the basin because most of the desert dust is deposited close to the coasts (Figure 7). Annual surface concentration of phosphate is on average  $22 \text{ } 10^{-6} \text{ g m}^{-3}$  greater in the "N+ $\text{PO}_4$ " simulation than in the "N" simulation. This represents locally 10 to 20% increase in surface phosphate concentrations (Figure 8b). We note a slight decrease (about 1%) in  $\text{NO}_3$  surface concentration following  $\text{PO}_4$  deposition from dust due to the extra consumption of nutrients induced by deposition (not shown).

Relative low contribution of atmosphere to  $\text{NO}_3$  budgets is observed in the areas where coastal inputs are high (e.g., in the Adriatic and Tyrrhenian basins). We also observe similar seasonal cycles of atmospheric nitrogen deposition and river nitrogen inputs in our forcing fields (not shown). Atmospheric deposition of dust-derived phosphate is much more variable in space and time and is 7 times lower than coastal inputs at the basin scale when considering inter-annual average. However, most of the mass dust sedimentation occurs close to the coasts, only wet deposition or exceptional deposition events occur in offshore areas.

The impacts of atmospheric deposition on nutrient budget vary throughout the year (Table 4 and see Louis et al. (2015) for experimental study). The highest atmospheric contribution to nutrient budget is observed during the stratified period (i.e., in summer). Seasonally averaged surface  $\text{NO}_3$  enhancement for the summer period is 43% in the Ionian basin, even though atmospheric deposition in this season is weak. Surface  $\text{PO}_4$  enhancement following deposition can be as high as 50% during the summer period whereas maximal surface enhancement observed throughout the rest of the year is 30%.

375 3.5. *The biological response to atmospheric deposition*

Figure 9 shows the average relative impacts of nitrogen and dust-derived  $\text{PO}_4$  deposition on surface chlorophyll *a* for different seasons. In general, we observe important relative effects of nitrogen deposition on surface chlorophyll concentration (more than 30% enhancement over the Eastern basin in summer). The relative effects of dust-derived phosphate deposition on surface chlorophyll are lower (10% at most). The effects of nitrogen deposition span over large and deep basins such as the Ionian, Levantine and Tyrrhenian whereas P-deposition effects are mainly observed in shallow areas such as the Adriatic, Aegean and coastal western basin. Deposition fluxes are minimal in summer, yet, in both cases, the impacts are maximal compared to other seasons. The characteristics of the biological response of the Mediterranean Sea are not strictly determined by the atmospheric deposition supply. This can be explained by the fact that impacts of atmospheric deposition are dependent on the underlying biogeochemical state of the Mediterranean which varies according to season and region. In our model study, we are able to separate the effects of nitrogen and phosphate deposition. We chose to separate the effects of nitrogen and dust-derived phosphate in this study, but we keep in mind that deposition events (most of the time through rain events) bring many chemically different aerosol components at once to the Mediterranean waters. The biological response being maximal in summer, the rest of our study will focus on this season.

Figure 10 shows the relative impacts of nitrogen and phosphate deposition on the simulated living organisms from the PISCES model: phytoplankton, zooplankton, on productivity variables: total primary productivity, grazing (preying) of zooplankton on phytoplankton, bacterial concentration and carbon export at 100m for the summer season. The effects of nitrogen deposition are overall much higher than those of dust-derived phosphorus deposition. Moreover, depositions of the two nutrients affect different regions. Nitrogen effects are maximal in the Tyrrhenian and Ionian waters whereas phosphate effects are located in the western basin, close to the African coast. Phosphate deposition impacts on marine productivity are weak, but still significant and mainly located in the areas of deposition. However we observe that zooplankton, primary production and grazing are also enhanced in the Adriatic and Aegean basins, even though there is very little deposition in these areas (Figure 7). This response is due to the intense nutrient limitations occurring during summer in the Mediterranean Sea, hence, even very small inputs of nutrient can unravel the nutrient limitation and fertilize quickly the biological chain.

In both cases the effects of atmospheric deposition observed are comparable in location and magnitude in all biological compartments. Effects of atmospheric deposition are higher on primary production and grazing than on phytoplankton. This shows that the increase of phytoplankton biomass due to intensification of primary production is directly consumed by zooplankton. Primary production through the use of nutrients in order to produce chlorophyll is the first step of the marine biological chain described by the PISCES model. Grazing is the consumption of phytoplankton organisms by primary producers (zooplankton). Phytoplankton concentration is affected by the difference between those two fluxes, as a result, some compensation effects are leading to lower phytoplankton concentration changes.

### 3.6. Maximal potential response of biological variables to atmospheric inputs

We report seasonal average responses to atmospheric inputs obtained with hindcast simulations of realistic conditions of the Mediterranean. However these results are constrained by the spatial and temporal characteristics of our N and P gridded deposition fields. Since these results are dependent of the characteristics of our forcing, we made an attempt to characterize the potential biological response of the Mediterranean Sea on a more general way using idealized experiments. The idealized simulation described in section 2.4 provides the modeled potential maximal response of biogeochemical variables to exceptional deposition events from different periods of the year.

We computed the maximal impacts of nitrogen and phosphate deposition on different variables (total phytoplankton concentration, total zooplankton concentration, primary production, grazing and chlorophyll *a* concentration) for each month of the year 1997. Figures 11 and 12 display results for February and June as a representation of respectively a non stratified state and a stratified, ultra-oligotrophic state of the Mediterranean. Maximum response of the biological tracers to a  $40 \cdot 10^{-3} \text{ g m}^{-2} \text{ day}^{-1}$  atmospheric N deposition event is high throughout the year, with a maximum spatial extent of the effects between May and December (not shown). The relative effects of N deposition reach their maximum intensity in June. Intense enhancement of primary productivity is found in the major part of the basin, an exception being almost no effect on productivity observed in the Algero-Provençal basin and the Adriatic Sea where there are sufficient external N inputs.

Simulated maximum response to P deposition seem to occur in June (Figure 12) and remains high until December (not shown), spanning all over the basin. Maximum productivity enhancement varies between 50 and 100% depending on the region. In particular, we observe a local doubling of primary productivity during the summer in the remote Ionian, Levantine and Tyrrhenian Seas and near the Gulf of Gabes.

The results from this sensitivity study show that the effects of atmospheric deposition observed in the "N" historical run are close to the potential maximal response of the Mediterranean with regards to N deposition (Figure 10). These results show however that dust-derived  $\text{PO}_4$  deposition can have effects on any part of the Mediterranean for a sufficient amount and if the deposition event occurs in summer or autumn. These findings seems to confirm the N-P-colimitation hypothesis for the Mediterranean (Tanaka et al., 2011).



## 4. Discussion

### 4.1. *The importance of atmospheric nutrients in the Mediterranean*

445 During winter and spring, when coastal inputs and vertical nutrients supply are maximal, atmospheric deposition has a very low influence on nutrient budgets. In the regions influenced by coastal and river inputs such as the Adriatic and Aegean Seas and in the western basin influenced by Atlantic inputs, atmospheric deposition has a low influence all year long on both nitrogen and phosphorus budgets (see Figure 8 and Table 4). During summer, the Mediterranean is highly stratified and the surface is virtually depleted in  
450 nutrients. The influence of coastal inputs is limited to shallow zones close to the river mouths. As a consequence, atmospheric deposition even in very small quantity becomes the main source of nutrient for biological production in pelagic areas.

### 4.2. *Specific effects of nitrogen and phosphorus deposition*

455 The enhancement of total primary production occurs mainly through new primary production. Similar results were found during the mesocosms studies in the DUNE project by Ridame et al. (2014). Nanophytoplankton and diatoms concentrations increase in the same proportions: 50 to 100% enhancement triggered by N deposition, 10 to 15% triggered by dust-derived P deposition but nanophytoplankton remains the most abundant compartment (~80% of total phytoplankton biomass). In the areas of high deposition effects on  
460 primary producers, we also observe enhancement of zooplankton concentrations due to grazing. Enhancement of zooplankton concentration is stronger than enhancement of phytoplankton, we observe an amplification effect towards zooplankton. Finally, the atmospheric deposition of nitrogen triggered a 20 to 30% enhancement of carbon export at 100 m in the eastern coastal areas.

Guieu et al. (2014b) and Guieu et al. (2014a) showed that fertilizing effects of aerosol deposition trigger fast  
465 and important increases of surface primary production, chlorophyll production, plankton concentration and also carbon export from the surface waters to depth. We found globally lower increases than these authors in biological variables following deposition, except for carbon export for which we find similar values. This 20 to 30% increase in carbon export triggered by atmospheric N deposition is consistent with the carbon export increase hypothesized by Krishnamurthy et al. (2010) and Christodoulaki et al. (2013). However,  
470 unlike Bressac et al. (2014) and Guieu et al. (2014c), we found very low effects of natural dust deposition on export and even a reduction of export during some months (not shown). The ballast effect hypothesized by these authors is not represented in PISCES, this may explain the low export we observe after intense dust deposition. Moreover Christodoulaki et al. (2013) found that primary production and phytoplankton biomass increase up to 30% in the eastern basin following atmospheric deposition of inorganic N and P. They also show  
475 that maximal impacts occur during summer and fall which is in good accordance with our results. Overall, our results suggest that the impacts of nitrogen deposition have much more consequences for the biology of the Mediterranean than natural dust deposition. In the global ocean, impacts of natural dust deposition

are more important because most of the productive regions are Fe-limited. Dust deposition can relieve this limitation (Desboeufs et al. (2005), Tagliabue et al. (2012)) in most oceanic waters, but the Mediterranean  
480 Sea is likely not Fe-limited (see Bonnet and Guieu (2006) for transient Fe-limitation hypothesis).

Even though bacterial activity is not explicitly modeled in the current version of PISCES, we are able to calculate a proxy of bacterial concentration (see Figure 10). Since this proxy is derived from zooplankton concentration, we observe enhancement of heterotrophic bacteria concentration in the areas of zooplankton concentration. Pulido-Villena et al. (2008) conducted laboratory experiments to observe the response of  
485 Mediterranean waters to Saharan dust addition. They found a 1.5 fold increase in heterotrophic bacteria abundance following high deposition of dust. In our long-term study, we do not find such an increase but we do detect a local increase of up to 12% in bacterial concentration following dust deposition in the Aegean Sea.

The effects of high deposition events are transitory. We noted a peak in most biological variables following  
490 the deposition. The peak was observed 4 to 10 days after N deposition event. Maximum response to P deposition is quicker (1 to 5 days). These results are coherent with Ridame et al. (2014) from in-situ experimental seedings.

#### *4.3. Implications of atmospheric deposition for nutrient limitations*

In general, the biological response to atmospheric deposition is higher in the most oligotrophic regions.  
495 The biological response reveals some structures that are different from the atmospheric nutrient deposition fluxes. The N/P ratio of aerosol deposition used in this study differs from the Redfield ratio for plankton growth imposed in PISCES (C:N:P = 122:16:1) with excess nitrogen deposited in the North of the basin and excess phosphate deposited in the South. In this version of the PISCES model, the biological compartments use phosphate and nitrate in the Redfield ratio proportions to grow. In such a redfieldian configuration,  
500 biological production is always limited by the availability of other nutrients that is controlled by the Redfield ratio. In a non redfieldian version, non-fixed in ratios could contribute to enhanced production when adding one element in excess. Nitrogen deposition is maximal in the Adriatic Sea due to the proximity of source regions located in the North of Italy (Im et al., 2013) but we observe negligible effects on nutrient budgets and biological production. Coastal inputs of nitrogen in this region are also important (Ludwig et al., 2009).  
505 The Adriatic basin is, at least in its northern region, not limited only by N (N-P colimitation may however exist). In general, impacts of nitrogen deposition are maximal in pelagic areas, far from coastal sources of nutrients (Figure 10). The relative impacts of N deposition on Chl *a* are maximal in the pelagic Tyrrhenian and Ionian basins, where deposition is minimal (see Figure 6). These two regions are among the least productive in the Mediterranean (Uitz et al., 2012; Lazzari et al., 2012). These results indicate that according to  
510 our model these areas are, at least part of the year, N-limited which is consistent with findings from Tanaka et al. (2011).

Deposited phosphate mass is very small in comparison to deposited nitrogen mass. Only a few episodic deposition events can bring nutrients to the surface and those events main influence is located close to the

coasts. The impacts we simulate on average seasonal biological production are weak and cover small areas  
515 corresponding in most cases to the areas of maximal deposition. We observe primary production enhance-  
ment of 8 to 10 % in the Adriatic basin and in the Gulf of Gabès (Figure 10). In these areas, atmospheric  
dust-derived P deposition is very weak (Figure 7). The importance of the biological reaction to such small  
deposition indicates that these areas are P-limited.

Finally, the differences in the response to N and dust-derived  $\text{PO}_4$  deposition from the atmosphere are the  
520 consequence of nutrient limitations. N or P fertilization is likely observed in N or P limited areas. We hy-  
pothesize that the N and P-fertilized zones observed in Figures 11 and 12 correspond respectively to N and  
P-limited areas. For instance, the Tyrrhenian and Ionian basins are responsive to both nitrogen and phos-  
phorus deposition, we can conclude that these regions are N and P co-limited, whereas primary production  
is highly increased in the Adriatic only by P deposition. This region is probably primarily P-limited. We  
525 do not know any study having done a basin-scale assessment of nutrient limitations in the Mediterranean.  
Numerous studies have investigated nutrient limitations in various stations of the Mediterranean, but these  
studies reflect the biological production state at specific places and times (Bonin et al., 1989; Herut et al.,  
2005; Tanaka et al., 2011). The use of model studies allows to investigate the state of biological production  
for periods of times extending over many years. We did our own assessment of nutrient limitation at the basin  
530 scale using our model outputs and we found that phytoplankton production is N-P-colimited in large parts  
of the basin, especially in the winter period (difference between the limiting terms for N and P <1 %, not  
shown). During the stratified season, productivity of nanophytoplankton is generally limited by one nutrient  
and N and P-fertilized areas in Figure 9 match the areas limited by N and P. Nevertheless, the use of a  
redfieldian model to assess nutrient limitation is not optimal, but it provides an interesting first approach.  
535 A perspective from this work would be to perform a similar analysis using a non redfieldian model in order  
to revisit our conclusions. In a non-Redfieldian configuration, the use of each nutrient for plankton growth  
is not determined by the other nutrients availability which may lead to different responses in production and  
different nutrient limitations.

## 5. Conclusions

540 This study presents a consistent estimation of atmospheric nutrient fluxes, issued from state-of-the-art  
modeling of sources and sinks and transport of phosphate and nitrate at the basin scale. Our estimation stands  
credible when compared to observations (see Figures 4 and 5) and represents a new and independent constraint  
and quantification of the atmospheric nutrient deposition over the Mediterranean basin. However, our values  
have still to be taken with caution ; aerosol and dust atmospheric transport and deposition simulation differ  
545 widely from model to model, regarding the total mass of nutrient deposition as well as its spatial and temporal  
structure (Basart et al., 2016). Moreover, we only found rare deposition fluxes measurements to constrain  
our model products. The development of intercalibrated networks for data acquisition such as the CARAGA  
and MOOSE frameworks developed in the frame of ChArMEx are an important step to confront model

outputs to data, but such initiatives are still rare in the Mediterranean. Recent analysis of P deposition  
550 samples in northwestern and Eastern Mediterranean sites highlighted the importance of organic forms in  
soluble phosphorus deposition (Violaki et al., this issue), such inputs could be taken into account in further  
modeling studies. We provide a first estimation but point out the need to compare different model products  
in the future. Comparing deposition fields from different models and newly available measurements would  
be necessary to better assess atmospheric deposition but is beyond the scope of this study, which is targeted  
555 to the biogeochemical response to atmospheric nutrient deposition.

We show that the atmosphere is a local scale, transitory source of bioavailable nutrients. Yet it can be the  
dominant source of nutrients during spring and summer, especially for pelagic areas far from coastal inputs.  
The use of a high resolution atmospheric deposition forcing allowed us to quantify the importance of the  
atmosphere as a source of major nutrients. We show that atmospheric deposition has greater importance on  
560 nutrient budgets in the Ionian and Levantine basins. The magnitude of these impacts are determined by the  
seasonal cycle of deposition and the underlying biogeochemical state of the waters.

Our simulations show that phosphate deposition from desert dust (mainly from Sahara) has a low but  
significant effect on biological productivity. The most significant effects occur during summer, when the  
Mediterranean is at its most oligotrophic stage. They are located in the areas of high deposition (which  
565 are located in the western basin) with a decreasing gradient towards the North, and in the Adriatic where  
deposition is relatively low but the area is P-limited. We also show that N deposition that mainly comes from  
combustion sources has large impacts on biological production in wide areas of the basin. Fertilizing effects  
of nitrogen deposition are observed in the Tyrrhenian, Ionian and Levantine basins throughout the year but  
maximal response is reached during summer. Maximal potential influence of dust-derived P deposition is not  
570 reached in our "N+PO<sub>4</sub>" simulation (effects observed in Figure 10 are lower than those of Figure 12), showing  
that the effects of dust deposition on marine productivity may be greater in case of extreme deposition event.  
The effects of nitrogen and dust-derived phosphate deposition observed in this work reflect the nutrient  
limitations in the Mediterranean. We hypothesize that the effects of nitrogen and phosphate are maximal in  
areas and during periods of nitrogen (respectively phosphate) limitations. However, the use of a redfieldian  
575 model in an oligotrophic area such as the Mediterranean is a possible limitation of our study on atmospheric  
deposition impacts because the use of nitrogen and phosphorus by plankton are linked by the Redfield ratio.  
Hence, the addition of one element in excess in comparison to the other might lead to an incomplete response  
of the biology in this model. Further work will aim at disentangling the effects of aerosols on nutrient  
limitations with a non redfieldian version of the PISCES model that is currently under development.

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Name	Rivers + Gibraltar	N deposition	P deposition
No dep		Monthly	No
N		Monthly	Monthly
N+PO <sub>4</sub>		Monthly	3-hourly
PO <sub>4</sub> only (not shown)		Monthly	No

Table 1: Description of the biogeochemical forcings used for our different simulations.

	[PO <sub>4</sub> ] <sub>inflow</sub>	[PO <sub>4</sub> ] <sub>outflow</sub>	[NO <sub>3</sub> ] <sub>inflow</sub>	[NO <sub>3</sub> ] <sub>outflow</sub>
This work	0.19 (0.06)	0.39 (0.003)	3.5 (1.0)	6.3 (0.06)
Huertas <i>et al.</i>	0.27 (0.04)	0.49 (0.05)	3.1 (0.4)	8.7 (0.6)

Table 2: Average concentrations ( $10^{-6}$  mol L<sup>-1</sup>) in Atlantic inflowing and Mediterranean outflowing waters. Our calculations are averages for the whole simulation period, Huertas *et al.* (2012) are for 2005 – 2008. Parentheses values are the standard deviation.

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

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

Basin	contribution to NO <sub>3</sub>	contribution to PO <sub>4</sub>
Krom <i>et al.</i> east	61–70	28
Whole Med.	10	9.1 (5-30)
West	13 (10-18)	10 (5-25)
Adriatic	5.0	3.0 (2-10)
Aegean	3.0	3.5 (0-20)
Ionian	29 (25-45)	36 (20-80)
Levantine	9.0 (8-11)	11 (1-30)
East	6.0	10 (2.0-23)

Table 4: Atmospheric contribution (%) to NO<sub>3</sub> and PO<sub>4</sub> inputs in different sub-basins (atmospheric inputs/(atmospheric inputs + coastal inputs + Gibraltar inputs)). Calculations are made for the entire simulation period (1997 - 2012). The first row is literature estimates and the seven next are from our model simulations. Two values in parentheses show the minimum and maximum contribution values over the year, one interannual average value is given when temporal variability of contribution is low (<2%).

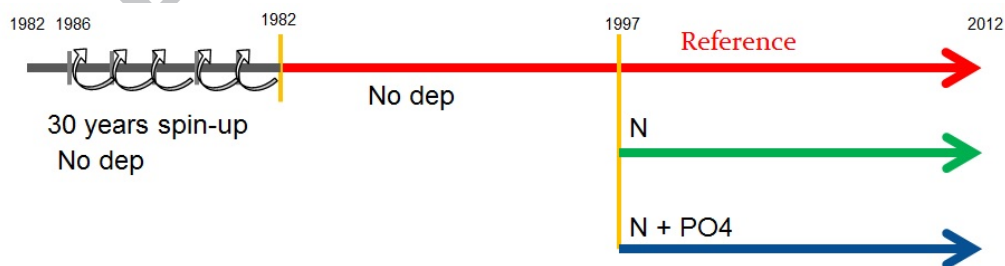


Figure 1: Schematics of our modelling strategy.



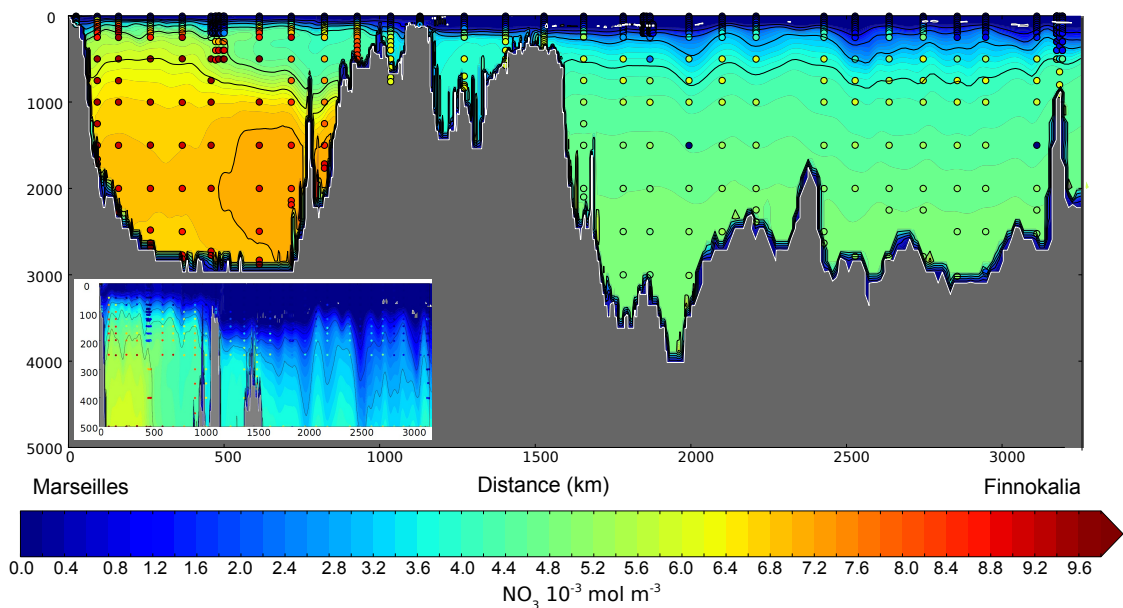
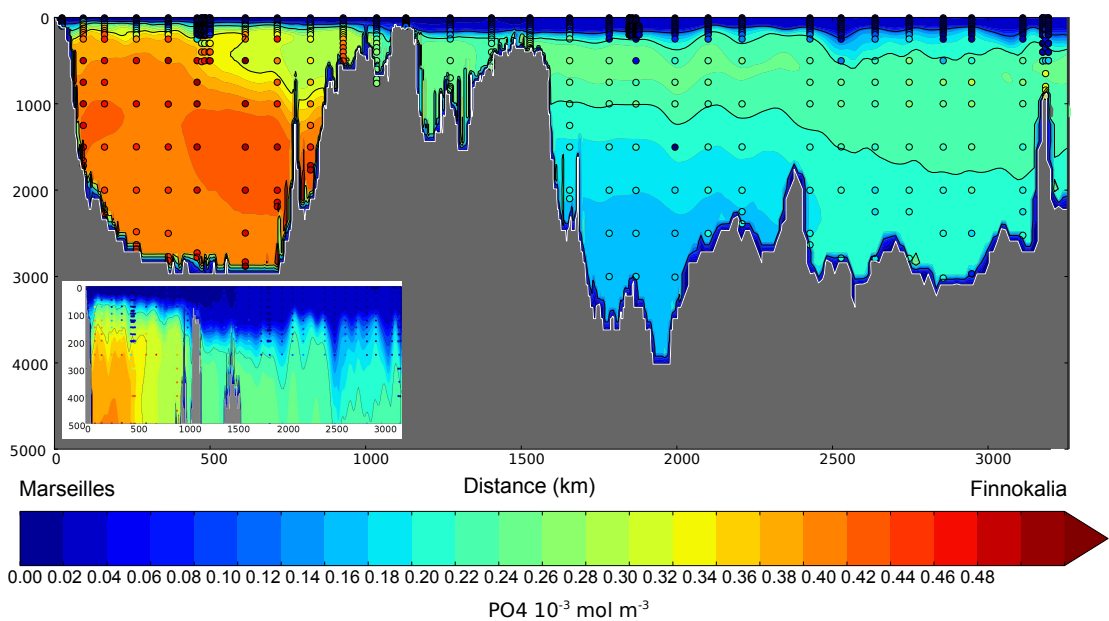
(a)  $\text{NO}_3$ (b)  $\text{PO}_4$ 

Figure 2: Evaluation of the model NEMOMED12/PISCES. Concentration calculated by the model (background) and measures from the BOUM cruise (dots) Moutin et al. (2012). Values represent the month of June 2008 which is the period of the cruise. Inserts show the top 500m of the water column.

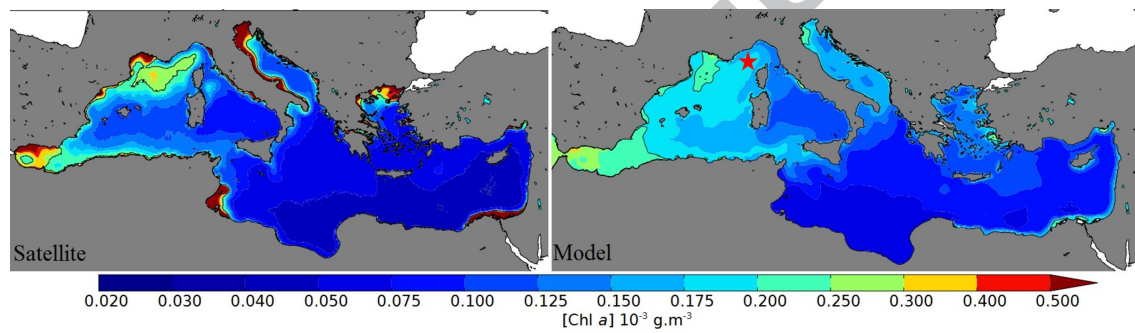
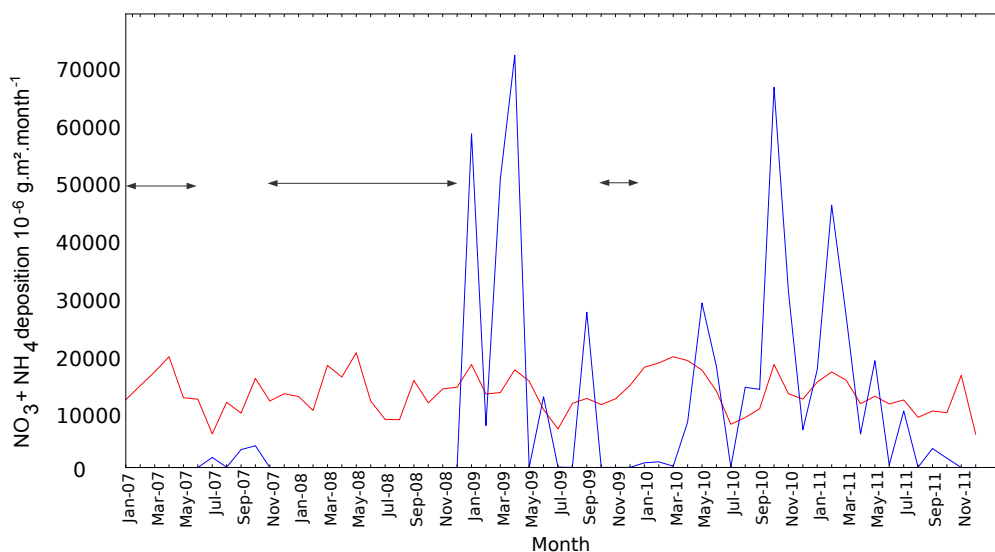
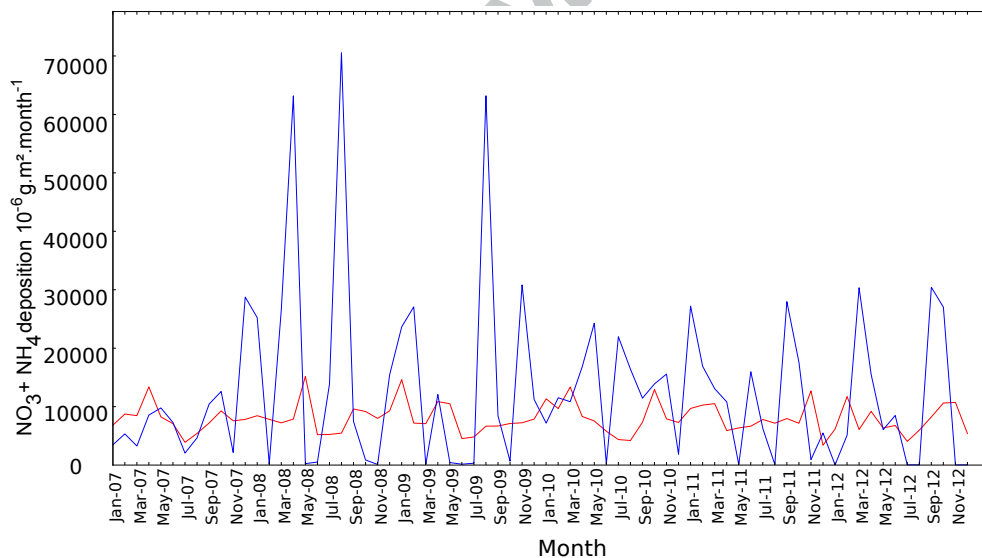


Figure 3: Evaluation of the model NEMOMED12/PISCES. Satellite (left) and modeled (right) surface chlorophyll *a* averaged over the period 1997–2012. The satellite observation comes from ESA–CCI reflectance products interpolated with the MedOC4 algorithm Volpe et al. (2007). Red star indicates the location of the DYFAMED measurement station.

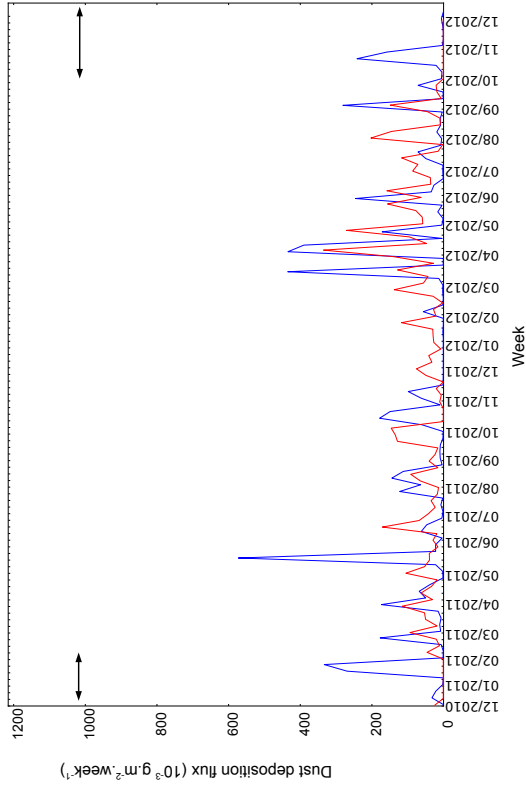


(a) Evaluation of N deposition at the Cap Ferrat station. Standard deviations of the modeled and measured time series are respectively 2 530 and 14 550  $10^{-6} \text{g m}^{-2} \text{month}^{-1}$

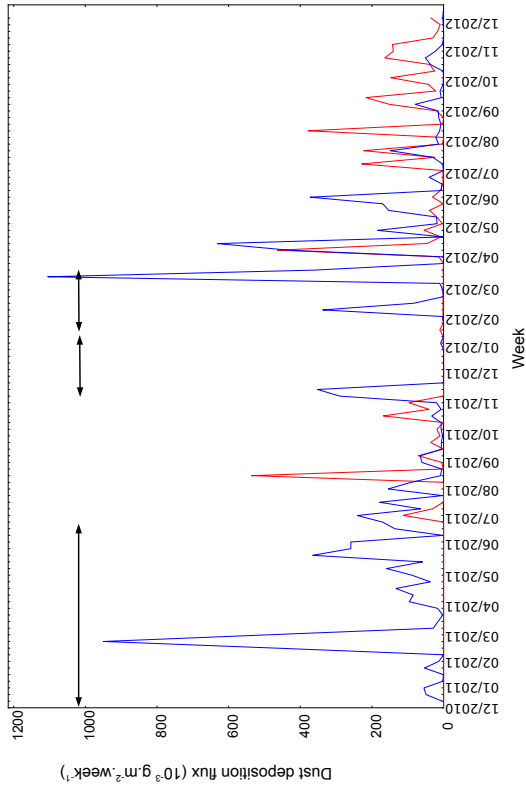


(b) Evaluation of N deposition at the Frioul station. Standard deviations of the modeled and measured time series are respectively 3 460 and 16 970  $10^{-6} \text{g m}^{-2} \text{month}^{-1}$

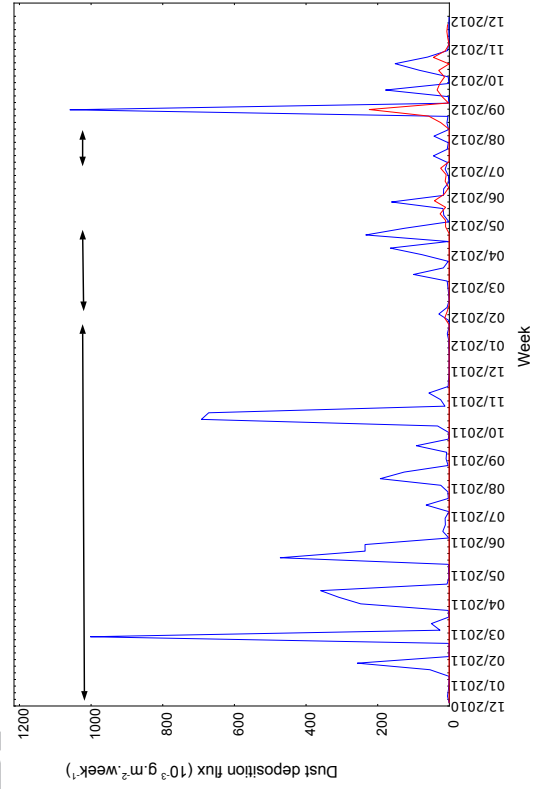
Figure 4: Evaluation of atmospheric nitrogen deposition. Measured (blue) and modeled with LMDz-INCA (Wang et al., 2015) (red) at the MOOSE stations. Arrows indicate periods without measurements. See Section 2.2.1 for details.



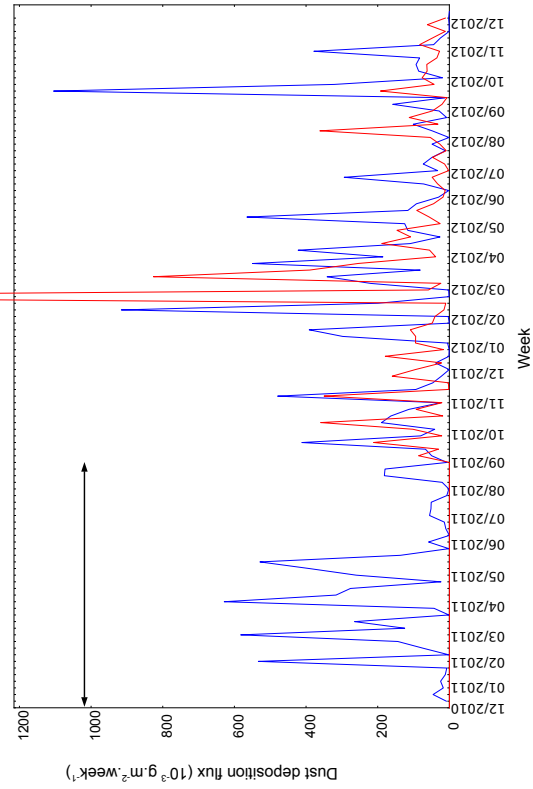
(a) Evaluation of dust deposition at Majorca station. Standard deviations of the modeled and measured time series are respectively  $181$  and  $107 \cdot 10^{-6} \text{ g m}^{-2} \text{ week}^{-1}$



(b) Evaluation of dust deposition at Frioul station. Standard deviations of the modeled and measured time series are respectively  $105$  and  $60.4 \cdot 10^{-6} \text{ g m}^{-2} \text{ week}^{-1}$



(c) Evaluation of dust deposition at Lampedusa station. Standard deviations of the modeled and measured time series are respectively  $211$  and  $338 \cdot 10^{-6} \text{ g m}^{-2} \text{ week}^{-1}$



(d) Evaluation of dust deposition at Corsica station. Standard deviations of the modeled and measured time series are respectively  $178$  and  $25.0 \cdot 10^{-6} \text{ g m}^{-2} \text{ week}^{-1}$

Figure 5: Evaluation of atmospheric deposition. Measured (red) and modeled with ALADIN-Climat (Nabat et al., 2015) (blue) total dust deposition fluxes for the period 2011–2012 at the CARAGA stations. The horizontal arrows at the top of the plots indicate missing measurement periods. See Section 2.2.2 for details

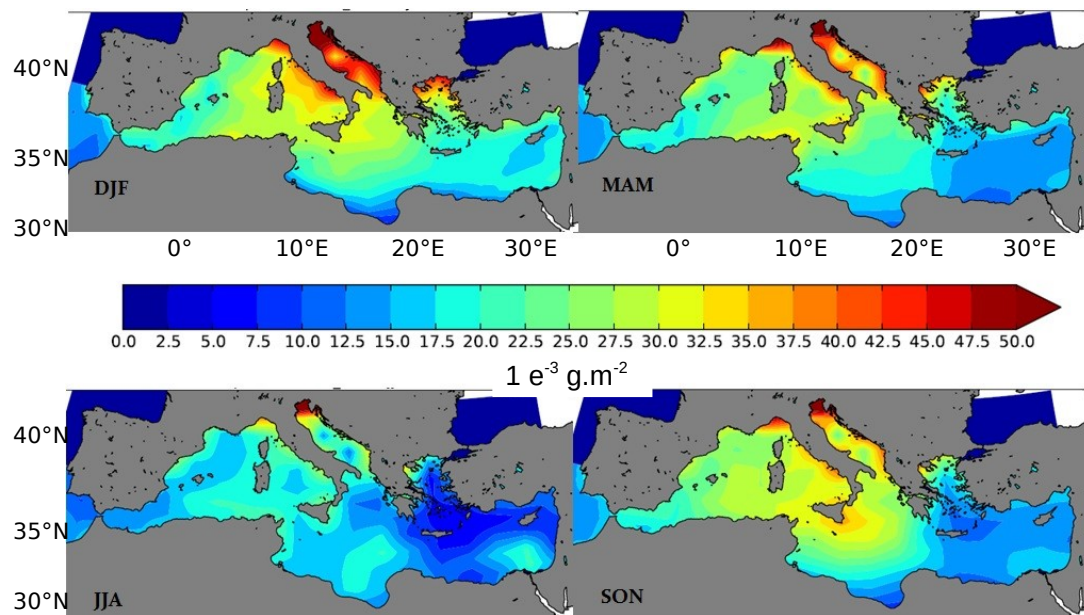


Figure 6: Average seasonal (1997–2012) nitrogen (i.e. NO<sub>3</sub> + NH<sub>4</sub>) mass flux (10<sup>-3</sup> gN m<sup>-2</sup>) from the LMDz-INCA model.

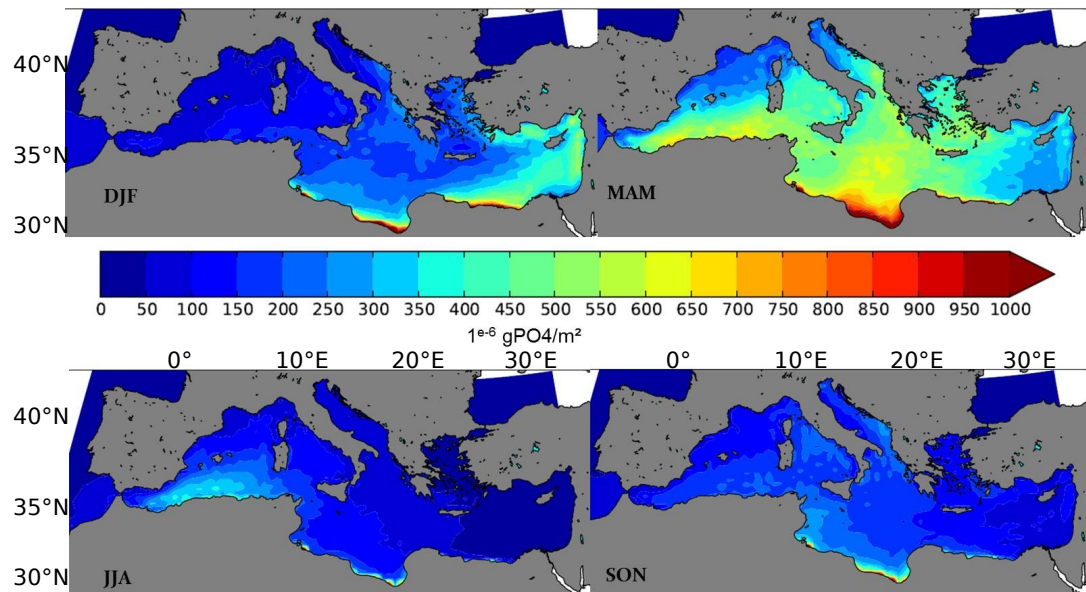


Figure 7: Average (1997–2012) seasonal PO<sub>4</sub> mass flux (10<sup>-6</sup> gPO<sub>4</sub> m<sup>-2</sup>) from the ALADIN-Climate model.

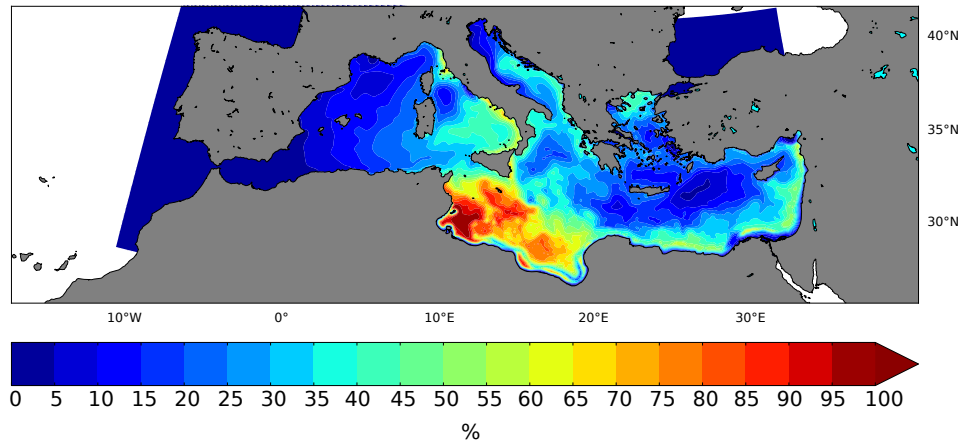
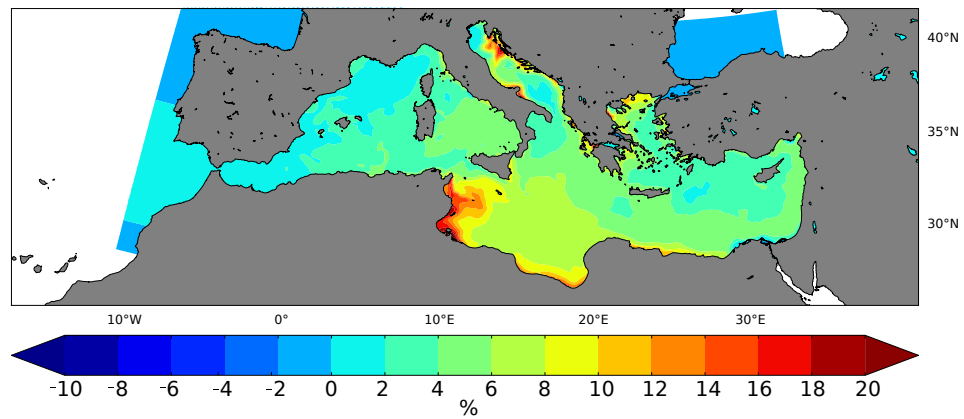
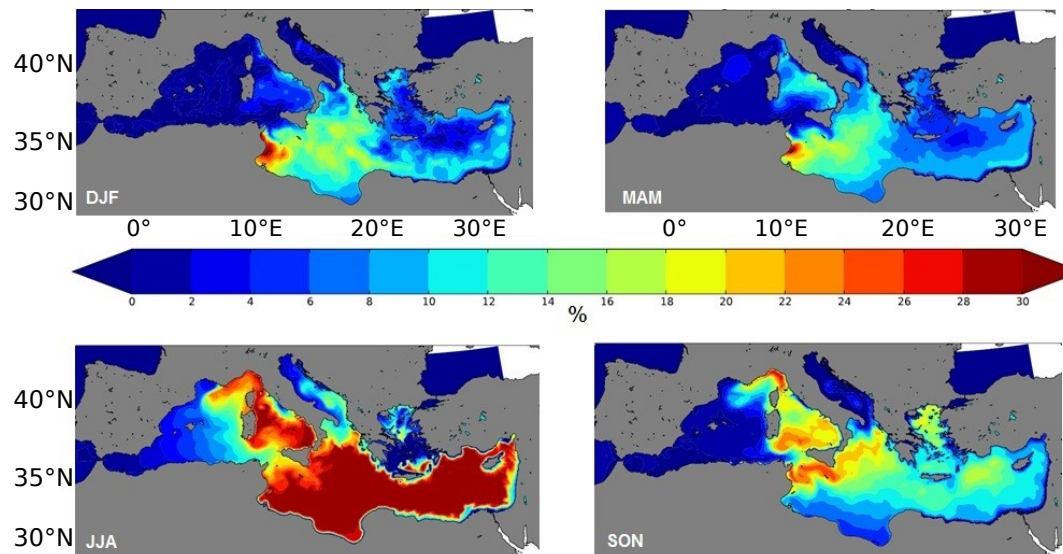
(a) Interannual relative surface change in  $\text{NO}_3$ (b) Interannual relative surface change in  $\text{PO}_4$ 

Figure 8: Interannual relative surface (0–10m) changes in  $\text{NO}_3$  (a) and  $\text{PO}_4$  (b). The differences are computed by difference between the interannual average concentrations in the N-Reference simulations for 8a and N+ $\text{PO}_4$ -N simulations for 8b.



(a) Influence of N deposition on surface Chl *a* concentration



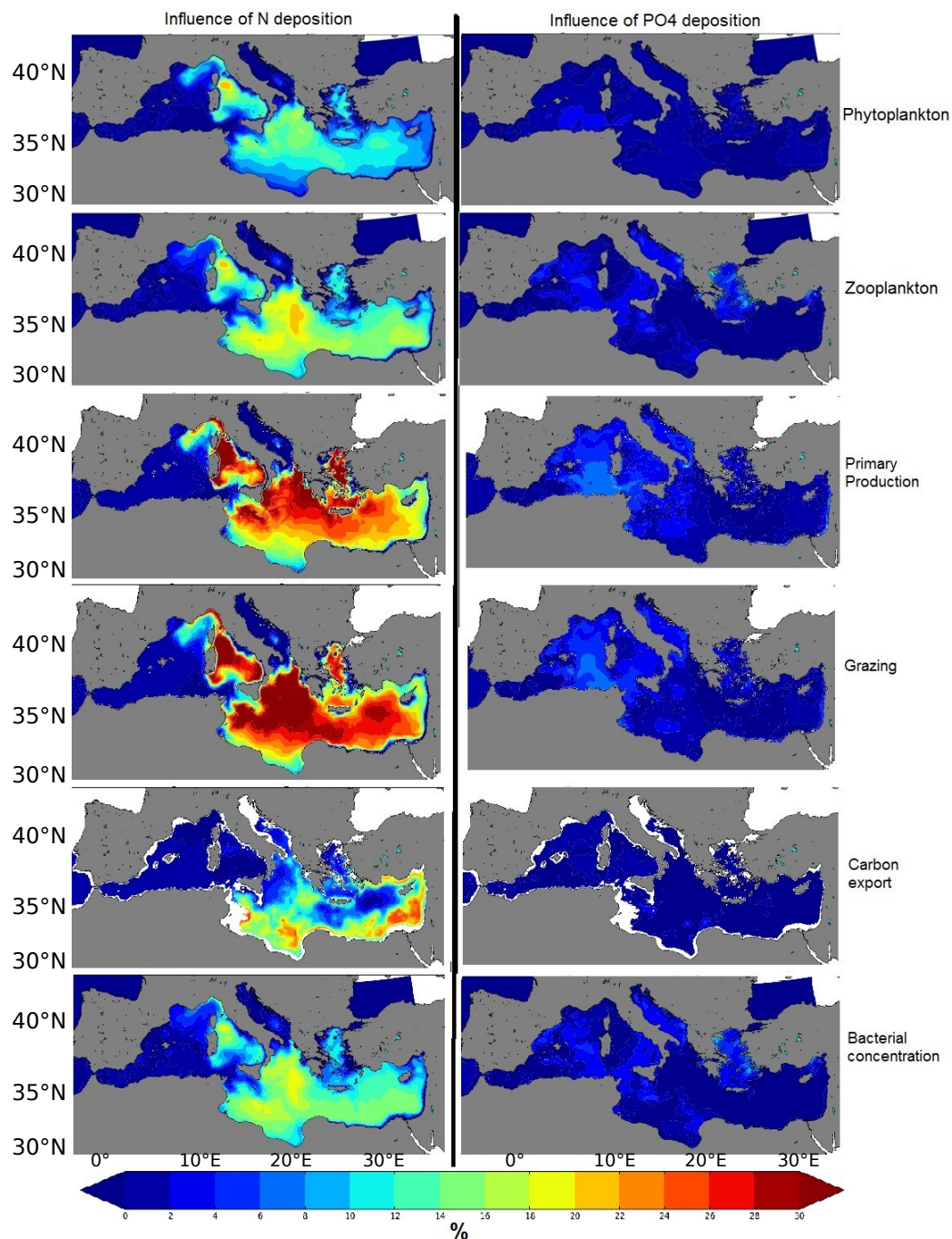


Figure 10: Relative influence (%) of atmospheric deposition on biological tracers for the summer season (JJA). Bacterial concentration is calculated as  $[Bact]=0.7([Microzoo]+2[Mesozoo])$ . This relation has been constructed from a version of PISCES including explicit bacterial biomass calculation (Aumont et al., 2015). Carbon export is the export of carbon particles at 100m, white zones are shallower than 100m.

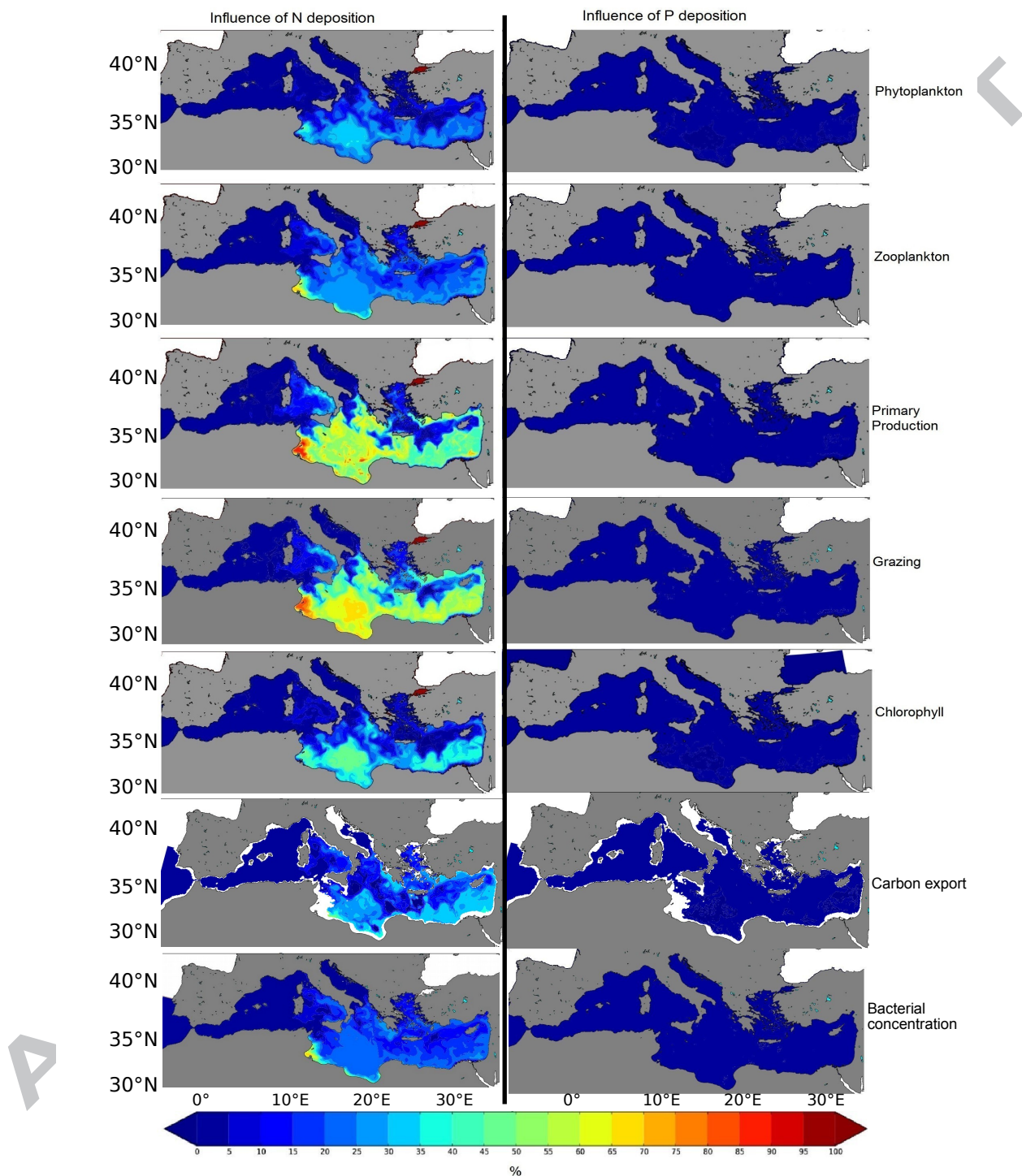


Figure 11: Maximal relative impacts (%) of atmospheric deposition on biological variables observed for the month of February.

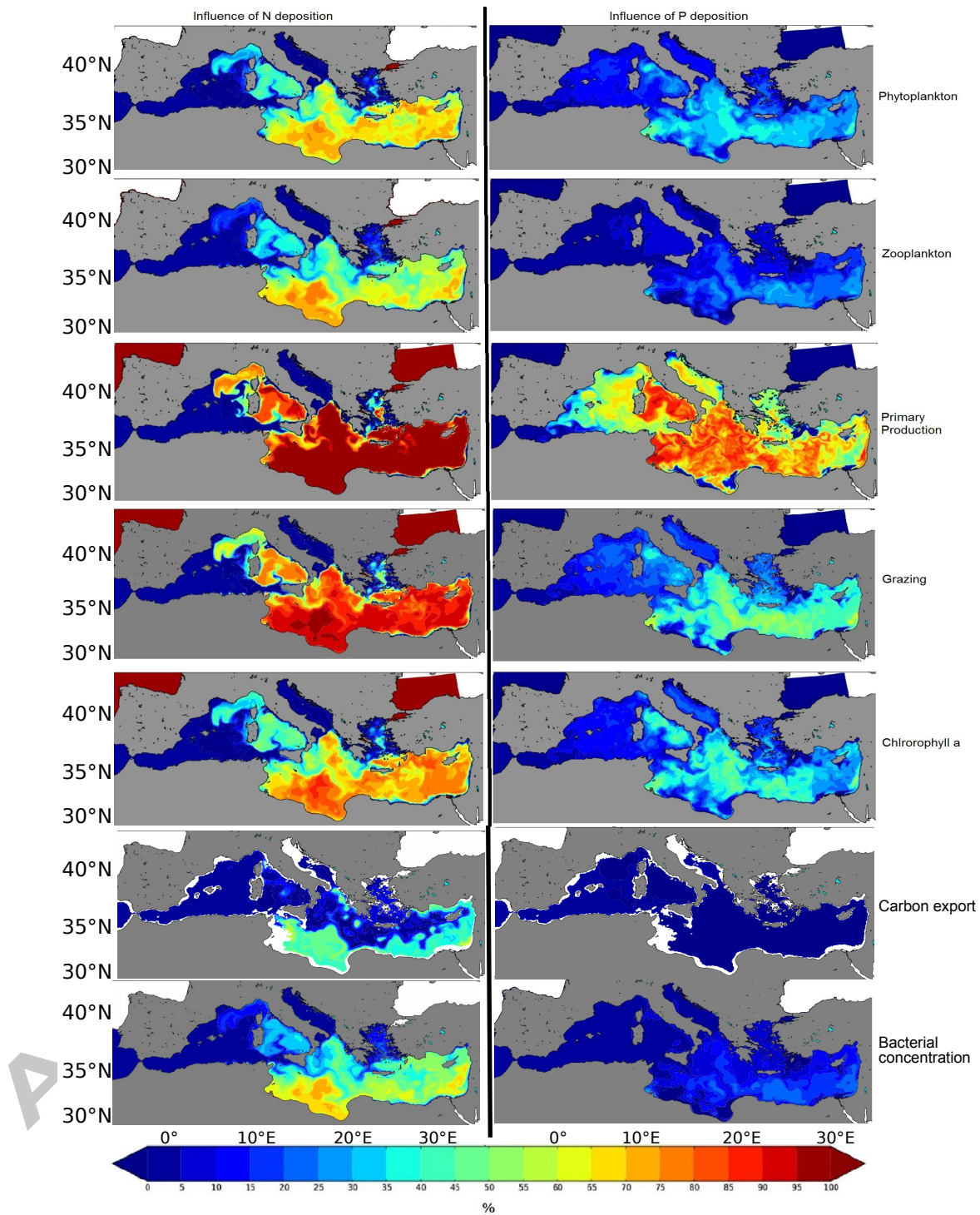


Figure 12: Maximal relative impacts (%) of atmospheric deposition on biological variables observed for the month of June.