#### Dear Editor,

Please find below the authors' responses to the reviewers' comments.

We have improved the manuscript by incorporating changes suggested by the reviewers. In particular, we have reorganized the results section and tried to express more clearly our findings in section 3.3.5. We also have reorganized the discussion by emphasizing all limitations of the study into section 4.1, and summarizing and discussing the impacts of our results in section 4.2.

As suggested by reviewer 2, we incorporated the results concerning atmospheric deposition effects in the methods and results section.

The authors would like to emphasize that the English has been corrected throughout the manuscript in an effort to make it easier to read.

You can find a track version of the manuscript highlighting all changes attached to these answers. Best regards,

The authors

Authors sufficiently addressed all of my comments, and the quality of the manuscript is significantly improved. The contribution of this modeling study in the effort to understand the impacts of climate change on the Mediterranean Sea biogeochemistry is clearer than before. However, some points in the present version of the manuscript have to be restructured. This mainly concerned the uncertainties and limitations associated to this study, and one part of the results (see my major comments). In addition, several minor corrections are necessary (see my minor comments). Therefore, I believe that this paper will likely be a significant contribution and reach the quality standards for publication in the Biogeosciences journal after minor revisions of the manuscript.

We thank the reviewer for their comments, which we have answered fully, while reorganizing and clarifying the manuscript accordingly.

#### Major comments:

1) Sections "4.2 Climate Change Scenario" and "4.3 Uncertainties from the PISCES model": these subsections need to be regrouped, completed, and rearranged at the end (or at the beginning) of the discussion section. In order to have a better insight into all uncertainties and limitations of the study (most of them already mentioned by the authors), to consider their influences on the results and processes discussed in the other sub-sections of the discussion (i.e., 4.1 and 4.4), and maybe to discuss some perspectives. For this, this sub-section should have a more general title, for example, "Limitations of the Study", and divided into three parts (at least):

a. Climate change scenario: mostly section 4.2

b. PISCES model: mostly section 4.3

*c. External sources of nutrients used (riverine and Atlantic inputs) and missing (atmospheric deposition): scattered in the manuscript* 

For example, I was expecting to have information in the discussion section about the influence of the following uncertainties and/or limitations on the biogeochemical response of the Mediterranean Sea, - How the Atlantic condition and Atlantic nutrient concentrations used in the model could influence the biogeochemical results obtained, mainly for the Western Basin? For example, the authors wrote, Line 646 - "...the choice of atmospheric and Atlantic conditions has a strong influence on the MTHC." with no information or discussion about possible consequences on the biogeochemical results obtained in their study.

# - How the atmospheric deposition (not represented in the model, line 157 - "We did not include atmospheric deposition...") could influence the fact that the Mediterranean Sea is becoming more P-limited at the end of the century in your study?

We have rearranged and modified the Discussion to address these comments. We regrouped sections 4.2 and 4.3 in one section named "Sources of uncertainties" (lines 585 to 685). This section is divided into 3 subsections covering:

#### - 1: Uncertainties linked with the climate change scenario (lines 593 to 623)

In this subsection, we discuss the potential impacts of different climate change scenarios on the Mediterranean Sea. Our HIS/A2 simulation increased stratification in Mediterranean leads to lower surface nutrient concentrations and hence lower primary productivity (Figures 7a,d and 8a,d). Conversely, Macias et al (2015) found that the RCP4.5 and RCP8.5 scenarios reduced the simulated vertical stratification , which led to slightly increased surface primary productivity. This large sensitivity to the choice of climate change scenario, calls for the use of a greater number of scenarios when modeling related changes in the Mediterranean Sea.

See lines 599 to 611: "Overall, the increase in stratification in our A2 climate change scenario leads to different evolutions of nutrient concentrations between the surface and the intermediate and deep waters with surface waters becoming more sensitive to external nutrient sources (Figures 7 and 8). On the other hand, Macias et al. (2015) found that primary productivity slightly increased as a result of decreased stratification in the climate change scenarios RCP 8.5 and RCP 4.5. The A2 scenario that we used was the only one available with 3–D daily forcings, as necessary for coupling with the PISCES biogeochemical model. However, Adloff et al. (2015) showed that other SRES scenarios such as the A1B or B1 may lead to a future decline in the vertical stratification with probably different consequences on the Mediterranean Sea biogeochemistry. It should be complemented by new simulations that explore the various sources of uncertainty (model choice, internal variability, scenario choice) once appropriate forcings become available for multiple models as expected from the Med–CORDEX initiative (Ruti et al., 2016)."

#### - 2: Uncertainties from the PISCES model (lines 625 to 646)

#### - 3: Uncertainties linked with external nutrient sources (lines 648 to 685).

In this subsection, we discuss the importance of accurately representing the external nutrient sources in future Mediterranean biogeochemical simulations. In an effort to facilitate reading, we divided this section in 3 paragraphs:

#### Fluxes through the Strait of Glbraltar

The Atlantic conditions and nutrient concentrations were shown with the CTRL\_RG simulation to have a strong influence on the western Mediterranean biogeochemistry. Hence it is important to have realistic scenarios for the evolution of the Atlantic boundary conditions to simulate the Mediterranean. In our

case, the Atlantic conditions are simulated with the same model (NEMO/PISCES) and the same climate change scenario (A2). Therefore, they are entirely compatible with our scenario.

Lines 675 to 680: "Results from our CTRL\_RG simulation show that the increase in nitrate and phosphate incoming fluxes through the Strait of Gibraltar leads to higher surface concentrations in the western Mediterranean. The Atlantic nutrient concentrations are derived from a global version of the same model used our simulations (NEMO/PISCES) and forced under the same A2 climate change scenario). Therefore, there is no incompatibility issue between for the forcing and model."

#### **Riverine nutrient fluxes**

The river fluxes of freshwater and nutrients are derived from different models and there are uncertainties with the nutrient flows we represent. However, this is the only scenario for river inputs available.

We modified the text: "For the riverine nutrient inputs, scenarios from the MEA report are based on different assumptions from the IPCC SRES scenarios used to compute freshwater runoff in the HIS/A2 simulation. Freshwater discharge from Ludwig et al. (2010) is based on the SESAME model reconstruction and differs

from freshwater runoff in the ARPEGE–Climate model used to force our physical model. This may lead to incoherences between water and nutrient discharges, but the nutrient discharges from Ludwig et al. (2010) are the only ones that are available. Furthermore, the SESAME model is not coupled with NEMO/PISCES. Associated discrepancies and the uncertainties linked with the use of inconsistent scenarios in our simulation should be addressed by developing a more integrated modelling framework to study the impacts of climate change on the Mediterranean Sea biogeochemistry." Lines 658 to 674.

#### Potential effects of aerosol deposition

There is no scenario for the evolution of aerosol deposition during the 21<sup>st</sup> century. It is difficult to forecast the evolution of aerosol emissions and deposition as it is influenced by socio-economic decisions, land-use change, winds and rains. Measurements of atmospheric deposition remain sparse and it is difficult to interpret any tendency from them. In this context of very sparse data and modeling, it is difficult to derive robust scenarios for the evolution of aerosol deposition over the Mediterranean.

However, as we noticed important effects of external nutrient sources on the Mediterranean surface biogeochemistry, atmospheric deposition may also influence on the response of the Mediterranean biogeochemistry to future changes. Our results using present day phosphate deposition from natural dust (see figure 17) show that atmospheric deposition of phosphate enhances surface primary production in the surface Mediterranean. Thus enhanced phosphate fluxes from aerosols may limit the surface decrease of phosphate concentrations and limit phosphorus limitation. However, in the HIS/A2\_NALADIN simulation, the surface Mediterranean is still P-limited over most of the Mediterranean because the atmospheric nutrient fluxes are low in comparison to the fluxes from rivers and across the Strait of Gibraltar. Therefore, it appears unlikely that changes in aerosol deposition from natural dust supply will have a larger impact on the Mediterranean biogeochemistry in the future. However, there are multiple sources of aerosols that are not included in atmospheric models such as anthropogenic, volcanic and volatile organic compounds. They may constitute important nutrient fluxes for surface waters of the

Mediterranean Sea. Moreover, aerosols affect radiative forcing over the Mediterranean and hence also climate conditions. It seems therefore important to try to represent this nutrient source in the Mediterranean models.

We added in the text (lines 711 to 723): "Results from these simulations show that enhanced phosphate fluxes from aerosols may limit the surface decrease of phosphate concentrations and limit phosphorus limitation. However, in the HIS/A2\_NALADIN simulation, the surface Mediterranean is still P-limited in most of the Mediterranean because the atmospheric nutrient fluxes are low in comparison to riverine nutrient fluxes from rivers and the nutrient flux through the Strait of Gibraltar (see Richon et al., 2017). Therefore, it appears unlikely that changes in aerosol deposition from natural dust would greatly influence future Mediterranean biogeochemistry. However, there are multiple sources of aerosols that are not included in atmospheric models, e.g., anthropogenic, volcanic and volatile organic compounds (e.g. Wang et al., 2014; Kanakidou et al., 2016). Their combined influence could perhaps constitute an important nutrient flux to the Mediterranean, thus altering the evolution of its biogeochemistry. Moreover, aerosols affect radiative forcing over the Mediterranean and may impact the climate conditions (Nabat et al., 2015b). Thus, efforts should be made to accurately represent this nutrient source in Mediterranean models to assess the effect on Mediterranean Sea biogeochemistry with regards to climate change."

2) In the conclusion – between lines 755 and 763 – authors highlighted the differences between the western and eastern basins in their biogeochemical responses to the climate change. This part of the results, which, I believe, is one of the major contribution of the manuscript, was absent in the discussion. There were even some contradictory statements, between the results, discussion and conclusion sections (see below), which make difficult to determine the author's opinion on some key aspects:

Line 449 – "But the strong difference between CTRL\_R and HIS/A2 at the end of the century indicates that vertical stratification leads to a decrease in surface layer nitrate concentrations, probably linked both with lower winter mixing and nutrient consumption by phytoplankton."

*Line 739 – "Stratification may lead to increased productivity in the surface because of the nutrient concentration increase (see also Macias et al., 2015)..."* 

*Line* 751 – *"increased stratification, and changes in Atlantic and river inputs, can lead to a significant accumulation of nitrate and a decrease in biological productivity in the surface..."* 

*Line 760 – "the eastern basin is more sensitive to vertical mixing and river inputs than the western basin [...] and the stratification observed in the future leads to a reduction in surface productivity..."* 

Please, clarify your interpretation of the results in the discussion, in order to support all your statements in the conclusion.

Our HIS/A2 simulation is the first to consider the joint evolution of physical aspects and external nutrient sources. In some regions, there are antagonistic effects of stratification and nutrient discharge. For instance, in the eastern basin, the increase in nitrate discharge from rivers tends to increase the surface nitrate concentration (see the evolution of nitrate concentrations in CTRL\_R). However, changes in physical conditions seem to lower surface nitrate concentrations in the eastern basin by the end of the 21<sup>st</sup> century (see concentrations in HIS/A2).

See lines 431-440: "In the eastern basin, the impacts of river discharges of nitrate seem to have large influence on the nitrate accumulation as shown by the similar evolution of HIS/A2 and CTRL\_R simulations (Figures 8d, 8e and 8f). Figure 8d shows the contrasted effects of climate and biogeochemical changes. The strong difference between CTRL\_R and CTRL concentrations at the beginning of the

simulation (almost 0.4 mmol m<sup>-3</sup>) indicates that riverine nutrient discharge has a strong influence on surface nitrate concentrations in the eastern basin and is responsible for an important part of the eastern Mediterranean nitrate budget (see also Table 3). But the strong difference between CTRL\_R and HIS/A2 at the end of the century indicates that vertical stratification leads to a decrease in surface layer nitrate concentrations, probably linked both with lower winter mixing and nutrient consumption by phytoplankton."

Our results suggest that the western Mediterranean basin is more sensitive to changes in nutrient fluxes through the Strait of Gibraltar and to the physical changes linked with climate change. The eastern basin is generally less sensitive to the fluxes through the Strait of Gibraltar, but some regions such as the Adriatic Sea and the Levantine basin are more sensitive to evolution of nutrient fluxes from rivers and to the physical changes.

See lines 742 to 748: "Results from our different control simulations indicates the extent to which the choice of the biogeochemical forcing scenario may influence the future evolution of the Mediterranean Sea biogeochemistry. Considering only our climate change scenario, less vertical mixing causes a basin-wide decline in surface nutrient concentrations, causing a reduction in primary productivity. But the combined effects of nutrient fluxes from external sources and climate change that lead to the surface accumulation in nitrate (figures 8a and 8d and Table 3). In particular, nutrient inputs through the Strait of Gibraltar have substantial influence on the western basin."

Our results show 2 main conclusions regarding the effects of climate change versus biogeochemical forcings: 1) surface biogeochemistry is influenced by the evolution of both external nutrient fluxes and climate change, and climate change effects on biogeochemistry are more visible in the intermediate and deep waters; 2) nutrient fluxes through the Strait of Gibraltar primarily influence the western basin whereas riverine nutrient input primarily influences the eastern basin.

We tried to emphasize these observations in the result section by adding a paragraph (lines 447 to 451): "Evaluating separately the evolution of nutrient concentrations in different layer of the Mediterranean Sea shows that external nutrient fluxes primarily affect the surface in the western basin whereas climate change affects the entire water column. Also, climate and nutrient fluxes may have opposite effects on surface nutrient concentration. This leads to different trends in nutrient concentrations in the surface layer and in the intermediate and deep layers. In particular, surface nitrate in the eastern basin is observed to increase as a result of increased river discharge, but climate change effects lower concentrations in HIS/A2 (see figure 8d). On the other hand, climate and river discharge of nitrate have similar effects on the intermediate and deep eastern layers, leading to the simulated increase in nutrient content (Tables 2 and 3)."

See also lines 738-741:" The difference between HIS/A2 and CTRL\_RG phosphate concentrations in the intermediate and deep layers (Figure 7b, 7e, 7c, 7f) indicates that variations of phosphate concentrations during the 21st century are primarily driven by climate change while nitrate concentration is equally sensitive to changes in biogeochemical forcings."

Also, in an effort to make to results section 3.3.5 easier to read, we clearly separated the results concerning phosphate and nitrate in 2 different paragraphs. In each of these paragraphs, we first

describe the results in the western basin (with the order: surface, intermediate and deep waters) and then in the eastern basin.

#### Minor comments:

1) Line 68 – "Macias et al. (2015) simulated [...]. They found that [...] primary productivity over the eastern Mediterranean basin may increase as a result of density changes (increased stratification isolating the upper layer from the rest of the water column)."

Need to be corrected, because in the abstract of Macias et al. (2015),

*"In the eastern basin, on the contrary, all model runs simulate an increase in surface production linked to a density increase (less stratification) because of the increasing evaporation rate."* 

We thank the reviewer for pointing out this mistake, we meant "decreased stratification". The wording in that sentence has been changed accordingly.

2) Line 81 – "This study aims at understanding the biogeochemical response of the Mediterranean to a "business–as–usual" climate change scenario throughout the 21st century." The objective of the study should be more detailed. Please, highlight, for example, that the study mainly focus on the influence of the external sources of nutrients (rivers, Atlantic).

We changed this sentence to "This study aims at understanding the biogeochemical response of the Mediterranean to a "business-as-usual" climate change scenario throughout the 21st century, **separating the effects of climate, nutrient inputs from rivers and changes in nutrient fluxes through the Strait of Gibraltar**."

3) Lines 127 - 139 – "Temperature and salinity increase strongly, leading to a decrease in surface density and an overall increase in vertical stratification. Average sea surface temperature of the Mediterranean rises by up to 3°C by the end of the century. However, the temperature rise is not homogeneous in the basin, regions such as the Balearic, Aegean, Levantine and North Ionian undergo a more intense warming (over 3.4 °C) probably due to the addition of the atmosphere-originated quasi-homogeneous warming with the local effect of surface current changes. The salinity increases by 0.5 (practical salinity units) on average across the basin. In the A2 simulation, the entire Mediterranean basin is projected to become more stratified by 2100 and deep water formation is generally reduced. These variations in hydrological characteristics of the water masses generate important changes in the circulation and in particular in the vertical mixing intensity. The strong reduction in vertical mixing observed in all deep water formation areas of the basin is linked with the changes in salinity and temperature of the water masses."

This paragraph needs to be re-write because there are some repetitions, for example, about the stratification and vertical mixing.

We separated this paragraph and reformulated it:

"Average sea surface temperature of the Mediterranean rises by up to 3°C by the end of the century.

However, that warming is not homogeneous across the basin, with regions such as the Balearic, Aegean, Levantine and North Ionian undergoing greater warming (>3.4 °C) probably due to the addition of the atmosphere-originated quasi-homogeneous warming being combined with local changes in surface currents. The salinity increases by 0.5 (practical salinity units) on average across the basin. These changes in hydrological characteristics generate substantial changes in the circulation and in particular the

vertical mixing intensity. Under the A2 scenario, the Mediterranean basin is projected to become more stratified by 2100. Consequently, deep-water formation is generally reduced."

## 4) Line 166 – "Nutrients input from rivers are derived from Ludwig et al. (2010) before 2000, Dissolved inorganic carbon (DIC) and Si are derived from Ludwig et al. (2009)."

What are included in "Nutrients"? Replace "Dissolved inorganic carbon" by "Dissolved Inorganic Carbon". "Si" means silicates? DIC and Si inputs are derived from Ludwig et al. 2009?

The nutrients that are mentioned include nitrate ( $NO_3$ ), phosphate ( $PO_4$ ), and dissolved organic carbon (DOC). We added these precisions.

"Nutrient inputs from rivers, including  $NO_3^-$ ,  $PO4_3^-$  (hereafter noted  $NO_3$  and  $PO_4$ ), and dissolved organic carbon (DOC) are derived from Ludwig et al. (2010). Dissolved inorganic carbon (DIC) and Si are derived from Ludwig et al. (2009)."

5) Line 190 – "This may result in higher nutrient concentrations at the river mouth. [...] However, nutrients from river discharge are consumed rapidly at proximity of the river mouth and we believe these potential higher concentrations don't have a large impact of the results."

## If all nutrients from river discharge are consumed near the river mouths, why did you study the influence of the river inputs at the scale of the Mediterranean basin?

At the time that the above-mentioned sentence was written, we meant that the effects of a particular river would be locally confined and would not substantially alter the mean state of the entire basin. However, our results do show that river inputs do have important effects on the regional scale. Hence we have now deleted this confusing sentence. For further clarification we also rearranged other sentences of the same paragraph.

## 6) Line 205 – "This period was chosen in order to avoid including in the CTRL years with too important warming such as the 1980s and 1990s." Need to be corrected.

Warming trends were already observed in the Mediterranean in the 1980s and 1990s (Adloff et al., 2015, HIS period), we chose to use a CTRL basis that does not include this trend.

Figure 12 from Adloff et al (2015), reproduced below, shows the simulated Mediterranean average SST between 1961 and 2100. It illustrates that in the HIS simulation (black line), i.e., the physical forcing we used in this article, the Mediterranean surface waters have a warming trend from 1980 onward. In the revised manuscript, we mention the same figure in this context.



Fig. 12 Yearly mean time series of sea surface temperature anomalies (vs. 1961–1990) averaged over the Mediterranean basin. A2-F, A2-RF and A2-ARF curves are overlapping. The spread of the ensemble is *shaded* in *grey* 

## 7) Line 212 – "present-day conditions [...] present-day conditions". Need to remove these expressions, or to define them.

We changed the sentence to "In order to separately quantify the effects of climate and biogeochemical changes, we made 2 additional control simulations: (1) CTRL\_R with climatic and Atlantic conditions corresponding to present–day conditions (no scenario for climate change or nutrient fluxes through the Strait of Gibraltar) and river nutrient discharge following the scenario evolution, and (2) CTRL\_RG with present-day climatic conditions, but river nutrient discharge and Atlantic buffer–zone concentrations following the scenario conditions."

8) Line 235 – "from satellite estimations from MyOcean Dataset (http://marine.copernicus.eu)". Which product of MyOcean, quote the full link to access this dataset. Done

9) Line 236 – "All chlorophyll values in the article and the data are chlorophyll–a." I do not understand. Do you mean that the word "chlorophyll" stands for "chlorophyll-a" throughout the manuscript? If so, it needs to be define when you use the word chlorophyll for the first time.

This is the first time we use chlorophyll in the article, we define here that we only use chlorophyll-a data. "Whenever we refer to chlorophyll, we always mean chlorophyll–a (hereafter noted chl-a)."

#### 10) Line 240 – "with values that", are you talking about the difference in magnitude?

In the revised manuscript, we have now clarified that sentence, saying "approximately 50% decrease in average chlorophyll concentration between the western and the eastern basin in the satellite data and 30 to 50% in the model."

11) Line 243 – "Moreover, several studies (see e.g. Claustre et al., 2002; Morel and Gentili, 2009) show that satellite estimates have a systematic positive bias in the coastal regions because of the high concentrations of colored dissolved organic matter and the presence of dust particles in seawater back scattering light."

*Need to be corrected. The bias in the coastal regions is due to the presence of sediment: turbid water (case-2 water, higher concentration of inorganic particles). The "general bias" over the Mediterranean* 

## basin is due to the presence of colored dissolved organic matter in seawater and other components (not well known yet) that modify the optical properties of the seawater.

Corrected to "because of the presence of particulate matter (for instance, sediments). The general bias observed in the Mediterranean is linked with colored dissolved organic matter and the presence of dust particles in seawater, which cause light back scattering."

#### 12) Line 247 – "the average chlorophyll surface concentration". That has been normalized?

Yes, we added precision: "Figure 2 provides an evaluation of the average normalized chl-a surface concentration evolution over the entire basin for the period 1997–2005."

## 13) Line 252 – "2 independent datasets" Replace "2" by "two". These datasets are not independent, the original data are from the same satellite sensors.

Reviewer is right, in the revised manuscript, we now write "two different datasets".

## 14) Line 267 – "200 m is 233 ± 146 10-9 g L-1 (average over the 1991–2005 period), while the model value for the HIS/A2 simulation over the same period is $159 \pm 87 \ 10-9 \ g \ L-1$ ." The unit should be modified into: $\mu g \ L-1 \ or \ mg \ m-3$ , and this sentence should be in the previous paragraph.

We changed all chlorophyll units to ng  $L^{-1}$  and put the sentence in the previous paragraph.

## 15) Maybe the paragraphs in the method (Section 2.2, lines 122 - 139) should be located into the Section 3.2.

We placed this paragraph in the Methods because these are not the results from our study but from Adloff et al (2015).

#### 16) Lines 290 – 295, already mentioned lines 210 – 219.

These sentences have now been removed.

## 17) Line 301 – "Phosphate content in the entire Mediterranean has increased in our simulation by 6 % over the 21st century..."

*Line 310 – "However, climate change effects lead to a global enhancement of 10 % in phosphate content in 2080–2099 in comparison to 1980–1999."* 

#### Is it 6 % or 10 %?

The difference between CTRL and HIS/A2 indicates an increase in phosphate concentrations of 6% ("Phosphate content in the entire Mediterranean has increased in our simulation by 6 % over the 21st century, as determined by the difference between CTRL and HIS/A2 simulations between 1980-1999 and 2080—2099"). This enhancement is the result of both climate and external nutrient fluxes changes (we added this precision in the text).

The effects of climate change alone (difference between HIS/A2 and CTRL\_RG) yield a more important increase in phosphate concentrations. This shows that future changes in climate and external nutrient supply may have opposite effects on nutrient concentrations in the Mediterranean.

We modified this paragraph lines (340-355): "Total phosphate content in the entire Mediterranean grew in our HIS/A2 simulation by 6 % over the 21st century, as determined by the difference between CTRL and HIS/A2 simulations between 1980-1999 and 2080–2099. The increase is larger in the eastern basin than in the western basin. In particular, there is an 8 % increase in phosphate content in the Ionian–Levantine sub–basin. Nutrient content in the HIS/A2 simulation is affected by both climate and nutrient fluxes from external sources (rivers and fluxes via the Strait of Gibraltar). The effects of changes in riverine input of phosphate are derived from the CTRL\_R simulation (see also Figure 5). The difference of phosphate content between CTRL and CTRL\_R are substantial over the first half of the century. We observe 3 % decrease in phosphate content in the entire Mediterranean between 1980–1999 and 2030–2049 due to river input changes (difference between CTRL\_R and CTRL). Changes in phosphate fluxes thought the Strait of Gibraltar seem to have limited effect on the global Mediterranean phosphate content as revealed by the small difference between the beginning and the end of simulation CTRL\_RG. Conversely, climate change enhances the basin-wide phosphate content by 10 % in 2080–2099 relative to 1980–1999 (HIS/A2 minus CTRL\_RG). Thus future changes in climate and external nutrient supply may have opposite effects on nutrient concentrations in the Mediterranean."

## 18) Line 384 - "P and N", you wrote phosphate and nitrate before, and now P and N. Please, stay consistent throughout the manuscript.

We corrected to 'phosphorus' and 'nitrogen'. However, the sedimentation fluxes in PISCES are total (inorganic and organic) phosphorus and nitrogen and not just phosphate and nitrate because these fluxes are calculated from organic carbon.

*19) Line 417 – "Nutrient concentrations in the eastern part…" Replace nutrient by phosphate.* Done

**20)** Line 422 – "a large annual variability" Replace by "a large interannual variability". Done

21) Line 427 – In the previous paragraph, about the western basin, some suggestions/interpretations were included. However, in this paragraph about the eastern basin, there were no suggestions to explain your results, why?

We added a sentence about the eastern basin indicating that its phosphate concentrations are probably influenced primarily by climate change throughout the 21<sup>st</sup> century: "*These results show that the 21st century evolution of phosphate concentrations in the eastern Mediterranean is mainly driven by climate change. Figure 7 shows that average PO4 concentrations in CTRL, CTRL\_R and CTRL\_RG are similar for all periods in the eastern basin.*"

22) Line 459 – "In the Mediterranean Sea, primary productivity is mainly limited by these 2 nutrients and their evolution in the future may impact the productivity of the basin." Not necessary, the sentence can be removed.

It has now been removed.

23) Line 461 – "showing an accumulation of nitrate in large zones of the basin," add "by the end of the century".

Done

24) Line 463 – "...and a small area in the southeastern Levantine" Also in the Gulf of Lion and south of Crete.

We have now added references to these two areas mentioned by the reviewer.

25) Line 465 – "...except near the mouth of the Nile and in the Alboran Sea." Also in the Ionian Sea, in the Tyrrhenian Sea, Algerian basin and between Crete and Cyprus.

We have now also mentioned these other areas.

26) Line 466 – "the N:P ratio (i.e. increase in P discharge and decrease in N discharge) in this river in our scenario." Replace by "the N:P ratio in this river in our scenario (i.e. increase in P discharge and decrease in N discharge).".

#### Done

27) Line 471 – "All the most productive zones of the beginning of the century are reduced in size and intensity by the end of the century." This statement does not convince me... Too broad and unclear... We have now clarified this sentence, writing as follows: "All the most productive zones at the beginning of the century are reduced in size and intensity by the end of the century. For instance, we observe a 10 to 40 % decrease in primary production in the Gulf of Lion and around the Balearic Islands, and more than a 50 % reduction in the North Adriatic basin, in the Aegean Sea and in the eastern Levantine basin around Cyprus."

#### 28) Line 481 – You already mentioned this area around Cyprus in the paragraph before.

We integrated this paragraph to the previous one. See lines 473 to 479: *"For instance, there is a 10 to 40 % decrease in primary production in the Gulf of Lion and around the Balearic Islands, more than 50 % reduction in the North Adriatic basin, in the Aegean Sea and in the eastern Levantine basin around Cyprus. There is also a reduction in primary productivity from 40-50 gCm<sup>-2</sup> year<sup>-1</sup> to 20-30 gC m<sup>-2</sup> year<sup>-1</sup> around Cyprus. These mesoscale changes may be linked with changes in local circulation (e.g., mesoscale eddies). These observations show that the evolution of the Mediterranean biogeochemistry is influenced by both meso- and large-scale circulations patterns."* 

#### 29) Line 499 – "South Adriatic", North Adriatic?

We meant more the north eastern Ionian basin. We also observe more P limitation in the coastal South Adriatic.

30) Line 501 – "One specificity of Mediterranean biology is that most planktonic productivity occurs below the surface at a depth called the deep chlorophyll maximum (DCM). Hence, most of the chlorophyll concentration is not visible by satellites (Moutin et al., 2012)." I do not think that these sentences are necessary, and I do not think that this satellite limitation is discussed in Moutin et al., 2012.

We removed these sentences.

*31)* Line 508 – "as the South Ionian and the Tyrrhenian basin." Not visible in these areas, mainly visible in the south of Crete.

We changed the sentence to refer to the North Ionian and the South of Crete.

#### 32) Line 511-524 – statements in this paragraph are not supported by the results. For example,

<sup>2</sup> "but surface concentration is enhanced by about 25 10-9 g L-1", that means 0.025  $\mu$ g L-1, which represents almost nothing. We agree this is a very small variation and make no conclusions about this value. We changed the sentence to "but surface concentration is decreased by about 25 10<sup>-9</sup> g L<sup>-1</sup>, which is negligible."

<sup>2</sup> "This shows that local variability in the Mediterranean circulation and biogeochemistry is important." A general statement not supported by results. We deleted this sentence

**"** "the average chlorophyll concentration is reduced by almost 50 %", looking at figure 14a, there is not a 50 % decrease in the chlorophyll concentration.

We thank the reviewer for this remark and deleted this sentence that indeed was referring to previous results that have been corrected since. We rearranged some sentences in this paragraph in an effort to make it clearer.

See lines 510 to 523: "Figure 14 shows the average vertical profiles of chl-a at the DYFAMED station (43.25° N, 7.52° E) and the average profiles for the western and eastern basins for the 1980–1999 and 2080–2099 periods. The subsurface chl-a maximum persists through to the end of the century. At the DYFAMED station, the average DCM depth remains unchanged, while the surface chl-a concentration is decreased by about 25 ng L<sup>-1</sup>, which is negligible. Thus the average chl-a profile at DYFAMED changes little throughout the simulation. However, at that station there is approximately 40 % variability in the chl-a concentration profile and depth of DCM for some month (not shown). In the western basin, the subsurface maxima at present and in the future are located at the same depth (100–120 m), but the average chl-a concentration in the DCM increases by about 15 to 20 ng L<sup>-1</sup> during the simulation. However, where there are small changes in the average chl-a profiles in the 520 western basin, those are often accompanied by greater local changes in the depth and intensity of the DCM (Figure 13). In the eastern basin, subsurface chl-a concentration is reduced by about 50 ng L<sup>-1</sup> and the subsurface chl-a maximum deepens from 100–120 m to below 150 m."

## *33)* Lines *527, 532, 533 – all percentage quoted in the text are different in Table 4. Please, double-check the values.*

We thank the reviewer. We have now corrected these values that were the surface difference values and not the integrated values.

#### *34) Line 546 and 556 – lack of quantitative estimates.*

#### We have no added some numeric values.

See for instance lines 448-450: "In HIS/A2, the biomass of both phytoplankton classes declines during the simulation (-0.01 mmolC  $m^{-3}$  for nanophytoplankton and - 550 0.03 to -0.04 mmolC  $m^{-3}$  for diatoms)." And lines 558-560: "In HIS/A2 in all basins there is a decrease in microzooplankton concentration during 1980–2000 (from 0.165 to 0.114 mmol  $m^{-3}$ ), after which it remains stable and consistently below the CTRL values until the end of the simulation in 2100."

## *35)* Section 4.1 Biogeochemical forcings – Not a good title. This section mainly discussed the influences of external sources of nutrients

We have rearranged the discussion. Section 4.1 is now titled "Sources of uncertainties" and subdivided in 3 subsections: 4.1.1 Climate change scenario, 4.1.2 Uncertainties from the PISCES model and 4.1.3 External nutrient sources.

36) Line 617 – "these regions" replace by "this region". Done

37) Line 624 - "2" replace by "two" Done

38) Line 701 - close parenthesis. Done

*39)* Figure A3a – It is impossible to read the colorbar.

We have changed the colorbar of this figure.

This revised manuscript is improved from the previous version and the authors have taken on board the comments by the reviewers. There is now quantitative arguments throughout the manuscript and I accept what the authors say regarding the limitations of the model and that this should not stop the manuscript from being published as it is the best model and model inputs that they have available. They have clarified what is in the model and now acknowledge that organic P and N may also affect the budget. In addition they have created a nice figure summarising the inputs and outputs of phosphate and nitrate to the basin. However, I have still found this manuscript relatively difficult to read in places with the keys points lost within the text. There are now places with extremely long paragraphs (i.e lines 166-195, 331-362, 589-620, 696-731) and in these paragraphs it becomes unclear which key point the author wanted the reader to get from the it. I think both the results and discussion sections can still be improved and there should be increased emphasis on the impact of the results in the discussion. I feel that the results the authors present are important and should be published but at present their impact does not come across strongly enough in the discussion.

We thank the reviewer for their comments. In this revised manuscript, we tried to improve the presentation of the results by separating section 3.3.5 in 2 different paragraphs: one for phosphate, and one for nitrate. In each of these paragraphs, we first describe the results in the western basin (with the order: surface, intermediate and deep waters) and then in the eastern basin.

We reorganized the discussion and in particular emphasize the implications and impact of our results in section 4.2.

Lines 166-195: There is now a very long paragraph in the method section discussing the riverine input. The authors have addressed my concerns in this paragraph but have additionally added further sentences which in my opinion become too detailed in regards to differences between the different inputs (Ludwig vs Adloff etc ) in different models which I find confusing. In addition, there is now a lot of repetition between this section and the first paragraph of the discussion. Although the authors obviously do need to acknowledge the potential errors in their results I think these sections can be reduced.

#### reduced.

We moved the part of this paragraph describing the uncertainties of the river nutrient fluxes to the discussion as these arguments were repeated.

Lines 665 to 680: "For the riverine nutrient inputs, scenarios from the MEA report are based on different assumptions from the IPCC SRES scenarios used to compute freshwater runoff in the HIS/A2 simulation. Freshwater discharge from Ludwig et al. (2010) is based on the SESAME model reconstruction and differs from freshwater runoff in the ARPEGE–Climate model used to force our physical model. This may lead to incoherences between water and nutrient discharges, but the nutrient discharges from Ludwig et al. (2010) are the only ones that are available. Furthermore, the SESAME model is not coupled with NEMO/PISCES. Associated discrepancies and the uncertainties linked with the use of inconsistent scenarios in our simulation should be addressed by developing a more integrated modelling framework to study the impacts of climate change on the Mediterranean Sea biogeochemistry. As there is no consensus nor validated scenario for nutrient fluxes from riverine runoff in the Mediterranean, we chose to use one scenario from Ludwig et al. (2010). This scenario has the advantage of being derived from a coherent modeling framework. However, the Ludwig et al. (2010) nutrient discharge transient scenario does not represent the interannual variability of nutrient runoff from rivers. Moreover, according to these authors, the socio–economic decisions made in the 21st century will influence nitrate and phosphate discharge over the Mediterranean. It is difficult to forecast these decisions and the resulting changes in nutrient fluxes are uncertain."

# Section 3.3: I still find this section hard to follow. The authors have now added quantitative metrics in this section but they are generally put in brackets rather than integrated into the text which is making it awkward to read. In addition although I appreciate that the authors have tried to change this section, the authors still explain trends such as decrease in phosphorus content by decrease in riverine inputs before presenting the river inputs and therefore are having to repeat things. I suggest putting the results on P and N content after discussing the other terms in the budget.

We moved the 3.1 section after the description of nutrient fluxes in and out of the basin. We also tried to rephrase parts of the section to include the quantitative metrics in the text.

Discussion: The discussion feels dominated by statements about the limitations/uncertainties of the model with weak statements on the impact and interpretation of results. I think it would improve the manuscript if there was a better integration of the literature with the authors own arguments and conclusions from this study within the discussion. In this revised manuscript the authors have added additional comparisons with other literature which is important and I think was needed, but it currently reads as a list (Lines 696-731). The authors are trying to justify why their results are different than what is in the literature rather than using the literature to put their results into context and strengthen their arguments. For example, in lines 727-731 rather than explaining why you can not observe the effects of temperature on nutrient recycling within this model, you can maybe say how an increase in nutrient cycling due to warmer temperatures may strengthen or weaken your conclusions.

We rearranged and reformulated the discussion following recommendations from another reviewer. We have separated the discussion on the uncertainties of the study in section 4.1. We try to emphasize the originality and the impact of our results in section 4.2.

In particular, our results show that the effects of climate change, riverine input and nutrient fluxes through the Strait of Gibraltar may have synergistic or antagonistic effects depending on the depth and the region.

See lines 737 to 744: "Results from our transient simulations show that nutrient concentrations may evolve differently depending on the region and the depth in response to climate change and external nutrient inputs. In the surface western Mediterranean, the effects of climate change and enhanced nutrient fluxes via Gibraltar both concur to the increase in nutrient concentrations (Figures 7a and 8a). In the surface eastern basin, river fluxes of nitrate and stratification have opposing effects on nitrate concentrations whereas phosphate concentrations are mainly driven by climate change effects (Figures 7d and 8d). The difference between HIS/A2 and CTRL\_RG phosphate concentrations in the intermediate and deep layers (Figures 7b, 7e, 7c, 7f) indicates that variations of phosphate concentrations during the 21st century are primarily driven by climate change while nitrate concentration is equally sensitive to changes in biogeochemical forcings (Figures 8b, 8e, 8c, 8f)."

We also report that the changes in nutrient concentration have consequences on the entire ecosystem. See lines 789 – 794: "Chust et al. (2014) have shown that regional seas and in particular the Aegean and Adriatic were sensitive to trophic amplification. Our results appear to agree, showing signs of trophic amplification (see Figures 15 and 16). Assessing the sensitivity of the Mediterranean to trophic amplification would require more simulations focused on the evolution of Mediterranean planktonic biomass under different climate change scenarios."

In general, we tried to use the literature to strengthen our conclusions instead of simply comparing our results. See for instance lines 661-664: "Herrmann et al. (2014) simulated an increase in chl-a

concentration associated with climate change effects in a small region of the north western basin by the end of the century. Thus, the separate effects of climate change and external nutrient inputs may have synergetistic effects on the evolution of the western Mediterranean chl-a."

Lines 673-675:" Lazzari et al. (2014) also conclude that the river mouth regions are highly sensitive because the Mediterranean Sea is influenced by external nutrient inputs. The choice of river runoff scenario will likely influence the evolution of nutrient concentrations and the biogeochemistry in many coastal regions such as the Adriatic Sea (see also Spillman et al., 2007)."

Along these lines the discussion on N and P limitation (Lines 713-726) could be a paragraph/section to itself. In this section the authors state the their results are "in contrast with previous literature on the matter" (Lines 715-716). However there is evidence within the literature for P and N co-limitation in the Mediterranean. Whilst generally the spring phytoplankton bloom is P limited, N and P co-limitation has been observed, especially during the stratification period (Thingstad et al., 2005; Tanaka et al., 2011) and there is some evidence of the spring phytoplankton bloom being N and P co-limited in the Western Mediterranean (Pasqueron de Fommervault et al., 2015) or even N limited (Marty et al. 2002). In addition N limitation has been predicted in the Alboran Gyre as well (Ramirez et al., 2005; Lazzari et al., 2016). What time period do you calculate the N and P limitation for (i.e annual mean, spring bloom etc)? This may also affect what you are predicting compared to the literature

We separated this paragraph from the rest of the text (lines 782-791) and modified in an effort to focus it more on the nutrient co-limitation:

"The modifications of chl-a production and plankton biomass are linked to changes in nutrient limitation (Figure 12). Finding no clear definition of nutrient co-limitation, we consider that N and P are co-limiting when the difference in limitation factors is less than 1 %. This definition of nutrient co-limitation applies well to the Mediterranean case because of its very low nutrient concentrations. Our results are confirmed by some studies (Thingstad et al 2005, Tanaka et al. 2011). However, our nutrient limitations are calculated from 20–years average nutrient concentrations and nutrient limitation may vary greatly during the seasonal cycle (Marty et al. 2002, Diaz et al 2001)."

Finally, at other reviewers suggestion the authors have now included a scenario on atmospheric deposition but only present this within the discussion (Lines 621-638). I feel it should be fully integrated into the text (i.e in the methods and results section) rather than tagged onto the discussion. It does provide some important insite on the effect of climate change despite only considering a climatology of atmospheric inputs rather than potential future ones. The authors could further hypothesise what potential future changes in atmospheric deposition may have on the results in the discussion based upon regional projections of atmospheric inputs into the future (i.e Lambarque et al., 2013). Whilst I appreciate they can't actually run a scenario they may be able to comment on whether it is likely to enhance/dampen the trend they see.

We initially decided to keep the description of these simulations in the discussion as they are not the main focus of the study, and the aerosol deposition forcings are not evolving during the 21<sup>st</sup> century.

As suggested, we included these simulations in the main text. In the methods section see lines 175 to 183: "There is, to our knowledge, no transient scenario for the evolution of atmospheric deposition over the Mediterranean Sea. However, in order to evaluate the potential effects of aerosol deposition on the future Mediterranean Sea, we used deposition fields of total nitrogen deposition ( $NO_3 + NH_4$ ) from the global model LMDz-INCA (Hauglustaine et al 2014) and phosphate deposition from natural dust modeled with the regional model ALADIN-Climat (Nabat et al 2015) respectively (see Richon et al 2017 and references therein for the description and evaluation of the atmospheric models) The atmospheric

deposition fields represent present-day aerosol deposition fluxes (1997-2012 and 1980-2012 for total nitrogen and dust deposition respectively) that are repeated over the 1980-2099 simulation period."

And lines 211 to 216: "We made two supplementary simulations, one with total nitrogen deposition (HIS/A2\_N) and another with total nitrogen and natural dust deposition (HIS/A2\_NALADIN). These simulations include climate change and nutrient fluxes from rivers and via the Strait of Gibraltar that follow the scenario conditions. The results from these simulations should be considered as exploratory. Nonetheless, they provide insight into the potential effects of future aerosol deposition."

## We added a result section lines 595 to 604: "Effects of aerosol deposition on surface primary productivity"

"Figure 17 shows the relative effects of total nitrogen and natural dust deposition on surface primary production in 1980-1999 and 2080-2099. As shown in Richon et al 2017, dust deposition enhances surface primary productivity in the southern part of the basin in 1980-1999 whereas nitrogen deposition enhances primary productivity in the northern part of the basin. As our HIS/A2 simulation shows a decrease in surface PO<sub>4</sub> concentrations, thus accentuating phosphate limitation over the Mediterranean basin by the end of the 21st century, the relative impact of phosphate deposition from dust would be enhanced in the 2080–2099 period relative to the 1980–1999 period. Conversely, nitrogen atmospheric deposition has very little effect on Mediterranean primary production at the end of the simulation period because most of the basin is not N-limited."

In the discussion section, we tried to emphasize the fact that even if phosphate deposition seems to relieve a part of the phosphorus limitations at the end of the century, the Mediterranean is still mainly P-limited in 2100. However, many aerosol sources such as volcanoes, anthropogenic sources and organic phosphorus and nitrogen are not included in our sources. Therefore, it is important to develop models and measurements of these aerosols as they may have important impacts on the Mediterranean surface biogeochemistry.

#### See lines 706 to 729:

"The biogeochemistry of the Mediterranean is significantly influenced by aerosol deposition (e.g. Krom et al., 2010; Dulac et al., 1989; Richon et al., 2018, 2017; Guieu et al., 2014). The future evolution of the multiple aerosol sources surrounding the Mediterranean will likely influence the response of the Mediterranean to climate change. Results from the HIS/A2 NALADIN simulation show that enhanced phosphate fluxes from aerosols may limit the surface decrease of phosphate concentrations and limit phosphorus limitation. However, in the HIS/A2 NALADIN simulation, the surface Mediterranean is still Plimited in most of the Mediterranean because the atmospheric nutrient fluxes are low in comparison to riverine nutrient fluxes from rivers and the nutrient flux through the Strait of Gibraltar (see Richon et al., 2017). Therefore, it appears unlikely that changes in aerosol deposition from natural dust would greatly influence future Mediterranean biogeochemistry. However, there are multiple sources of aerosols that are not included in atmospheric models, e.g., anthropogenic, volcanic and volatile organic compounds (e.g. Wang et al., 2014; Kanakidou et al., 2016). Their combined influence could perhaps constitute an important nutrient flux to the Mediterranean, thus altering the evolution of its biogeochemistry. Moreover, aerosols affect radiative forcing over the Mediterranean and may impact the climate conditions (Nabat et al., 2015b). Thus, efforts should be made to accurately represent this nutrient source in Mediterranean models to assess the effect on Mediterranean Sea biogeochemistry with regards to climate change. Our results show that the state of the Mediterranean biogeochemistry at the end of the 21st century is the result of the combined evolutions of both climate and external nutrient fluxes. Therefore, it is very difficult to predict the future evolution of the Mediterranean based on the evolution of one of these components only. This is why it is important, in the case of semi-enclosed basins, to produce reliable estimates of the evolution of all the components influencing the biogeochemistry."

Figure 18: I suggest reversing the input and output arrows through the Strait of Gibraltar so that they are the same as the actual water flow. Currently it is suggesting an estuarine flow rather than anti estuarine.

We thank the reviewer for this suggestion. We changed the arrows.

Manuscript prepared for Biogeosciences with version 2015/09/17 7.94 Copernicus papers of the LATEX class copernicus.cls. Date: 15 December 2018

### **Biogeochemical response of the Mediterranean Sea to the transient SRES–A2 climate change scenario**

Camille Richon<sup>1,\*</sup>, Jean-Claude Dutay<sup>1</sup>, Laurent Bopp<sup>2</sup>, Briac Le Vu<sup>2</sup>, James C. Orr<sup>1</sup>, Samuel Somot<sup>3</sup>, and François Dulac<sup>1</sup>

 <sup>1</sup>LSCE/IPSL, Laboratoire des Sciences du Climat et de l'Environnement, CEA-CNRS-UVSQ, Gif-sur-Yvette, France
 <sup>2</sup>Laboratoire de Météorologie Dynamique, LMD/IPSL, Ecole Normale Supérieure - PSL Research Univ., CNRS, Sorbonne Université, Ecole Polytechnique, Paris, France
 <sup>3</sup>CNRM, Université de Toulouse, Météo-France, CNRS, Toulouse, France
 \*Now at: Department of Earth, Ocean and Ecological Sciences, School of Environmental Sciences, University of Liverpool, Liverpool L69 3GP, UK

Correspondence to: Camille Richon (crichon@liverpool.ac.uk)

Abstract. The Mediterranean region is a climate change hot-spot. Increasing greenhouse gas emissions are projected to lead to a significant warming of Mediterranean Sea waters, substantial warming of the Mediterranean Sea as well as major changes in its circulation, but the subsequent effects of such changes on marine biogeochemistry are still-poorly understood. Our Here, our aim is to inves-

- 5 tigate the changes in how climate change will affect nutrient concentrations and biological productivity in response to climate change in the Mediterranean region Mediterranean Sea. To do so, we perform transient simulations with the coupled high resolution model NEMOMED8/PISCES using the high-emission IPCC SRES-A2 socio-economic scenario and corresponding Atlantic, Black Sea, and riverine nutrient inputs. Our results indicate that nitrate is accumulating in the Mediter-
- 10 ranean Sea over the 21st century, whereas no tendency found for phosphorus. These contrasted variations while phosphorus shows no tendency. These contrasting changes result from an unbalanced nitrogen-to-phosphorus input from fluxes through nitrogen-to-phosphorus input from riverine discharge and fluxes via the Strait of Gibraltarand riverine discharge and , which lead to an expansion of phosphorus-limited phosphorus-limited regions across the Mediterranean. In addition, phy-
- 15 toplankton net primary productivity is reduced by 10 % in the 2090s in comparison to the present state, with reductions of up to 50 % in some regions such as the Aegean Sea as a result of nutrient limitation and vertical stratification. We also perform sensitivity tests in order to study separately to separately study the effects of climate and biogeochemical input changes on the Mediterranean future state of the Mediterranean Sea. Our results show that changes in nutrient supply
- 20 from the Strait of Gibraltar and from rivers and circulation changes linked to climate change may have opposing antagonistic or synergistic effects on nutrient concentrations and surface primary productivity. In some regions such as the Adriatic Sea, half of the biogeochemical changes observed throughout simulated during the 21st century are linked with external nutrient input changes changes

in nutrient input while the other half can be are linked to climate changeeffects. This article is a first

25 step in the study of . This study is the first to simulate future transient climate change effects on the Mediterranean Mediterranean Sea biogeochemistry, but calls for presently missing atmospheric deposition scenario and coordinated multi-model efforts to explore the various uncertainty sources of such a future projection further work to characterize effects from atmospheric deposition and to assess the various sources of uncertainty.

#### 30 1 Introduction

The Mediterranean basin-Sea is enclosed by three continents, with and is surrounded by mountains, deserts, rivers, and industrialized cities. This area evaporative basin is known as one of the most oligotrophic marine environment environments in the world (Béthoux et al.) [1998). Because of its high anthropogenic pressure and low biological productivity, this region is predicted-likely to be highly

- 35 sensitive to future climate change impacts (Giorgi, 2006; Giorgi and Lionello, 2008). Records of the past evolution of the Mediterranean circulation show that the Mediterranean has undergone abrupt changes in its circulation patterns over ancient times. In particular, high stratification events, characterized by the preservation of organic matter in the sediments known as sapropels, have been recorded several times through geological history. The most recent was
- 40 recorded of such event occurred 10 000 years ago and lasted about 3 000 years. This accumulation of organic matter in the sediments is interpreted as the result of a strong stratification of the water column leading to suboxic deep layers (e.g. Rossignol-Strick et al., 1982; Rohling, 1991, 1994; Vadsaria et al., 2017). In more recent times, abnormal winter conditions have led to changes in deep water formation, such as the Eastern Mediterranean Transient (EMT) event that occurred during
- 45 the early nineties and had chemical impacts such as an increase in the Levantine basin salinity (see Theocharis et al.) [1999; Lascaratos et al.] [1999; Nittis et al.] [2003; Velaoras and Lascaratos, [2010; Roether et al.] [2014). Also, changes in the North Ionian Gyre circulation triggered the so-called Bimodal Oscillating System (BiOS) that influenced phytoplankton bloom in the Ionian Sea through the modification of by modifying water transport that led to modified nutrient distribution and altered
- 50 local productivity (Civitarese et al., 2010). These events show that a semi-enclosed basin with short residence time of water (around 100 years, see Robinson et al., 2001) such as the Mediterranean Sea is highly sensitive to climate conditions and that perturbations of these conditions can modify the circulation, ultimately leading to changes in the its biogeochemistry.

Future climate projections with greenhouse gases high-emission scenarios yield an increase in temperature
and a decrease in for greenhouse gases simulate warming and reduced precipitation over the Mediterranean region (Giorgi, 2006; IPCC, 2012) leading to warmer and saltier seawater (Somot et al.,
2006; Adloff et al., 2015). As a result of these changes, the Mediterranean thermohaline circulation (MTHC) may significantly change with a consistent weakening in the western basin and a less

certain response in the eastern basin in elimate change scenarios characterized by high greenhouse

- 60 gases emission for such high emission scenarios (Somot et al.) 2006; Adloff et al.) 2015). In all simulations under the A2 runs, Adloff et al. (2015) show an increase in the stratification index at the end of the 21st century. This scenario, Adloff et al. (2015) find an increased stratification index in 2100. This increase will likely weaken the vertical mixing and may reduce nutrient supply in to the upper layer of the Mediterranean, a supply that is essential for phytoplankton to bloom (d'Ortenzio)
- 65 and Ribera d'Alcalà, 2009; Herrmann et al., 2013; Auger et al., 2014). Primary productivity in the ocean is influenced by water-its circulation and vertical mixing that brings available nutrients to phytoplankton (Harley et al., 2006). Changes in oceanic physics physical processes such as modification of vertical mixing can have dramatic effects on plankton community dynamics and ultimately on the productivity of the entire oceanic food web (Klein et al., 2003; Civ-
- 70 itarese et al., 2010). Few studies have investigated the sensitivity of the oligotrophic Mediterranean Sea to future climate change (e.g. Herrmann et al.) 2014, for the northwestern Mediterranean). Lazzari et al. (2014) investigated the effects of the A1B SRES (Special Report on Emissions Scenarios) moderate climate change scenario on the Mediterranean biological productivity and plankton communities. They performed short (10–year) non–transient simulations at the beginning and the end of
- 75 the 21st century and found a decreasing trend of phytoplankton biomass in response to this climate change scenario. Macias et al. (2015) simulated a "baseline" of expected consequences of climate change alone on the Mediterranean primary productivity. They found that according to the RCP 4.5 and RCP 8.5 scenarios, Under the RCP4.5 and RCP8.5 scenarios, their simulated integrated primary productivity over the eastern Mediterranean basin may increase increased as a result of density
- 80 changes (increased stratificationisolating the upper layer from the rest of the water columnchanges in density (decreased stratification). However, their results are based those results depend on non-transient simulations and present-day nutrient inputs. The response of the Mediterranean biogeochemistry to transient climate and biogeochemical change scenarios has never not been evaluated. Being As a semi-enclosed oligotrophic basin, the Mediterranean is highly sensitive to external nu-
- 85 trient inputs. Their origins are mainly from sources. Those sources include coastal runoff, river discharge (Ludwig et al.) [2009), Atlantic inputs through the inputs from the Atlantic via the Strait of Gibraltar (Gómez, 2003; Huertas et al.) [2012), and atmospheric deposition (see e.g. Dulac et al.) [1989; Christodoulaki et al.) [2013; Gallisai et al.) [2014; Guieu et al.) [2014; Richon et al.) [2017] [2018). Recent studies also showed that Other potentially important sources of nutrient supply include direct
- 90 wastewater discharge (Powley et al., 2016) and transfer by submarine groundwater (Rodellas et al., 2015)may be important sources of nutrient for the Mediterranean. However, these sources are to date not two potential sources have yet to be well quantified. This study aims at understanding to assess the biogeochemical response of the Mediterranean to a "business-as-usualbusiness-as-usual" climate change scenario throughout during the 21st century. For this purpose, we use, while distinguishing
- 95 effects from climate change, nutrient input from rivers, and changes in nutrient transport across the

Strait of Gibraltar. Thus we used the high resolution coupled physical-biogeochemical physical-biogeochemical model NEMOMED8/PISCES .- We model to simulate the evolution of biogeochemical tracers (nutrients, chlorophyll-a e.g., nutrients, chlorophyll-a concentration, plankton biomass and primary production) under the SRES A2 climate change scenario for over the 21st century (IPCC and Working

- 100 Group III, 2000). The choice of the A2 that scenario was driven by the availability of daily 3–D forcings for the biogeochemical model (physical forcings such as ocean currents, temperature and salinity, see Adloff et al., 2015). We are aware that using a single simulation will limit the robustness of our results. However the computer power required Although results from single simulation scenario must be used with caution, it is not currently feasible to make a more extensive assessment because
- 105 of the computational requirements to perform large ensembles with **PISCES** and the unavailability of NEMOMED8/PISCES offline model and because the 3-D daily ocean transient scenario data currently prevent a more extensive assessment forcing data are not available from simulations made with other scenarios.

This article is organized as follows: the coupled model, forcings and the different simulations are 110 first described. We briefly evaluate the biogeochemical model in Section 3.1 and present the evolution of the physical and biogeochemical forcings in Section 3.2. In section 3.3, we expose the temporal evolution of the main nutrients, their budgets in present and future conditions and discuss their impact on the biogeochemistry of the Mediterranean Sea in section 4.

#### 2 Methods

120

#### 115 2.1 The ocean model

The oceanic ocean general circulation model used in this study is NEMO (Madec, 2008) in its regional configuration for the Mediterranean Sea (NEMOMED8 Beuvier et al., 2010). The NEMOMED8 grid has a horizontal resolution of 1/8° stretched in latitude (i.e., with a resolution from 9 km in the North to 12 km in the South of the domain). The model has 43 vertical levels with varying thicknesses (from 6 m in the surface layer to 200 m in the deepest layer). The Atlantic boundary is closed at 11°W and tracers are introduced in a buffer zone between 11°W and 6°W.

Air-sea fluxes (momentum, heat, water) and river discharges discharge used to force NEMOMED8 are prescribed by the atmospheric Regional Climate Model ARPEGE-Climate (Déqué et al., 1994; Gibelin and Déqué, 2003) using a global and stretched grid, which has a 50-km horizontal resolution over the area of interest.

125

#### 2.2 The SRES-A2 scenario simulation

ARPEGE-Climate is itself driven by greenhouse gases (GHG) and aerosol forcings following the observations (up to year 2000) and the SRES-A2 scenario afterwards and by SST (Sea Surface Temperature) coming from a previously run CNRM-CM coupled GCM (General Circulation Model)

- simulation (Royer et al., 2002). In addition, the ocean component of CNRM-CM (a low resolution 130 NEMO version) provides the near-Atlantic conditions (3-D potential temperature and salinity) for NEMOMED8. The various forcings and the modeling chain from the GCM to the ocean regional model are described in details in Somot et al. (2006) and Adloff et al. (2015). The NEMOMED8 simulation (ocean physics and forcings) used here corresponds to one of the sim-
- ulations used and studied in Adloff et al. (2015), and more specifically the by Adloff et al. (2015), 135 i.e. their simulations labeled HIS (historical period 1961 to 1999) and A2 (A2 scenario period 2000– 2099) as listed in their Table 1. This physical run has already been simulation was previously used to study elimate change impacts on Mediterranean marine-impacts of climate change on Mediterranean Sea ecosystems (Jordà et al., 2012; Hattab et al., 2014; Albouy et al., 2015; Andrello et al., 2015).
- The main changes on the Mediterranean Sea physical changes (SST, SSS -Sea Surface 140 Salinity-, surface circulation, deep convection and thermohaline circulation, vertical stratification, sea level) are detailed in Adloff et al. (2015). Briefly, changes in temperature and precipitation in the A2 scenario lead to increased evaporation in the basin at the end of the 21st century. Freshwater inputs by 2100. Freshwater input from rivers and the Black Sea decrease along with total precipita-
- tion. This consequently leads to a significant, which in turn contributes to a substantial increase in 145 net transport through the Strait of Gibraltar (+0.018 Sv by the end of the century). Temperature and salinity increase strongly, leading to a decrease in surface density and an overall increase in vertical stratification.

Average sea surface temperature of the Mediterranean rises by up to 3°C by the end of the century.

- However, the temperature rise that warming is not homogeneous in across the basin, with regions 150 such as the Balearic, Aegean, Levantine and North Ionian undergo a more intense warming (over undergoing greater warming (>3.4 °C) probably due to the addition of the atmosphere-originated quasi-homogeneous warming with the local effect of surface current changes being combined with local changes in surface currents. The salinity increases by 0.5 (practical salinity units) on average
- across the basin. In the A2 simulation, the entire Mediterranean basin is projected to become more 155 stratified by 2100 and deep water formation is generally reduced. These variations-These changes in hydrological characteristics of the water masses generate important generate substantial changes in the circulation and in particular in the vertical mixing intensity. The strong reduction in vertical mixing observed in all deep water formation areas of the basin is linked with the changes in salinity
- 160 and temperature of the water masses. Under the A2 scenario, the Mediterranean basin is projected to become more stratified by 2100. Consequently, deep-water formation is generally reduced.

#### 2.3 The biogeochemical model

Here, the physical model NEMOMED8 is coupled to the biogeochemical model PISCES (Aumont and Bopp, 2006), already used for investigations in the Mediterranean basin (Richon et al., 2017),

165 2018). This Monod–type model (Monod, 1958) has 24 biogeochemical compartments including 2 phytoplankton (nanophytoplankton and diatoms) and 2 zooplankton size classes (microzooplankton and mesozooplankton). Phytoplankton growth is limited by the external concentration of five different nutrients: nitrate, ammonium, phosphate, silicic acid and iron. In this version of PISCES, elemental ratios of C:N:P in the organic matter are fixed to 122:16:1 following Takahashi et al. (1985).

- 170 There is no explicit bacterial compartment but bacterial biomass is calculated using zooplankton biomass (see Aumont and Bopp, 2006, for details). Organic matter is divided in 2 forms: dissolved organic carbon (DOC) and particulate organic carbon. The biogeochemical model is ran\_was\_run in offline mode (see e.g. Palmieri et al., 2015): biogeochemical quantities are passive tracers, they are transported following an advection–diffusion equation using dynamical fields (velocities, mixing co-
- 175 efficients...) ealeulated beforehand pre-calculated in a separate simulation with only the dynamical model NEMOMED8.

#### 2.4 Boundary and initial physical and biogeochemical conditions

External nutrient supply for the biogeochemical model <u>includes</u> inputs from the Atlantic Ocean and from Mediterranean rivers. We did not include atmospheric deposition as there is currently

- 180 no scenario for its future evolution. Similarly, we did not include submarine groundwater discharge and direct wastewater discharge as there is to date no climatology for these sources. Atlantic input is prescribed from water exchange through the Strait of Gibraltar in the NEMO circulation model along with the concentrations of biogeochemical tracers in the buffer zone. Nutrient concentrations in the buffer zone are prescribed from a global ocean climate projection using the A2 simulation
- 185 values from IPSL–CM5–LR (Dufresne et al., 2013) performed within the framework of the CMIP5 project (Taylor et al., 2012). Nutrient concentrations in the buffer–zone are relaxed to these values with a time constant of one month.

Nutrients input from rivers Nutrient inputs from rivers, including  $NO_3^-$ ,  $PO_4^{3-}$  (hereafter noted  $NO_3$  and  $PO_4$ ), and dissolved organic carbon (DOC) are derived from Ludwig et al. (2010) before 2000,

- 190 Dissolved inorganic carbon (DIC) and Si are derived from Ludwig et al. (2009). For the 21st century, we use the estimations for nutrient discharge proposed by Ludwig et al. (2010) of following the "Order from Strength" scenario from the Millenium Ecosystem Assessment (MEA) (Cork et al., 2005), which gives nitrateand phosphate, phosphate and DOC discharge per sub-basin in 2030 and 2050. Yearly values are obtained by linear interpolation between 2000 and 2030 and between
- 195 2030 and 2050, after which they are held constant until the end of the simulation in 2100. Seasonal variability coming from four of the largest rivers for Mediterranean and Black Sea (Rhône, Po, Ebro and Danube) is also included. According to Ludwig et al. (2010), the future trends in nutrient discharge from the major rivers of the Mediterranean stay within the interannual variability over of the past 40 years. This The "Order From Strength" scenario is based on hypotheses of very
- 200 little efforts made towards mitigation of climate change. <u>Moreover</u>, <u>Ludwig et al.</u> (2010) point out some substantial changes in the nutrient and water budget in specific regions. <u>However, total riverine</u>

nutrient input is not drastically changed at the basin scale. In particular, according to their scenario, the northern part of the Mediterranean has decreasing trends in nitrate discharge whereas it is increasing in the southeastern Levantine . Freshwater discharge from Ludwig et al. (2010) isbased on

- 205 the SESAME model reconstruction and differs from the ARPEGE-Climate model used here. This may lead to incoherences between water and nutrient discharges, but the nutrient discharges from Ludwig et al. (2010) are the only available values, and the SESAME model is not coupled with NEMO/PISCES. Adloff et al. (2015) evaluate the changes in total freshwater runoff in the HIS/A2 simulation. Their Table 2 shows that the total freshwater runoff to basin.
- 210 There is, to our knowledge, no transient scenario for the evolution of atmospheric deposition over the Mediterranean Sea. However, in order to evaluate the potential effects of aerosol deposition on the future Mediterranean Sea, we used deposition fields of total nitrogen deposition (NO<sub>3</sub> + <u>NH<sub>4</sub>) from the Mediterranean is lower than the Ludwig et al.</u> (2009) estimate (by about 30 %). They found approximately 27 % decrease in total runoff by the end of the 21st century. This trend is
- 215 consistent with the deceasing trend found by Ludwig et al. (2010). However, the 2050 estimates of freshwater runoff from Ludwig et al. (2010) are only 13 % lower than the 1970 and 2000 estimates. The freshwater runoff decrease in the physical model is more important than in the nutrient runoff model. This may result in higher nutrient concentrations at the river mouth. We are also aware that the future evolution of river discharges into the Mediterranean Sea is highly uncertain and depends at
- 220 least on the scenario choice and on the model and modelling strategy choice (Sanchez-Gomez et al.) [2009; Dubois et al.], [2012; Adloft ... However, nutrients from river discharge are consumed rapidly at proximity of the river mouth and we believe these potential higher concentrations don't have a large impact of the resultsglobal model LMDz–INCA (Hauglustaine et al.), [2014) and phosphate deposition from natural dust modeled with the regional model ALADIN–Climat (Nabat et al.), [2015a) respectively (see Richon et al.), [2017], and references therein for the description.
- 225 . The atmospheric deposition fields represent present-day aerosol deposition fluxes (1997–2012 and 1980–2012 for total nitrogen and dust deposition respectively) that are repeated over the 1980–2099 simulation period.

Initial nutrient concentrations in the Mediterranean come from the SeaDataNet database (Schaap and Lowry, 2010) and initial nutrient concentrations in the buffer zone are prescribed from the

World Ocean Atlas (WOA) (Locarnini et al.) 2006). Salinity and temperature are initialized from the MEDATLAS II climatology of Fichaut et al. (2003).
All simulations begin began from a restart of a historical run starting that started in January 1965 following a spin-up spin-up of more than 115 years done made with a loop of the period 1966 to 1981 over the 1966-1981 period for the physical forcings and the river nutrient discharge.

#### 235 2.5 Simulation set-up

All simulations are performed were made for 120 years. The control run CTRL is performed was made with forcing conditions corresponding to the 1966–1981 period looped over the simulation

period. This period was chosen in order to avoid including in the CTRL years with too important the years with excessive warming such as the 1980s and 1990s (see Figure 12 in Adloff et al. 2015) for the surface temperature evolution

- 240 . The scenario simulation is referred to as HIS/A2 as in Adloff et al. (2015). HIS is the name of the The HIS refers to the historical period (in our case between 1980 and 1999), and while A2 is the name of the 2000–2099 2000–2099 scenario simulation.
   In order to quantify separately separately quantify the effects of climate and biogeochemical changes, we performed made 2 additional control simulations: (1) CTRL\_R with climatic and Atlantic con-
- 245 ditions corresponding to present-day conditions present-day conditions (no scenario for climate change or nutrient fluxes through the Strait of Gibraltar) and river nutrient discharge following the scenario evolution, and (2) CTRL\_RG with elimatic conditions corresponding to present-day conditions, and present-day climatic conditions, but river nutrient discharge and Atlantic buffer-zone buffer-zone concentrations following the scenario conditions. Table T describes the different simula-
- 250 tions. The effects of nutrient inputs from exchanges through different effects are computed by taking differences between simulations. The effects of nutrient input from exchange across the Strait of Gibraltar and from riverine discharge independent of climate effect are derived by taking CTRL\_R minus CTRL and CTRL\_RG minus CTRL\_R. Similarly, to derive the effects of climate change and nutrient input change on nutrient budgets, we use the difference between HIS/A2 and CTRL. To de-
- rive the effects of climate change only, we calculate the difference between HIS/A2 and CTRL\_RG.

We made two supplementary simulations, one with total nitrogen deposition (HIS/A2\_N) and another with total nitrogen and natural dust deposition (HIS/A2\_NALADIN). These simulations include climate change and nutrient fluxes from rivers and via the Strait of Gibraltar that follow the scenario

260 conditions. The results from these simulations should be considered as exploratory. Nonetheless, they provide insight into the potential effects of future aerosol deposition.

#### 3 Results

#### 3.1 Evaluation of the NEMOMED8/PISCES model

NEMOMED8 has already been used in a number of regional Mediterranean Sea modeling studies either in hindcast mode (Beuvier et al.) 2010; Herrmann et al., 2010; Sevault et al., 2014; Soto-Navarro et al., 2015; Dunić et al., 2016) or scenario mode (Adloff et al., 2015). It produces the main characteristics of the Mediterranean Sea circulation. Evaluation of the HIS simulation provided in Adloff et al. (2015) shows that the main physical characteristics of the Mediterranean are produced, in spite of a too cold upper layer (1°C colder than observations) and too little stratification in com-

parison to observations. In particular, the HIS simulation matches closely the observed thermohaline circulation in the Adriatic and Ionian basins (see Adloff et al., 2015).
 The regional NEMOMED physical model has already been coupled to the biogeochemical model

PISCES on a 1/12° grid horizontal resolution (Palmieri et al.) 2015; Richon et al.) 2017, 2018), but no future climate simulation has yet been performed. As a first study coupling NEMOMED8 with

- 275 PISCES, we compared the main biogeochemical features of our simulations with available data. Figure [] shows the surface average chlorophyll concentrations in the top 10 meters of the CTRL and HIS/A2 simulations, and from satellite estimations from MyOcean Dataset (http://marine.copernicus.eu). All chlorophyllvalues in the article and the data are chlorophyll\_ahttp://marine.copernicus.eu/services-portfolio/access-to-products/?option=com\_csw&view=details&product\_id=OCEANCOLOUR\_MED\_CHL\_
- 280 L4\_NRT\_OBSERVATIONS\_009\_041). Whenever we refer to chlorophyll, we always mean chlorophyll–a (hereafter noted chl-a). The model correctly reproduces the main high–chlorophyll\_high–chl-a regions such as the Gulf of Lions-Lion and coastal areas. However, Figure I shows an underestimation of about 50 % of the surface chlorophyll\_chl-a concentrations by the model in these productive areas. The west–to–east gradient of productivity is also reproduced by the model with values that agree with
- 285 satellite estimates (approximately 50 % decrease in average chl-a concentration between the western and the eastern basin in the satellite data and 30 to 50 % in the model). Moreover, this Figure shows that ehlorophyll-chl-a produced by the CTRL is stable over time. The model fails, however, to reproduce the observed ehlorophyll-rich chl-a-rich areas in the Gulf of Gabes and at the mouth of the Nile. This discrepancy is probably linked with insufficient simulated nutrient discharge from coastal
- runoff in these regions. Moreover, several studies (see e.g. Claustre et al., 2002; Morel and Gentili, 2009) show that satellite estimates have a systematic positive bias in the coastal regions because of the high concentrations of colored dissolved presence of particulate matter (for instance, sediments). The general bias observed in the Mediterranean is linked with organic matter and the presence of dust particles in seawater back scattering lightwhich cause light back scattering. Figure 2 provides
- an evaluation of the average <u>chlorophyll\_normalized chl-a</u> surface concentration evolution over the entire basin for the period 1997–2005. This Figure shows that the normalized <u>chlorophyll\_chl-a</u> surface concentration in the model is close to the estimates provided by the SeaWiFs satellite data (Bosc et al., 2004). Even though the interannual variability of the model is 50 % smaller than in the satellite product, the model captures the increase in <u>chlorophyll\_chl-a</u> concentration between 2002 and 2005
- 300 (approximately 15 % of increase in the model and 30 % in the satellite data). The evaluation of the model against 2-independent two datasets shows that the model yields satisfying estimates of surface chlorophyllchl-a.

The vertical distribution of nitrate and phosphate over a section crossing the Mediterranean from East to West as well as <u>chlorophyll-chl-a</u> and nutrient concentration profiles at the DYFAMED station are

305 shown in appendix Appendix (Figures A1 and A3). These figures show that the model produces some seasonal and interannual interannual variability of the nutricline depth and intensity. However, the nutricline depth and DCM depth are consistently overestimated by the model in comparison to the data. The nutricline intensities intensity seem to be underestimated by about 50 % and the its depth is overestimated. However, nutricline depth deepens from 100–120 m to 180–200 m between

- 310 the western and the eastern basins (see Figure A3). The average chl-a concentration observed at the DYFAMED station in the top 200 m is 233 ± 146 ng L<sup>-1</sup> (average over the 1991–2005 period), while the model value for the HIS/A2 simulation over the same period is 159 ± 87 ng L<sup>-1</sup> (Figure A1). In spite of some underestimation of nutrient concentrations that are probably linked with the features of the simulated intermediate and deep waters characteristics, the PISCES model reproduces
- 315 the main characteristics of the Mediterranean biogeochemistry, including a salient west-to-east gradient in nutrient concentrations, low surface nutrient concentrations and a deep ehlorophyll chl-a maximum (DCM). The average chlorophyll concentration observed at the DYFAMED station in the top 200 m is 233 ± 146 10<sup>-9</sup>g L<sup>-1</sup> (average over the 1991–2005 period), while the model value for the HIS/A2 simulation over the same period is 159 ± 87 10<sup>-9</sup>g L<sup>-1</sup>. These performances lend
- 320 credence to our efforts to investigate the evolution of the Mediterranean biogeochemistry under the A2 climate change scenario with the same modeling platform.

#### 3.2 Evolution of temperature and salinity

Average surface temperature and salinity (SST and SSS) evolution in the entire basin during the CTRL and HIS/A2 simulations are shown in Figure 3, which confirms results from Adloff et al.

- 325 (2015) and shows that the CTRL simulation is stable over time. Beyond this basin-wide average variation in SST and SSS, a more detailed analysis reveals much greater variability depending on the region (Somot et al., 2006; Adloff et al., 2015). For instance, the Balearic Sea is more sensitive to warming than the rest of the western basin, and the eastern basin has a more intense warming than the western basin (up to 3°C warming in the eastern basin and in the Balearic Sea). Also, the surface
- 330 salinity in the Aegean Sea increases more than the other regions.

#### 3.3 Evolution of the nutrient budgets in the Mediterranean Sea

The nutrient budgets of the semi-enclosed Mediterranean basin are highly dependent on external sources (e.g. Ludwig et al., 2009, 2010; Huertas et al., 2012; Christodoulaki et al., 2013). In-We

335 first looked at the evolution of phosphate and nitrate fluxes in and out of the Mediterranean during our simulations. Then, in order to map the effects of climate change and external nutrient flux evolution on the Mediterranean nutrient balance, we calculated mass budgets of inorganic nitrate and phosphate during the simulated period. These budgets take into account Finally we calculated the evolution of nutrient concentrations in different layers of the Mediterranean in order to point

out the different effects of climate and nutrient fluxes on surface, intermediate and deep waters. The nutrient budgets account for changes in Atlantic input, river discharge and sedimentation. Nitrate can also accumulate in the Mediterranean waters through N<sub>2</sub> fixation by cyanobacteria, but this process accounts for less than 1 % of the total nitrate budget (Ibello et al., 2010; Bonnet et al., 2011; Yogev et al., 2011), and is neglected here.

- In this Section, we refer to the period 1980-1999 as the beginning of the century, to the period 2030–2049 as the middle of the century and to the period 2080–2099 as the end of the century. Also, we derive the effects of river input changes as the difference of nutrient concentrations between CTRL\_R and CTRL over these time periods. Similarly, the effects of changes in nutrient fluxes through the Strait of Gibraltar are derived by the differences between CTRL\_RG and CTRL\_R, and
   the effects of climate change by the difference between HIS/A2 and CTRL\_RG.

## 3.3.1 Phosphate and nitrate budgets under climate and biogeochemical changes in the Mediterranean

Tables 2 and 3 summarize the average phosphate and nitrate content in all simulations for the 3 time periods described earlier. Phosphate content in the entire Mediterranean has increased in our
simulation by 6 % over the 21st century, as determined by the difference between CTRL and HIS/A2 simulations between 1980-1999 and 2080-2099. The increase is more important in the eastern basin than in the western basin. In particular, we observe an 8 % increase in phosphate content in the Ionian–Levantine sub–basin in 2080–2099 compared to 1980–1999. The effects of phosphate river input changes are substantial over the first half of the century. We observe 3 % decrease in

- 360 phosphate content in the entire Mediterranean between 1980–1999 and 2030–2049 due to river input changes (difference between CTRL\_R and CTRL). Changes in phosphate fluxes thought the Strait of Gibraltar seem to have limited effect on the global Mediterranean phosphate content. However, elimate change effects lead to a global enhancement of 10 % in phosphate content in 2080–2099 in comparison to 1980–1999. This result shows contrasted effects of physical and biogeochemical
- 365 conditions on the evolution of nutrient concentrations. Table 3 shows that in the model, the combined effects of climate, riverine and Atlantic nutrients input changes over the 21st century lead to a 17 % increase in nitrate content over the Mediterranean in the 2080–2099 period compared to 1980–1999 (derived from the calculation of HIS/A2–CTRL). Changes in river discharge in the Mediterranean over the century lead to 9 % enhancement of nitrate content by the end of the century (2080–2099)
- 370 compared to the beginning of the simulation period (1980–1999). The most important effects of river input changes are observed in the Adriatic basin (over 50 % nitrate accumulation by the end of the century). Over the entire Mediterranean, the effects of changes in the fluxes though the Strait of Gibraltar on nitrate content are weak (< 1 %). However, the comparison of nitrate content in CTRL\_RG in the western basin shows a 3 % decrease in nitrate content in the western</p>
- 375 basin during the 2030–2049 period followed by an increase resulting in +1 % nitrate by 2080–2099 compared to the 1980–1999 period. Finally, climate change effects lead to 7 % increase in nitrate content over the Mediterranean basin in the 2080–2099 period compared to 1980–1999 (computed by HIS/A2–CTRL\_RG). These results indicate that river inputs and climate change are the main drivers of nitrate content changes in the Mediterranean basin over the 21st century.

#### 380 3.3.1 Fluxes of nutrients through the Strait of Gibraltar

The Mediterranean is connected to the global ocean by the narrow Strait of Gibraltar. Water masses transport through this strait contributes contribute substantially to its water and nutrient budgets (e.g. Gómez, 2003; Huertas et al., 2012). The Mediterranean is a remineralization basin that has net negative fluxes of inorganic nutrients (i.e. organic nutrients enter the basin through the Gibral-

- 385 tar Strait surface waters and inorganic nutrient leave the Mediterranean through the deep waters of the Gibraltar Strait Huertas et al., 2012). Figure 4 shows the evolution of incoming and outgoing nitrate and phosphate fluxes through the Strait of Gibraltar in the HIS/A2 and in the CTRL simulations. We observe similar trends in phosphate and nitrate fluxes in the model. This commonality is linked to the Redfieldian behavior of the primary production in PISCES. According to the The
- 390 nutrient fluxes through the Strait of Gibraltar result from both the evolution of water fluxes, from NEMOMED8, and the evolution of nutrient concentrations in the buffer zone, from the A2 scenario from Dufresne et al. (2013).

In the HIS/A2 simulation, the incoming fluxes of nitrate and phosphate decrease slightly (flux of nitrate decreases from 50 the to 35 Gmol month<sup>-1</sup> for nitrate and while that for phosphate drops

- 395 from 2.5 to 1.55–1.6 Gmol month<sup>-1</sup> for phosphate) until the middle of the century (with despite a period of increased incoming fluxes of both phosphate and nitrate these nutrients in the 1990s) and then. After 2050, fluxes increase to reach values higher than the control in the last 25 years of simulations (Figure 4). Outgoing fluxes follow the same trends as incoming fluxes: total outgoing nitrate and phosphate fluxes decrease from 1980 to 2040 (flux values getting closer to zero) and
- 400 then increase until the end of the century. We observe a decreasing trend in the nitrate outgoing flux in the control (from -129 to -110 Gmol month<sup>-1</sup> representing about 18 %). At the end of the 21st century, incoming fluxes of nutrients By 2100, incoming nutrient fluxes have increased in the A2 scenario simulation by about 13 -% (difference between the 2080-2099 and 1980-1999 % (2080-2099 minus 1980-1999 periods). But this significant increase (linear regression reveals This increase is
- 405 statistically significant, with a linear regression having a positive slope with correlation coefficient greater than 0.75 and a p-value < 0.001for the second half of the simulation period for both nitrate and phosphate). Furthermore, this increase follows a decrease of over 20 % in incoming nutrient fluxes between the 1980–1999 and the 2030–2049 periods. Most of the decrease is observed between 2030 and 2040 (with a decrease of 15 and 1 Gmol month<sup>-1</sup> for nitrate and phosphate respectively
- 410 during this decade). Outgoing fluxes increase less between the beginning and. Outgoing fluxes through the Strait of Gibraltar follow the same trends as incoming fluxes: total outgoing nitrate and phosphate fluxes decrease from 1980 to 2040 with flux values getting closer to zero and then increase until the end of the century(3.5 and 3.9 % for phosphate and nitrate respectively,. We observe a decreasing trend in the nitrate outgoing flux in the control from -129 in to
- 415 -110 Gmol month<sup>-1</sup> representing about 15 % flux decrease. Over the simulation, outgoing nutrient fluxes increase less than incoming nutrient fluxes. The increase in outgoing nitrate and phosphate

flux is less than 5 % (Figure 4). If the The relative changes in incoming and outgoing fluxes seem to indicate an increase in the net incoming flux , the absolute values seem to show a rather steady net flux between the beginning and the end of the century. Net of about 5 %. The net flux at the beginning

- 420 of the century is around -83 Gmol month<sup>-1</sup> for nitrate and -3 Gmol month<sup>-1</sup> for phosphate. At the end of the century, the fluxes are about -80 Gmol month<sup>-1</sup> and -2.5 Gmol month<sup>-1</sup> for nitrate and phosphate respectively. Also, these net fluxes are close to the CTRL net fluxes. These trends result from the evolution of water fluxes through the Strait of Gibraltar computed by NEMOMED8 and the A2 scenario of nutrient concentrations in the buffer zone taken from Dufresne et al. (2013). The
- 425 imbalance between incoming and outgoing nutrient flux anomalies may be a cause for the observed accumulation of inorganic nutrients (in particular of nitrate) in the basin.

#### 3.3.2 River fluxes of nutrients

River discharge is the main external source of phosphate for the eastern part of the basin (Krom et al., 2004; Christodoulaki et al., 2013). Figure 5 shows the total discharge of phosphate and nitrate from rivers to the Mediterranean Sea.

Phosphate discharge decreases by 25 % between the beginning and the end of the simulation period. As suggested by Ludwig et al. (2010), phosphate discharge in the A2 period stays lower than in the HIS period, in spite of a small discharge enhancement between 2030 and 2049.

- Nitrate riverine discharge in the HIS/A2 simulation is significantly substantially higher than in CTRL (between the CTRL simulation by 30 and to 60 Gmol month<sup>-1</sup>difference). Nitrate total discharge in the Mediterranean has continuously increased. The total river discharge of nitrate into the Mediterranean Sea has increased continuously from the 1960s (see the CTRL values for the years 1966–1981). According to the HIS/A2 simulation, total river nitrate discharge is
- 440 24 % larger during 2080-2099 than during 1980-1999.

#### 3.3.3 Sedimentation

445

Sedimentation removes nutrients from the Mediterranean Sea. In this version of PISCES, the loss of nitrogen and phosphorus to the sediment is calculated from the sinking of organic carbon particles particulate organic carbon (POC) to the sediment (linked through the Redfield ratio). Sediment fluxes of phosphorus and nitrogen during the simulations are shown in Figure [6].

The nutrient loss to the sediment decreases rapidly during the HIS simulation (<del>1980–1999)</del> 1980–1999) and remains low during the 21st century although it exhibits substantial interannual variability in the sedimentation fluxes. By the end of the 21st century, sedimentation of <del>P and N phosphorus and</del>

450 <u>nitrogen</u> are almost 50 % lower relative to the 1980 fluxes.

- 3.3.4 Evolution of phosphate Phosphate and nitrate concentrationsIn order to observe budgets under climate and biogeochemical changes in the general evolution of nutrient concentrations over the Mediterranean
- 455 Tables 2 and 3 summarize the average phosphate and nitrate water column content in all simulations for the 3 time periods described earlier.

Total phosphate content in the entire Mediterranean grew in our HIS/A2 simulation by 6 % over the 21st century, we plotted the evolution of phosphate and nitrate concentrations for the entire
simulation period century, as determined by the difference between CTRL and HIS/A2 simulations between 1980-1999 and 2080–2099. The increase is larger in the eastern basin than in the western basin. In particular, there is an 8 % increase in phosphate content in the Ionian–Levantine sub–basin. Nutrient content in the HIS/A2 simulation is affected by both climate and nutrient fluxes from external sources (rivers and fluxes via the Strait of Gibraltar). The effects of changes in riverine

- 465 input of phosphate are derived from the CTRL\_R simulation (see also Figure ). The difference of phosphate content between CTRL and CTRL\_R are substantial over the first half of the century. We observe 3 % decrease in phosphate content in the entire Mediterranean between 1980–1999 and 2030–2049 due to river input changes (difference between CTRL\_R and CTRL). Changes in phosphate fluxes thought the Strait of Gibraltar seem to have limited effect on the global Mediterranean
- 470 phosphate content as revealed by the small difference between the beginning and the end of simulation CTRL\_RG. Conversely, climate change enhances the basin-wide phosphate content by 10 % in 2080–2099 relative to 1980–1999 (HIS/A2 minus CTRL\_RG). Thus future changes in climate and external nutrient supply may have opposite effects on nutrient concentrations in the Mediterranean.
- 475 Table shows that by 2100, in HIS/A2 the combined effects of climate change, riverine, and Atlantic nutrient input changes over the 21st century lead to a 17 % basin-wide increase in nitrate content when compared to CTRL. Changes in river discharge lead to 9 % larger nitrate content by the end of the century (2080–2099) compared to the beginning of the simulation period (1980–1999). The most important effects of river input changes are observed in the Adriatic basin (over 50 % nitrate
- 480 accumulation by the end of the century). The largest effects from river input changes are found in the Adriatic basin (>50 % nitrate accumulation by the end of the century). Across the Mediterranean, there is only a weak (<1 %) effect on nitrate content from changes in the fluxes through the Strait of Gibraltar. In the western basin, the comparison of CTRL\_R and CTRL\_RG reveals a 3 % decline in nitrate content during the 2030–2049 period followed by an increase, reaching +1 % in 2080–2099</p>
- 485 relative to 1980–1999. Finally, climate change enhances basin-wide nitrate content by 7 % between 1980–1999 and 2080–2099 period (HIS/A2 minus CTRL\_RG). Thus river inputs and climate change are the main causes for changes in basin-wide nitrate content during the 21st century. These global nutrient budgets reveal that climate change and external nutrient fluxes to the Mediterranean

can influence its nutrient content in different sometimes even in opposing directions. In particular,

490 river inputs have large effects on nutrient content in the eastern basin, while input through the Strait of Gibraltar has limited effects on the nutrient content even in the western basin.

#### **3.3.5** Continuous evolution of phosphate and nitrate concentrations

In order to observe the continuous evolution of nutrient concentrations in different layers over the 21st century, we plotted the evolution of phosphate and nitrate concentrations for the entire simulation period in the western and eastern basins. The separation between western and eastern basin is the Sicily Strait . Therefore, the eastern basinincludes the Ionian, Levantine, Adriatic and Aegean basins. Figure 2 shows the evolution of average phosphate concentrations in the western and eastern basin, respectively, for basins in the surface (0–200 m), intermediate (200–600 m) and bottom deep (> 600 m) layers of the water column. (Figures 2 and 3). With the separation between western and eastern basin being the Sicily Strait, the eastern basin includes the Ionian, Levantine,

500 western and eastern basin being the Sicily Strait, the eastern basin includes the Ionian, Levantine, Adriatic and Aegean basins.

#### Phosphate

495

Figures 77a and 47b show that the evolution of surface phosphate concentration in the western basin is linked with phosphate inputs through the Strait of Gibraltar (Pearson's correlation coefficient is

- 505 0.85, p-value < 1 %). We observe that As shown in Figure 7a and 7b until mid century, the phosphate concentration in HIS/A2 decreases in the surface and intermediate layers of the western basin until the middle of the century (concentration decreases by about 0.015 mmol m<sup>-3</sup> in the surface layer and by 0.017 mmol m<sup>-3</sup> in the intermediate layer ) and then of the western basin. After 2050, phosphate concentration increases again until
- 510 the end of the centuryto reach similar concentration than, reaching concentrations that are similar to those in 1980 in the surface layer and higher concentrations than in 1980 in the intermediate layer (but at intermediate depths reaching values that are about 0.01 mmol m<sup>-3</sup> higher ). This evolution is different than the controls, than in that reference year. Figures [7a] and [4b] show that the evolution of surface phosphate concentration in the western basin in CTRL\_RG and HIS/A2
- 515 are similar. The Pearson correlation coefficient between western basin phosphate concentration in HIS/A2 and incoming phosphate fluxes through the Strait of Gibraltar is 0.85, p-value < 0.01. Thus the phosphate concentration in the western basin appears to be influenced by phosphate inputs through the Strait of Gibraltar. However, Figure 7a shows that the difference in surface phosphate concentrations in the surface layer of the western Mediterranean in the CTRL\_RG and CTRL\_R</p>
- 520 simulations is important only at the end of the 21st century (approximately from 2070). Therefore, we hypothesize that the similar evolutions differ substantially only after about 2070. Thus the similar evolution of phosphate concentration in HIS/A2 and of incoming fluxes of phosphate the incoming phosphate fluxes through the Strait of Gibraltar throughout the simulation period are linked with

may be linked to changes in physical conditions. In this very dynamic part of the Mediterranean

525 <u>Mediterranean region</u>, changes in physical conditions linked with climate change are preconditioning precondition the western basin to become more sensitive to nutrient fluxes thought the Strait of Gibraltar.

A slight accumulation of about 0.015 mmol m<sup>-3</sup> of phosphate is <del>observed simulated</del> in the HIS/A2 simulation in the deep western basin. The <del>significant large</del> difference between the HIS/A2 simulation

- 530 and the control runs (over 0.010 mmol m<sup>-3</sup> by the end of the century) shows that the evolution of the Mediterranean physics linked with climate change is primarily responsible for the changes in phosphate concentration in the intermediate and deep western basin. Climate change effects lead to an accumulation of phosphate in the intermediate and deep layer that is probably linked to a decrease in surface primary productivity (hence, in nutrient consumption and export), decreased also decreases
- sediment fluxes (see Figure 6), and increased and increases stratification, thus isolating most of the phosphate pool from the surface.

Nutrient Phosphate concentrations in the eastern part of the basin are lower than in the western part<del>(</del>. Figure [] illustrates the 50 % lower phosphate concentration in the surface layer, about the roughly 30 % lower concentration in the intermediate layer and about the 15 to 20 % lower concentration

- 540 in the deep layer). In the surface layer, phosphate concentration decreases decline in the beginning of the simulation and remains low during the 21st century (from 0.022 mmol m<sup>-3</sup> in 1980 to less that 0.015 mmol m<sup>-3</sup> in 2000 , and remains low during the 21st century (Figure 7d). There is, however, a large annual interannual variability in surface phosphate concentration with peaks up to 0.025 mmol m<sup>-3</sup> in 2060. But the HIS/A2 simulation values are consistently below the CTRL
- 545 concentrations showing an important effect of climate change on surface phosphate reduction. We observe in Figures 7e and 7f an accumulation of phosphate in the intermediate and deep layers (of 17 and 13 % respectively), with large decennial variability of phosphate concentration in the deep eastern basin. In both of these layers, HIS/A2 concentrations are higher than the CTRL concentrations = (over 0.015 mmol m<sup>-3</sup> higher by the end of the century). These results show that the evolution of
- 550 phosphate concentrations in the eastern Mediterranean throughout the 21st century is mainly driven by climate change. Indeed, Figure Ashws that average PO<sub>4</sub> concentrations in CTRL, CTRL\_R and CTRL\_RG are similar for all periods in the eastern basin.

The evolution of nitrate concentration shows an accumulation over the century in all regions of the intermediate and deep Mediterranean waters (between 9 and 20 %)in the HIS/A2 simulation

555 (Figures 8b, 8c, 8e and 8f).

#### Nitrate

In the surface western basin, the evolutions of nitrate evolutions in the HIS/A2 and CTRL\_RG simulations are similar, showing confirming the regulating effects of fluxes through the Strait of Gibraltar (Figure 8d). We observe an accumulation of nitrate in the HIS/A2 simulation in the intermediate
- 560 and deep Mediterranean waters between 9 and 20 % (Figures 8b, 8c, 8c and 8f). In the intermediate layer, nitrate concentration in the HIS/A2 simulation is decreasing from 4.05 decreases from 4.1 to 3.6 mmol m<sup>-3</sup> between 1980 and 2030. After 2030, the concentration it increases again up to 4.3 mmol m<sup>-3</sup>. In the deep western basin, we observe a slight decrease in nitrate concentration in the controls from 4.4 to about 4.15 4.2 mmol m<sup>-3</sup> whereas. In HIS/A2, nitrate concentration follows
- the decrease of the controls until approximately 2020, then, there is a slight accumulation from 4.5 to 4.75-4.8 mmol m<sup>-3</sup> in the until the end of the century. Thus, physical changes linked with climate change have little effect on nitrate concentrations in the deep western Mediterranean until approximately 2020. Significant differences between HIS/A2 over the simulation period and CTRL appear in the middle of the 21st century.
- 570 In the eastern basin, the impacts of river discharges of nitrate seem to have large influence on the nitrate accumulation as shown by the similar evolution of HIS/A2 and CTRL\_R simulations (Figure & general &
- 575 on surface nitrate concentrations in the eastern basin and is responsible for an important part of the eastern Mediterranean nitrate budget (see also Table 3). But the strong difference between CTRL\_R and HIS/A2 at the end of the century indicates that vertical stratification leads to a decrease in surface layer nitrate concentrations, probably linked both with lower winter mixing and nutrient consumption by phytoplankton. In the intermediate and deep layers, the evolution of physical condi-
- 580 tions seems to have similarly large impacts has a similarly large impact on the nitrate concentrations in the eastern basin as shown by the difference between CTRL\_R and HIS/A2 (see also Table 3). In particular, nitrate concentrations increase by about 0.5 mmol m<sup>-3</sup> between 1980 and 2099 in the deep eastern basin. Approximately 50 % of this accumulation is due to river discharge. The large differences between the CTRL simulations and the HIS/A2 show that modification of circulation
- 585 resulting from climate change have substantial impacts on the deep and intermediate nutrient concentrations. Figure & shows the contrasted effects of climate and biogeochemical changes. The strong difference between CTRL\_R and CTRL concentrations at the beginning of the simulation (almost 0.4 mmol m<sup>-3</sup>) indicates that riverine nutrient discharge has a strong influence on surface nitrate-

590

Evaluating separately the evolution of nutrient concentrations in different layer of the Mediterranean Sea shows that external nutrient fluxes primarily affect the surface in the western basin whereas climate change affects the entire water column. Also, climate and nutrient fluxes may have opposite effects on surface nutrient concentration. This leads to different trends in nutrient concentrations in

595 the eastern basin and is responsible for an important part of the eastern Mediterranean nitrate budget (see also Table 3). But the strong difference between CTRL\_R and HISsurface layer and in the intermediate and deep layers. In particular, surface nitrate in the eastern basin is observed to increase as a result of increased river discharge, but climate change effects lower concentrations in HIS/A2 at the end of the century indicates that vertical stratification leads to a decrease in surface layer

600 nitrate concentrations, probably linked both with lower winter mixing and nutrient consumption by phytoplankton. A2 (see figure 8d). On the other hand, climate and river discharge of nitrate have similar effects on the intermediate and deep eastern layers, leading to the simulated increase in nutrient content (Tables 2 and 3).

# 3.4 Present and future surface Surface nutrient concentrations, primary productivity and nutrient limitationsin the surface Mediterranean

605

Figures and 10 show the average surface concentrations of nitrate and phosphate in the beginning of the century (1980–1999) and the relative concentration differences with for the end of the century in the HIS/A2 and CTRL simulations. In the Mediterranean Sea, primary productivity is mainly limited by these 2 nutrients and their evolution in the future may impact the productivity of the basin.

- Figure confirms the previous results showing an accumulation of nitrate in large zones of the basin by the end of the century, except for the southwestern part of the western basin (Alboran Sea)and a small area, the Gulf of Lion, the North Levantine around the Rhodes Gyre and Crete and small areas in the southeastern Levantine, Tyrrhenian and Algerian basins.
  On the contrary, Figure 10 shows that phosphate surface the surface phosphate concentration is
- 615 decreasing everywhere in the over most of the Mediterranean basin except near the mouth of the Nileand, the Ionian, Algerian, Tyrrhenian, between Crete and Cyprus and in the Alboran Sea. The specific concentrations observed next to the Nile mouth are linked with to an inversion of the N:P ratio in this river in our scenario (i.e. an increase in P discharge and a decrease in N discharge)in this river in our scenario. The distribution of surface phosphate concentration at the
- 620 end of the century (2080–2099) shows that all P–rich areas of the eastern basin at the beginning of our simulations are depleted by the end of the simulation. For instance, the <u>P-rich P-rich</u> areas around Crete and Cyprus are no longer observed in the 2080–2099 no longer exist in the 2080–2099 period (Figure 10). Moreover, Figure 11 shows that these areas match productive zones of high productivity. All the most productive zones of the beginning of the century are
- 625 reduced in size and intensity by the end of the century. The primary production integrated For instance, there is a 10 to 40 % decrease in primary production in the Gulf of Lion and around the Balearic Islands, more than 50 % reduction in the North Adriatic basin, in the Aegean Sea and in the eastern Levantine basin around Cyprus. There is also a reduction in primary productivity from 40-50 gC m<sup>-2</sup> year<sup>-1</sup> to 20-30 gC m<sup>-2</sup> year<sup>-1</sup> around Cyprus. These mesoscale changes
- 630 may be linked with changes in local circulation (e.g., mesoscale eddies). These observations show that the evolution of the Mediterranean biogeochemistry is influenced by both meso- and large-scale circulations patterns. The simulated basin-wide vertical integral of primary production over the eu-

photic layer (0–2000–200 m) is reduced in our simulation\_declines by 10 % on average between 1980–1999 and 2080–2099. However, Figure III shows 1980–1999 and 2080–2099. However, there

- 635 is a productivity decrease of more than 50 % in areas such as the Aegean Sea and the Levantine Sea (Figure 1). In general, the differences in surface biogeochemistry between the 1980–1999 and 2080–2099 periods biogeochemical variables between 1980–1999 and 2080–2099 are weaker in the western basin because of the strong regulating impact of nutrient exchange through the Strait of Gibraltar. The large scale reduction of surface primary productivity may be a cause for the observed
- 640 reduction in sedimentation (see Figure 6).

665

We also observe local changes in nutrient concentrations and primary production. For instance, around Cyprus, changes in local concentrations of nutrients (decrease of about 50 % in phosphate concentration) have substantial effects on primary productivity (decrease from 40-50 gC m<sup>-2</sup> year<sup>-1</sup>)

to 20-30 gC m<sup>-2</sup> year<sup>-1</sup>). These mesoscale changes may be linked with local circulation changes (such as mesoscale eddies). These observations show that the evolution of the Mediterranean biogeochemistry is influenced by both meso and large scale circulations patterns.

Figure 12 presents the limiting nutrient calculated using PISCES half-saturation coefficients (see Aumont and Bopp, 2006). The limiting nutrient is derived from the minimal value of limitation fac-

- 650 tors. In the Monod-type model PISCES, nutrient-based growth rates follow a Michaëlis-Menten evolution with nutrient concentrations. In the present period, most of the productive areas are N and P colimited in the simulation (Figure 12). This includes regions such as the Gulf of LionsLion, the South Adriatic Sea, the Aegean Sea and the northern Levantine basin. Future accumulation of nitrogen in the basin modifies the nutrient balance causing most eastern Mediterranean surface wa-
- 655 ters to become P-limited. The total balance of phosphate is more negative in the future than in the present period whereas we observe an inverse situation for nitrate. Therefore, phosphate would become becomes the major limiting nutrient in most of the regions where productivity is reduced such as the Aegean Sea, the northern Levantine basin and the South Adriatienorth eastern Ionian Sea.

## 660 3.5 Modifications of the Mediterranean deep chlorophyll maximum and chl-a budget

One specificity of Mediterranean biology is that most planktonic productivity occurs below the surface at a depth called the deep chlorophyll maximum (DCM). Hence, most of the chlorophyll concentration is not visible by satellites (Moutin et al., 2012). Figure [3] shows the average depth of the simulated DCM-deep chlorophyll maximum (DCM) for the period 1980–1999 and for the period 2080–2099.

We observe that the The DCM depth changes little during the simulation, even though the physical characteristics of the water masses do change (salinity and temperature )change. The DCM tends

to deepen deepens slightly in some regions such as the South North Ionian and the Tyrrhenian

670 basin. These results suggest that South of Crete. Although the DCM depth is not significantly altered changes little in the futurebut, the intensity of subsurface productivity seems is reduced (see Figure III).

Figure <sup>14</sup>/<sub>4</sub> shows the average vertical profiles of <del>chlorophyll chl-a</del> at the DYFAMED station (43.25° N,
7.52° E) and the average profiles for the western and eastern basins for the 1980–1999 and 2080–2099 periods. The results show that the subsurface chlorophyll maximum is still modeled at subsurface chl-a maximum persists through to the end of the century. At the DYFAMED station, the average DCM depth is unchangedbut surface concentration is enhanced remains unchanged, while the surface chl-a concentration is decreased by about 25 10<sup>-9</sup> g ng L<sup>-1</sup>, which is negligible. Thus

- 680 the average chl-a profile at DYFAMED changes little throughout the simulation. This shows that local variability in the Mediterranean circulation and biogeochemistry is important. However, we simulated seasonal at that station there is approximately 40 % variability in the chlorophyll concentration profiles at the station with intensity chl-a concentration profile and depth of DCM reduced by about 40 % for some month (not shown). In the western basin, the subsurface maximum in the
- 685 present and future periods is maxima at present and in the future are located at the same depth (100-120100-120 m), but the average chlorophyll concentration is reduced by almost 50 %, which confirms the results from chl-a concentration in the DCM increases by about 15 to 20 ng L<sup>-1</sup> during the simulation. However, where there are small changes in the average chl-a profiles in the western basin, those are often accompanied by greater local changes in the depth and intensity of the DCM
- 690 (Figure 13). In the eastern basin, subsurface <u>chlorophyll chl-a</u> concentration is reduced by about  $50 \text{ ng L}^{-1}$  and the subsurface <u>chlorophyll chl-a</u> maximum deepens from 100–120 m to below 150 m.

In the oligotrophic Mediterranean, the majority of the <del>chlorophyll chl-a</del> is produced within the DCM. The changes circulation. There is an 8.9 % reduction in integrated chl-a production between the

- 695 <u>1980–1999 and 2080–2099 due to circulation changes</u> combined with changes in fluxes through the Strait of Gibraltar and riverine inputs<del>result in 17 % reduction in integrated chlorophyll production between the 1980–1999 and 2080–2099 periods</del>. Table 4 reports total <del>chlorophyll production chl-a</del> <u>budgets</u> in the 1980–1999, 2030–2049 and 2080–2099 periods of all the simulations in all Mediterranean subbasins (Figure 2 from Adloff et al., 2015). Table 4 shows that chlorophyll production
- 700 It reveals that chl-a budget is stable over the CTRL simulation but decreases in all Mediterranean subbasins over the HIS/A2 simulation. The decrease in chlorophyll production is more important chl-a is larger in the eastern regions, in particular in the Adriatic and Aegean Seas (-29 and -15-17 and -19 % respectively). In the western basin, the loss of chlorophyll decline in chl-a is smaller (-13-5.1 %). The chlorophyll production chl-a budget is probably maintained by the enhancement of
- 705 <u>enhanced</u> nutrient fluxes through the Strait of Gibraltar (chlorophyll production chl-a in CTRL\_RG

does not significantly decrease in the western basin).

The results indicate that About 85 % of the reduction in chlorophyll production in the future period modeled in the future reduction in chl-a in HIS/A2 simulation is explained by elimate effects (difference

- 710 between the effects of climate change (HIS/A2 and minus CTRL\_RG). However, the effects of from increased nutrient inputs through the Strait of Gibraltar, decreased riverine phosphate inputs and increased riverine inputs of nitrate seem to have opposing effects to climate and circulation changes on chlorophyll-chl-a production. In particular, in the western basin, changes reductions in riverine discharge of nutrients seem to reduce chlorophyll production reduce chl-a by 3.5 % (see CTRL\_R
- 715 values), whereas changes in fluxes through the Strait of Gibraltar seem to enhance chlorophyll production (see enhance chl-a (only 1 % decrease in chlorophyll concentration in CTRL\_RG in the western basin).

## 3.6 Plankton biomass evolution

- Most of the biological activity in the marine environment is confined to found within the euphotic layer which is confined to the upper 200 m. Figure 15 shows the evolution of nanophytoplankton and diatom concentrations in (in terms of carbon content mmolC m<sup>-3</sup>) over the top 200 m for the entire simulation period in all simulations in in the western and eastern basins throughout the duration of all simulations. In HIS/A2, we observe lower biomass for the biomass of both phytoplankton classes across the Mediterranean Sea at the end of the century than at the beginning. In
- 725 general, diatoms seem declines during the simulation (-0.01 mmolC m<sup>-3</sup> for nanophytoplankton and -0.03 to -0.04 mmolC m<sup>-3</sup> for diatoms). Generally, diatoms appear more sensitive to climate change and as their biomass decreases more sharply than nanophytoplankton. Moreover, does that for nanophytoplankton. As shown in Figure [15c]shows that-, diatom concentrations in the western basin seem to be appear sensitive to changes in nutrient input across the Strait of Gibraltar as indi-
- 730 cated by the large difference between CTRL\_RG and CTRL\_R (0.04 mmolC m<sup>-3</sup>). However, the low diatom concentration observed in reduction in diatom concentration found with HIS/A2 indicates that the evolution of the diatom concentration over the 21st century-it is primarily influenced by climatic drivers.
- Figure 6 shows the evolution of zooplankton biomass in the top 200 m during the simulation. The same general evolution is found than for the simulated phytoplankton : for zooplankton as seen for phytoplankton (Figure 6). In HIS/A2 in all basins there is a decrease in microzooplankton during 1980–2000 concentration during 1980–2000 (from 0.165 to approximately 0.114 mmol m<sup>-3</sup>), after which it remains stable and consistently below the CTRL values until the end of the simulation
- 740 in 2100 in all basins. A large drawdown 2100. In the eastern basin, there is a large reduction in mesozooplankton levelsis simulated in the eastern basin. The average mesozooplankton concentra-

tion in the eastern part of the Mediterranean declines by almost 60 % in 2099 in comparison to that in 1980. However, the average mesozooplankton concentration over the 2080–2099 period is only slightly lower than the average concentration over the 1980–1999 period (0.10 and 0.11 mmol m<sup>-3</sup>)

- 745 respectively) because the decline in concentration occurs within the first years of the HIS/A2 simulation. In the western basin, we observe a marked decrease of there is a marked decline in mesozoo-plankton concentrations between 1980 and 2040 (a decrease of 0.05 mmol m<sup>-3</sup> is observed between these 2 periods). After 2040, mesozooplankton surface concentration the surface concentration of mesozooplankton increases regularly to similar values than values that are similar to those at the
- 750 beginning of the simulation. This evolution is similar to those of nutrient concentrations in surface waters of the western basin (Figure 7). In the PISCES model, zooplankton , and in particular mesozooplankton is highly and particularly mesozooplankton are especially sensitive to the variations of external climatic and biogeochemical conditionsbecause that is, being the highest trophic level that is represented. Owing to their bottom–up control, zooplankton canalize all changes at the
- 755 basin scale and ultimately displays the largest response. This behavior is similar to the trophic amplification observed by Chust et al. (2014) and Lefort et al. (2015).

Altogether, the analysis of plankton biomass evolution during the simulation period suggests that primary and secondary production in the eastern basin are more sensitive to climate change than in

- 760 the western basinin this simulation. The eastern part of the basin is more isolated from the open Atlantic Ocean than is the western part and western basin as it receives less nutrients from the Atlantic and from coastal inputs. The eastern basin is also deeper and less productive than the western basin (Crispi et al., 2001). The eastern basin exhibits a decline in the phytoplankton biomass that is similar to the decline in the phosphate concentration. Biological production is mainly P–limited in this basin
- 765 (see also Figure 12). Therefore, the constant low concentrations of phosphate observed throughout this century limit biological production and keep plankton biomass at low levels.

#### 4 Discussion

## 3.1 Biogeochemical forcingsEffects of aerosol deposition on surface primary productivity

Climate change may impact all drivers of biogeochemical cycles in the ocean. In the case of semi-enclosed

- 770 seas like the Mediterranean, the biogeochemistry is primarily influenced by external sources of nutrients (namely rivers, Atlantic and atmospheric inputs, see Ludwig et al., 2009; Krom et al., 2010) and modification of the physical ocean (vertical mixing, horizontal advection, ..., see Santinelli et al., 2012) . Nutrient fluxes from external sources (rivers, aerosols and fluxes through the Strait of Gibraltar) may evolve separately and differently depending on socio-economic decisions and climate feedbacks.
- 775 In this study, different scenarios were used for river inputs ("Order from Strength" from Ludwig et al., 2010, based on the Millenium and for Atlantic nutrient concentrations (SRES/A2 from Dufresne et al., 2013). Scenarios from the

MEA report are based on different assumptions from the IPCC SRES scenarios used to compute freshwater runoff in the HIS/A2 simulation. Moreover, the Ludwig et al. (2010) nutrient discharge transient scenario does not represent the interannual variability of nutrient runoff from rivers. Other

- 780 studies by Herrmann et al. (2014) and Macias et al. (2015) used continued present-day discharge of nutrients. As there is no consensus nor validated scenario for nutrient fluxes from riverine runoff in the Mediterranean, we chose to use one scenario from Ludwig et al. (2010). This scenario has the advantage of being derived from a coherent modeling framework. However, according to these authors, the socio-economic decisions made in the 21st century will influence nitrate and phosphate
- 785 discharge over the Mediterranean. It is difficult to forecast these decisions and the resulting changes in nutrient fluxes are uncertain. Moreover, our results emphasize that the biogeochemistry in many coastal regions of the Mediterranean Sea such as the Adriatic Sea are highly influenced by riverine nutrient inputs, which is in accordance with previous work (e.g. Spillman et al., 2007), and references therein) . In these regions, the effects of nutrient runoff from rivers seem more important than climate
- 790 change effects (see Table ). In the Adriatic basin, table is shows that riverine nitrate discharge is responsible for 41 % increase in nitrate concentration over the simulation period (difference between the 1980–1999 and the 2080–2099 periods). In the CTRL\_RG simulation, nitrate concentrations are similar to the CTRL\_R simulation showing no influence of fluxes through the Strait of Gibraltar in this region. Finally, nitrate concentrations in the HIS/A2 simulation are close to the CTRL\_R values
- 795 showing that most of the nitrate evolution in this region is linked with riverine discharge. Therefore, the choice of river runoff scenario will greatly influence the results in these regions. Associated discrepancies and the uncertainties linked with the use of inconsistent scenarios in our simulation should be addressed by developing a more integrated modelling framework to study the impacts of elimate change on the Mediterranean Sea biogeochemistry. No transient atmospheric deposition was
- 800 considered in this study because there is, to our knowledge, no transient scenario for atmospheric deposition evolution over the Mediterranean Sea. However, in order to evaluate the potential effects of aerosol deposition on the future Mediterranean Sea, we performed 2 simulations with respectively total nitrogen (NH<sub>4</sub> + NO<sub>3</sub>) deposition (labeled HIS/A2\_N) and total nitrogen and natural dust deposition (labeled HIS/A2\_NALADIN). The deposition fields are derived from the global model
- 805 LMDz–INCA and the regional model ALADIN–Climat respectively (see Richon et al.) 2017, and references therein for description in . The atmospheric deposition fields represent present–day aerosol deposition fluxes (1997–2012 and 1980–2012 for total nitrogen and dust deposition respectively) that are repeated over the 1980–2099 simulation period. The following figure (Figure 17) shows the relative effects of total nitrogen and natural dust deposition on surface primary production in the 1980–1999 and 2080–2099 periods. As
- 810 shown in Richon et al. (2017), dust deposition is a source of phosphate for the surface Mediterraneanenhances surface primary productivity in the southern part of the basin in 1980–1999 whereas nitrogen deposition enhances primary productivity in the northern part of the basin. As our HIS/A2 simulation shows a decrease in surface PO<sub>4</sub> concentrations, thus accentuating phosphate limitation over the Mediter-

23

ranean basin by the end of the 21st century, the relative effects impact of phosphate deposition from

815 dust are increased in the 2080–2099 period in comparison to the 1980–1999 would be enhanced in the 2080–2099 period relative to the 1980–1999 period. Conversely, nitrogen atmospheric deposition has very little effect on Mediterranean primary production at the end of the simulation period because most of the basin is not N–limited.

## 820 4 Discussion

#### 4.1 Climate change scenarioSources of uncertainties

The study represents the first transient long term simulations of the Mediterranean Sea with a coupled physical-biogeochemical high resolution model. It provides a first glimpse of the sensitivity of the Mediterranean Sea biogeochemistry to climate change and to the evolution of external nutrient

825 fluxes. As for all modeling studies, our conclusions are subject to some limitations that we attempt to underline in this section.

## 4.1.1 Climate change scenario

Although the physical model adequately represents the MHTC-MTHC (Adloff et al., 2015), there are many uncertainties linked with climate change projections. Some are discussed in Somot et al.

- 830 (2006), in particular, the need to using use different IPCC scenarios for climate change projections and MTHC changes. Adloff et al. (2015) apply an ensemble of SRES scenarios and boundary conditions to the Mediterranean Sea and discuss their effects on MTHC. In particular, their results suggests suggest that the choice of atmospheric and Atlantic conditions has a strong influence on the MTHC. This influence is mainly linked with the evolution of stratification index and vertical
- 835 mixing. Overall, the increase in stratification in our A2 climate change scenario leads to different evolutions of nutrient concentrations between the surface and the intermediate and deep waters with surface waters becoming more sensitive to external nutrient sources (Figures 7 and 8). On the other hand, Macias et al. (2015) found that primary productivity slightly increased as a result of decreased stratification in the climate change scenarios RCP 8.5 and RCP 4.5. The A2 scenario that we used
- 840 was the only one available with 3–D daily forcings, as necessary for coupling with the PISCES biogeochemical model. However, Adloff et al. (2015) showed that other SRES scenarios such as the A1B or B1 may lead to a future decrease decline in the vertical stratification with probably different consequences on the Mediterranean Sea biogeochemistry. Our study should be considered as is thus only a first step for transient modeling of the Mediterranean Sea biogeochemistrybut. It should be
- 845 complemented by new simulations that explore the various sources of uncertainty (model choice, internal variability, scenario choice) once appropriate forcings become available for multiple models as expected from the Med–CORDEX initiative (Ruti et al.) [2016).

## 4.2 Uncertainties from the PISCES model

The-

- 850 Freshwater runoff in the physical model may also influence the circulation and nutrient concentrations at the river mouth. Adloff et al. (2015) evaluated the changes in total freshwater runoff in the HIS/A2 simulation. Their Table 2 shows that the total freshwater runoff to the Mediterranean is lower than the Ludwig et al. (2009) estimate (by about 30 %). They found an approximately 27 % decrease in total runoff by the end of the 21st century. This trend is consistent with the deceasing trend found by
- 855 Ludwig et al. (2010). However, the 2050 estimates of freshwater runoff from Ludwig et al. (2010) are only 13 % lower than the 1970 and 2000 estimates. The freshwater runoff decrease in the physical model from Adloff et al. (2015) is more important than in the nutrient runoff model from Ludwig et al. (2010). This decrease may result in higher nutrient concentrations at the river mouth. We are also aware that the future evolution of river discharges into the Mediterranean Sea is highly
- 860 uncertain and depends on the choices for the scenario and the model (Sanchez-Gomez et al., 2009; Dubois et al., 2012; Adloff et al.,

## 4.1.1 Uncertainties from the PISCES model

The evaluation of the CTRL simulation showed that NEMOMED8/PISCES is stable over time in spite of a slight drift in nitrate concentrations (see Figure 8). Nutrient concentrations in the inter-

- 865 mediate and deep layers were shown to be underestimated in comparison to measurements (see appendix Appendix). Nutrient concentrations can be were underestimated by up to 50 %, in particular in the deep eastern basin. Moreover, nitrate fluxes from coastal discharge in CTRL are lower than in HIS/A2. The low riverine discharge and the imbalance in sources and sinks explains the loss of nitrate in the CTRL (see Figures 8 and 9). Organic forms of nutrients are not directly available
- 870 to phytoplankton in this version of PISCES, and are not included in our nutrient budgets. Powley et al. (2017) show that organic forms of nutrient are an important part of the Mediterranean elemental budgets. Therefore, we may be missing a appear to be missing part of the N and P budgets in our calculations. The simulated ehlorophyll-a chl-a vertical profiles at the DYFAMED station show a correct reasonable representation of the subsurface productivity maximum of the Mediter-
- 875 ranean in spite of a mismatch in the subsurface <u>chlorophyll\_chl-a</u> maximum depth between model and measurements. Model values were not corrected to match data, <del>and we are therefore conscious</del> that hence, the uncertainties in the representation of present-day biogeochemistry by the PISCES model may be propagated <u>in-into</u> the future.

In the **PISCES** version version of **PISCES** used in this study, variations in nitrate and phosphate concentration variations are linked by the Redfield ratio (Redfield et al., 1963). The Redfield hypothesis of a fixed nutrient ratio used for plankton growth and excretion holds true for most parts of the global ocean, but may not be true for oligotrophic regions such as the Mediterranean Sea (e.g. Béthoux and Copin-Montégut, 1986). Moreover, changes in nutrient balance influence the nutrient limitations as shown by Figure 12 The Yet, the results simulated with the Redfieldian hypothesis are

885 coherent with the observed variations of nutrient supply to the Mediterranean Sea and yield realistic biological productivity. But Nonetheless results concerning nutrient limitations might limitation could change in a non Redfieldian biogeochemistry model.

## 4.1.2 External nutrient sources

Climate change may impact all drivers of biogeochemical cycles in the ocean. In the case of semi-enclosed

- 890 seas like the Mediterranean, the biogeochemistry is heavily influenced by external sources of nutrients (namely rivers, Atlantic and atmospheric inputs, see Ludwig et al., 2009; Krom et al., 2010) and modification of the physical ocean (e.g. vertical mixing, horizontal advection, see Santinelli et al., 2012). Nutrient fluxes from external sources (rivers, aerosols and fluxes through the Strait of Gibraltar) may evolve separately depending on future socio-economic choices and climate feedbacks. In this study, different
- 895 scenarios were used for river inputs ("Order from Strength" from Ludwig et al., 2010) based on the Millenium Ecosystem Assessme and for Atlantic nutrient concentrations (SRES/A2 from Dufresne et al., 2013). It is important to accurately represent the incoming nutrient fluxes to the Mediterranean and their potential evolution with regards to climate change, as they have important influence on the Mediterranean Sea biogeochemistry.

## 900 Fluxes through the Strait of Gibraltar

Results from our CTRL\_RG simulation show that the increase in nitrate and phosphate incoming fluxes through the Strait of Gibraltar leads to higher surface concentrations in the western Mediterranean. The Atlantic nutrient concentrations are derived from a global version of the same model used our simulations (NEMO/PISCES) and forced under the same A2 climate change scenario). Therefore, there is no incompatibility issue between for the forcing and model.

### **Riverine nutrient fluxes**

905

Additionally, our CTRL\_R simulation shows that the increase in riverine nitrate fluxes leads to the accumulation of nitrate in the surface Mediterranean, in particular in the eastern basin and in the Adriatic. For the riverine nutrient inputs, scenarios from the MEA report are based on different

- 910 assumptions from the IPCC SRES scenarios used to compute freshwater runoff in the HIS/A2 simulation. Freshwater discharge from Ludwig et al. (2010) is based on the SESAME model reconstruction and differs from freshwater runoff in the ARPEGE-Climate model used to force our physical model. This may lead to incoherences between water and nutrient discharges, but the nutrient discharges from Ludwig et al. (2010) are the only ones that are available. Furthermore, the SESAME model is
- 915 not coupled with NEMO/PISCES. Associated discrepancies and the uncertainties linked with the use of inconsistent scenarios in our simulation should be addressed by developing a more integrated

modelling framework to study the impacts of climate change on the Mediterranean Sea biogeochemistry. As there is no consensus nor validated scenario for nutrient fluxes from riverine runoff in the Mediterranean, we chose to use one scenario from Ludwig et al. (2010). This scenario has the advantage

920 of being derived from a coherent modeling framework. However, the Ludwig et al. (2010) nutrient discharge transient scenario does not represent the interannual variability of nutrient runoff from rivers. Moreover, according to these authors, the socio–economic decisions made in the 21st century will influence nitrate and phosphate discharge over the Mediterranean. It is difficult to forecast these decisions and the resulting changes in nutrient fluxes are uncertain.

## 925 Potential effects of aerosol deposition

The biogeochemistry of the Mediterranean is significantly influenced by aerosol deposition (e.g. Krom et al., 2010; Dulac et al., 1989). The future evolution of the multiple aerosol sources surrounding the Mediterranean will likely influence the response of the Mediterranean to climate change.

Results from the HIS/A2\_NALADIN simulation show that enhanced phosphate fluxes from aerosols
may limit the surface decrease of phosphate concentrations and limit phosphorus limitation. However, in the HIS/A2\_NALADIN simulation, the surface Mediterranean is still P-limited in most of the Mediterranean because the atmospheric nutrient fluxes are low in comparison to riverine nutrient fluxes from rivers and the nutrient flux through the Strait of Gibraltar (see Richon et al., 2017).

Therefore, it appears unlikely that changes in aerosol deposition from natural dust would greatly

- 935 influence future Mediterranean biogeochemistry. However, there are multiple sources of aerosols that are not included in atmospheric models, e.g., anthropogenic, volcanic and volatile organic compounds (e.g. Wang et al., 2014; Kanakidou et al., 2016). Their combined influence could perhaps constitute an important nutrient flux to the Mediterranean, thus altering the evolution of its biogeochemistry. Moreover, aerosols affect radiative forcing over the Mediterranean and may impact the climate
- 940 conditions (Nabat et al., 2015b). Thus, efforts should be made to accurately represent this nutrient source in Mediterranean models to assess the effect on Mediterranean Sea biogeochemistry with regards to climate change.

Our results show that the state of the Mediterranean biogeochemistry at the end of the 21st century is the result of the combined evolutions of both climate and external nutrient fluxes. Therefore, it is

945 very difficult to predict the future evolution of the Mediterranean based on the evolution of one of these components only. This is why it is important, in the case of semi–enclosed basins, to produce reliable estimates of the evolution of all the components influencing the biogeochemistry.

## 4.2 Climate versus biogeochemical changes effects

Figure 18 summarizes the fluxes of phosphate and nitrate in and out of the Mediterranean considered 950 in this study. In general, the sum of nitrogen net fluxes into the Mediterranean basin (Riverine, Gibraltar Strait and sedimentary sources and sinks) increases by 39 % at the end of the century in the scenario (HIS/A2) whereas it is increased by 23 % in the control (CTRL) in comparison to the beginning of

- 955 the simulation(1980). The balance between inputs and outputs of phosphorus increases by 9 % in the scenario and by 11 % in the control(net gain of phosphorus in the basin). These results suggest a significant substantial accumulation of nitrogen in the Mediterranean basin over the century when phosphorus fluxes can be considered roughly stable. The strong decrease in sedimentation (Figure 6) occurring in spite of an enhancement in nutrient flux, coming enhanced nutrient flux from the
- Atlantic and an enhanced nitrate river flux may be linked to the decrease in vertical water fluxes (upwelling and downwelling). This would explain the accumulation of phosphate and nitrate in the deep layers of the Mediterranean Sea (Figures 7c, 7f 8c and 8f).
   Results from our transient simulations show that nutrient concentrations may evolve differently
- 965 In the surface western Mediterranean, the effects of climate change and enhanced nutrient fluxes via Gibraltar both concur to the increase in nutrient concentrations (Figures 7a and 8a). In the surface eastern basin, river fluxes of nitrate and stratification have opposing effects on nitrate concentrations whereas phosphate concentrations are mainly driven by climate change effects (Figures 7d and 8d). The difference between HIS/A2 and CTRL\_RG phosphate concentrations (Figure 2n the intermediate

depending on the region and the depth in response to climate change and external nutrient inputs.

970 and deep layers (Figures 75, 7c, 7c, 7f) indicates that the variations of phosphate concentrations during the 21st century are primarily linked with climate change whereas nitrate concentration seems driven by climate change while nitrate concentration is equally sensitive to changes in biogeochemical forcings (Figures 85, 8c, 8c).

Results from our different control simulations indicates the extent to which the choice of the biogeochemical

- 975 forcing scenario may influence the future evolution of the Mediterranean Sea biogeochemistry. Results from Table (CTRL\_RG minus CTRL\_R) indicate that the increase in nutrient input through the Strait of Gibraltar at the end of the century is responsible for a 2.5 % increase in chl-a concentration in the western basin by the end of the century. Herrmann et al. (2014) simulated an increase in chl-a concentration associated with climate change effects in a small region of the north western basin by
- 980 the end of the century. Thus, the separate effects of climate change and external nutrient inputs may have synergetistic effects on the evolution of the western Mediterranean chl-a. In some parts of the eastern basin, the effects from riverine nutrient fluxes on chl-a appear more important than those from climate change (see Table 4). In the Adriatic Sea, Table 3 shows that riverine nitrate discharge is responsible for 41 % increase in nitrate concentration over the simulation
- 985 period (2080–2099 minus 1980–1999). In the CTRL\_RG simulation, nitrate concentrations are similar to those in the CTRL\_R simulation, indicating no influence of fluxes through the Strait of Gibraltar in the Adriatic Sea. Finally, nitrate concentrations in the HIS/A2 simulation are close to the CTRL\_R values showing that most of the nitrate evolution in the Adriatic Sea is linked

with riverine discharge. Lazzari et al. (2014) also conclude that the river mouth regions are highly

- sensitive because the Mediterranean Sea is influenced by external nutrient inputs. The choice of river runoff scenario will likely influence the evolution of nutrient concentrations and the biogeochemistry in many coastal regions such as the Adriatic Sea (see also Spillman et al., 2007).
   To our knowledge, this is the first attempt to study the basin–scale biogeochemical evolution using a transient business–as–usual (A2) climate change scenario. Lazzari et al. (2014) tested the effects
- of several land-use change scenarios on the A1B SRES climate change scenario over 10-years time slices. They found a general decrease in phytoplankton and zooplankton biomasses\_biomass (about 5 %)that-, which is lower than in our severe climate change scenario. Considering only changes in climate, Herrmann et al. (2014) and Macias et al. (2015) studied the transient biogeochemical evolution of the Mediterranean Sea (Herrmann et al.) 2014, only studied a small region in the north western Mediterranean).
- studied the transient biogeochemical evolution of the Mediterranean Sea under different climate change scenarios, with the former study focusing on a small region in the northwestern Mediterranean. Both studies found that chlorophyll concentration and plankton biomass increase slightly due to changes in vertical stratification. In our simulations, average phytoplankton biomass decreases by about 2 to 30 % (see Figure 15) and average zooplankton biomass decreases by about 8 and
- 1005 to 12 % (see Figure 6). However, our transient simulations revealed non linear trends in plankton biomass evolution . Lazzari et al. (2014) also conclude that the river mouth regions are highly sensitive because the Mediterranean Sea is influenced by external nutrient inputs. Our results show the same sensitivity of the Mediterranean to external nutrient inputs. Herrmann et al. (2014) studied the transient biogeochemical evolution of the northwestern Mediterranean Sea under the A2 and A1B
- 1010 scenarios with the coupled ECO3M–S/SYMPHONIE model. But they used present–day conditions for biogeochemical forcings, as did Macias et al. (2015). Results from Herrmann et al. (2014) indicate that chlorophyll concentration and plankton biomass in as a results of the northwestern Mediterranean increase slightly as a result of vertical stratificationinfluence of external nutrient fluxes. Chust et al. (2014) have shown that regional seas and in particular the Aegean and Adriatic were sensitive to trophic
- 1015 amplification. Our results indicate that the contrasting effects of vertical stratification biogeochemical changes may lead to a decrease in chlorophyll concentration, phytoplankton and zooplankton biomass content of up to 50 % locally and between 2 and 30 % at the basin scale as indicated by Figures 14 appear to agree, showing signs of trophic amplification (see Figures 15 and 16-). Assessing the sensitivity of the Mediterranean to trophic amplification would require more simulations focused on the evolution
- 1020 of Mediterranean planktonic biomass under different climate change scenarios. The modifications of chlorophyll chl-a production and plankton biomass are linked to changes in nutrient limitation (Figure 12). Our finding that most of the Mediterranean basin is N and P co-limited seems in contrast with previous literature on the matter (see Krom et al.) [2004] [2010]; Pujo-Pay et al.] [2011], and references therein) . These authors found from analyses of the N:P ratio of the waters a clear phosphorus limitation in the
- 1025 major part of the Mediterranean. The discrepancy between our results and literature estimates comes

from the way we calculate nutrient limitations. Considering how low nutrient concentrations are in the Mediterranean and how low the nutrient limitation factors are, the small difference between the limitation factors indicate that the Mediterranean is both limited in P and N. Finding no clear definition of nutrient co–limitation, we propose to consider that N and P are co-limiting when the

- 1030 difference in limitation factors is less than 1 %. This definition of nutrient co–limitation applies well to the Mediterranean case because of the its very low nutrient concentrations. Chust et al. (2014) have shown that regional seas and in particular the Aegean and Adriatic were sensitive to trophic amplification. Our results seem to agree with these conclusions by showing sign of trophic amplification (see Figures [5] and [6]). Finally, Luna et al. (2012) hypothesise Our results are confirmed by some
- 1035 studies (Thingstad et al., 2005; [Tanaka et al., 2011). However, our nutrient limitations are calculated from 20-years average nutrient concentrations and nutrient limitation may vary greatly during the seasonal cycle (Marty et al., 2002; [Diaz et al., 2001). It has also been hypothesized by [Luna et al.] (2012) that the warm temperature of the deep Mediterranean may be a cause for important enhance nutrient recycling via prokaryotic metabolism. In the version of PISCES used in this study, nutrient
- 1040 recycling is dependant on oxygen, depth, plankton biomass and bacterial activity. Therefore, we could not observe the effects of temperature on nutrient recycling. Results from our different control simulations indicates the extent to which the choice of the biogeochemical forcing scenario may influence the future evolution of the Mediterranean Sea biogeochemistry. In particular, nutrient inputs through the Strait of Gibraltar have substantial consequences on the western basin. Results
- 1045 from Figures 7a and 8a and Table 1 indicate that the increase in nutrient inputs through the Strait of Gibraltar at the end of the century is responsible for a 2.5 % increase in chlorophyll concentration in the western basin during the 2080–2099 period. Moreover, climate and nutrient forcing changes may have contrasting influences on the Mediterranean Sea biogeochemistry. Stratification may lead to increased productivity in the surface because Therefore, a part of the nutrient concentration increase
- 1050 (see also Macias et al., 2015), while decreasing coastal discharges of phosphate may decrease the productivity in the basin. accumulation we observed may be linked with the increase in temperature.

## Conclusion

- This study aims at assessing the transient effects of climate and biogeochemical changes on the 1055 Mediterranean Sea biogeogeochemistry under the high-emission IPCC-SRES A2 scenario. The NEMOMED8/PISCES model adequately reproduces the main characteristics of the Mediterranean Sea: the west-to-east gradient of in productivity, the main productive zones, and the presence of a DCM. Hence, it appears reasonable to use it to study the future evolution of the biogeochemistry of the Mediterranean basin in response to increasing atmospheric CO<sub>2</sub> and resulting climate change.
- 1060 For the first time, we performed Our study is the first to offer a continuous simulation over the entire

period of the future IPCC scenario (A2), between 1980-2000 and 2099.

This study illustrates Its results illustrate how future changes in physical and biogeochemical conditions, including warming, increased stratification, and changes in Atlantic and river inputs, can lead to a significant accumulation of nitrate and a decrease in biological productivity in the surface, thus

- 1065 affecting the entire Mediterranean ecosystem. Our results also illustrate how climate change and nutrient inputs from riverine sources and fluxes through the Strait of Gibraltar have contrasted contrasting influences on the Mediterranean Sea productivity. In particular, the biogeochemistry in the western basin displays similar nutrient trends as does its trends as that for nutrient input across the Strait of Gibraltar. Therefore, it appears critical
- 1070 to correctly represent the future variations of external biogeochemical forcings of the Mediterranean Sea as they may have an equally important influence on <u>surface</u> biogeochemical cycles as <u>does</u> climate. The biogeochemistry of the eastern basin is more sensitive to vertical mixing and river inputs than <u>is</u> the western basin<del>(that receives regulating effects from exchanges, which is regulated by input</del> through the Strait of Gibraltar<del>) and the stratification observed in the future leads to a reduction in</del>
- 1075 surface productivity. Increased future stratification also reduces surface productivity in the eastern basin.

Finally, this study accounts Although this study does account for the changes in fluxes through the Strait of Gibraltar and riverine inputs, but some potentially important sources are missing such as direct wastewater discharge, submarine groundwater input and atmospheric deposition. Measurements

- 1080 and models are still missing in order to include These additional nutrient sources are poorly known, with a general lack of both measurements and models as needed to build comprehensive datasets for past and future evolution of these nutrient sources. The HIS/A2\_N and HIS/A2\_NALADIN simulations presented in the discussion section include continued present-day this study include continued present-day nitrogen and phosphate deposition. Although these atmospheric fluxes have been eval-
- 1085 uated previously and were shown to represent correctly the deposition fluxes, there is no guarantee that these fluxes are going to will remain constant over the next century. Results showed indicate that the future sensitivity of the Mediterranean to atmospheric deposition depends on the surface nutrient limitationslimitation, which may in part be influenced by aerosol deposition. However, there is to our knowledge no available transient scenario for the 21st century evolution of atmospheric depo-
- 1090 sition and no ensemble simulations to assess the future evolution of the Mediterranean Sea under different climate change scenarios. A new generation of fully coupled regional models have been developed and used to study aerosols climatic impacts (Nabat et al., 2015b). These models include a representation of the ocean, atmosphere, aerosols and rivers and should could eventually be used to perform make consistent future climate projections at the Mediterranean regional scale regional
- 1095 scale of the Mediterranean.

## acknowledgmentsAcknowledgments

The authors would like to thank Florence Sevault for the physical simulations of NEMOMED8, Pierre Nabat for the dust deposition with ALADIN-Climat, Yves Balkanski and Rong Wang for the N deposition with the LMDz-INCA model and Wolfgang Ludwig for the river inputs. This work was

1100 funded by CEA (C. Richon PhD grant) and is part of the MISTRALS project. Simulations were made using HPC resources from the French GENCI program (grant x2015010040).

#### Appendix A: Evaluation of the NEMOMED8/PISCES model

The comparison of modeled surface <u>chlorophyll-a</u> <u>chl-a</u> concentration with satellite estimates has 1105 revealed that the model correctly simulates the main characteristics observed in the Mediterranean Sea (Figures 1] and 2). Comparison with in situ observation provides more refined estimates.

Figure AI presents the average chlorophyll-a\_chl-a profiles at the DYFAMED station (43.25°N, 7.52°E) compared with measured concentrations for the month of February (low stratification, high

- 1110 productivity) and May (end of spring bloom, beginning of stratification and DCM appearance). There are few data points below 200 m. The model produces the characteristic of the deep chlorophyll chl-a maximum generated in May, even if its depth is too importantit is too deep. The colors show that the model represents some interannual variability in chlorophyll chl-a production in spite of the a consistent bias.
- 1115

The overestimation of the overestimated DCM depth may be due to the overestimation of nitracline and phosphacline as shown by figure A2 This Figure shows the vertical profiles of nitrate and phosphate at the DYFAMED station in May, colors represent the different years in the model and in the observation.

## 1120

The vertical distribution of nitrate and phosphate concentrations along a West-to-East-west-to-east transect is shown in Figure A3. The model produces the salient West-to-East-west-to-east gradient of nutrient concentrations. Concentrations in the surface layer seem correctappear realistic. The nutricline is located between 100 to and 150 m deep in the western basin and deepens to around 180 to

1125 200 m in the eastern basin. Although the model represents the spatial variability of the nutricline, it is too smooth, leading to the underestimation of deep water concentrations (by about 30 to 50 %).

## References

Adloff, F., Somot, S., Sevault, F., Jordà, G., Aznar, R., Déqué, M., Herrmann, M., Marcos, M., Dubois, C.,

- Padorno, E., Alvarez-Fanjul, E., and Gomis, D.: Mediterranean Sea response to climate change in an ensemble of twenty first century scenarios, Climate Dynamics, 45, 2775–2802, doi:10.1007/s00382-015-2507-3, http://link.springer.com/article/10.1007/s00382-015-2507-3, 2015.
  - Albouy, C., Leprieur, F., Le Loc'h, F., Mouquet, N., Meynard, C. N., Douzery, E. J. P., and Mouillot, D.: Projected impacts of climate warming on the functional and phylogenetic components of coastal Mediter-
- 1135 ranean fish biodiversity, Ecography, 38, 681–689, doi 10.1111/ecog.01254, http://doi.wiley.com/10.1111/ ecog.01254, 2015.
  - Andrello, M., Mouillot, D., Somot, S., Thuiller, W., and Manel, S.: Additive effects of climate change on connectivity between marine protected areas and larval supply to fished areas, Diversity and Distributions, 21, 139–150, doi:10.1111/ddi.12250, http://onlinelibrary.wiley.com/doi/10.1111/ddi.12250/abstract, 2015.
- 1140 Auger, P. A., Ulses, C., Estournel, C., Stemmann, L., Somot, S., and Diaz, F.: Interannual control of plankton communities by deep winter mixing and prey/predator interactions in the NW Mediterranean: Results from a 30-year 3D modeling study, Progress in Oceanography, 124, 12–27, doi:10.1016/j.pocean.2014.04.004, http://www.sciencedirect.com/science/article/pii/S0079661114000524, 2014.
- Aumont, O. and Bopp, L.: Globalizing results from ocean in situ iron fertilization studies: GLOBALIZING
- 1145 IRON FERTILIZATION, Global Biogeochemical Cycles, 20, n/a–n/a, doi:10.1029/2005GB002591, http: //doi.wiley.com/10.1029/2005GB002591, 2006.
- Beuvier, J., Sevault, F., Herrmann, M., Kontoyiannis, H., Ludwig, W., Rixen, M., Stanev, E., Béranger, K., and Somot, S.: Modeling the Mediterranean Sea interannual variability during 1961–2000: Focus on the Eastern Mediterranean Transient, Journal of Geophysical Research: Oceans, 115, C08017, doi:10.1029/2009JC005950, http://onlinelibrary.wiley.com/doi/10.1029/2009JC005950/abstract, 2010.
- Bonnet, S., Grosso, O., and Moutin, T.: Planktonic dinitrogen fixation along a longitudinal gradient across the Mediterranean Sea during the stratified period (BOUM cruise), Biogeosciences, 8, 2257–2267, doi:10.5194/bg-8-2257-2011, http://www.biogeosciences.net/8/2257/2011/, 2011.
  - Bosc, E., Bricaud, A., and Antoine, D.: Seasonal and interannual variability in algal biomass and
- 1155 primary production in the Mediterranean Sea, as derived from 4 years of SeaWiFS observations: MEDITERRANEAN SEA BIOMASS AND PRODUCTION, Global Biogeochemical Cycles, 18, n/a–n/a, doi;10.1029/2003GB002034, http://doi.wiley.com/10.1029/2003GB002034, 2004.

Béthoux, J. P. and Copin-Montégut, G.: Biological fixation of atmospheric nitrogen in the Mediterranean Sea,

- Limnology and Oceanography, 31, 1353–1358, doi:10.4319/lo.1986.31.6.1353, http://onlinelibrary.wiley.
- 1160 com/doi/10.4319/lo.1986.31.6.1353/abstract, 1986.
  - Béthoux, J. P., Morin, P., Chaumery, C., Connan, O., Gentili, B., and Ruiz-Pino, D.: Nutrients in the Mediterranean Sea, mass balance and statistical analysis of concentrations with respect to environmental change, Marine Chemistry, 63, 155–169, doi:10.1016/S0304-4203(98)00059-0, http://www.sciencedirect.com/science/
  - article/pii/S0304420398000590, 1998.
- 1165 Christodoulaki, S., Petihakis, G., Kanakidou, M., Mihalopoulos, N., Tsiaras, K., and Triantafyllou, G.: Atmospheric deposition in the Eastern Mediterranean. A driving force for ecosystem dynamics, Journal of Ma-

rine Systems, 109-110, 78–93, doi:10.1016/j.jmarsys.2012.07.007, http://linkinghub.elsevier.com/retrieve/ pii/S0924796312001583, 2013.

Chust, G., Allen, J. I., Bopp, L., Schrum, C., Holt, J., Tsiaras, K., Zavatarelli, M., Chifflet, M., Cannaby, H.,

- Dadou, I., Daewel, U., Wakelin, S. L., Machu, E., Pushpadas, D., Butenschon, M., Artioli, Y., Petihakis, G., Smith, C., Garçon, V., Goubanova, K., Vu, B. L., Fach, B. A., Salihoglu, B., Clementi, E., and Irigoien, X.: Biomass changes and trophic amplification of plankton in a warmer ocean, Global Change Biology, 20, 2124–2139, doi:10.1111/gcb.12562, https://onlinelibrary.wiley.com/doi/abs/10.1111/gcb.12562, 2014.
- Civitarese, G., Gacic, M., Lipizer, M., and Borzelli, G. L. E.: On the impact of the Bimodal Oscillating System (BiOS) on the biogeochemistry and biology of the Adriatic and Ionian Seas (Eastern Mediterranean),
  - Biogeosciences, 7, 3987–3997, doi:10.5194/bg-7-3987-2010, wOS:000285574100006, 2010.
    Claustre, H., Morel, A., Hooker, S. B., Babin, M., Antoine, D., Oubelkheir, K., Bricaud, A., Leblanc, K., Quéguiner, B., and Maritorena, S.: Is desert dust making oligotrophic waters greener?, Geophysical Research
    - Letters, 29, 107–1–107–4, doi 10.1029/2001GL014056, https://agupubs.onlinelibrary.wiley.com/doi/abs/10.

1180

- Cork, S., Peterson, G., Petschel-Held, G., Alcamo, J., Alder, J., Bennett, E., Carr, E., Deane, D., Nelson, G., Ribeiro, T., and others: Four scenarios, Ecosystems and human well-being: Scenarios, 2, http://www.academia.edu/download/31005604/Millennium\_assessment.pdf, 2005.
- Crispi, G., Mosetti, R., Solidoro, C., and Crise, A.: Nutrients cycling in Mediterranean basins: the role
- 1185 of the biological pump in the trophic regime, Ecological Modelling, 138, 101–114, doi:10.1016/S0304-3800(00)00396-3, http://www.sciencedirect.com/science/article/pii/S0304380000003963, 2001.
  - Diaz, F., Raimbault, P., Boudjellal, B., Garcia, N., and Moutin, T.: Early spring phosphorus limitation of primary productivity in a NW Mediterranean coastal zone (Gulf of Lions), Marine Ecology Progress Series, 211, 51– 62, doi:[10.3354/meps211051], http://www.int-res.com/abstracts/meps/v211/p51-62/, 2001.
- 1190 d'Ortenzio, F. and Ribera d'Alcalà, M.: On the trophic regimes of the Mediterranean Sea: a satellite analysis, Biogeosciences, 6, 139–148, http://www.biogeosciences.net/6/139/2009/bg-6-139-2009.html, 2009.
  - Déqué, M., Dreveton, C., Braun, A., and Cariolle, D.: The ARPEGE/IFS atmosphere model: a contribution
     to the French community climate modelling, Climate dynamics, 10, 249–266, <a href="http://cat.inist.fr/?aModele=afficheN&cpsidt=4250659">http://cat.inist.fr/?aModele=afficheN&cpsidt=4250659</a>, 1994.
- 1195 Dubois, C., Somot, S., Calmanti, S., Carillo, A., Déqué, M., Dell'Aquilla, A., Elizalde, A., Gualdi, S., Jacob, D., L'Hévéder, B., Li, L., Oddo, P., Sannino, G., Scoccimarro, E., and Sevault, F.: Future projections of the surface heat and water budgets of the Mediterranean Sea in an ensemble of coupled atmosphere-ocean
  - regional climate models, Climate Dynamics, 39, 1859–1884, doi:10.1007/s00382-011-1261-4, http://adsabs. harvard.edu/abs/2012ClDy...39.1859D, 2012.
- 1200 Dufresne, J.-L., Foujols, M.-A., Denvil, S., Caubel, A., Marti, O., Aumont, O., Balkanski, Y., Bekki, S., Bellenger, H., Benshila, R., Bony, S., Bopp, L., Braconnot, P., Brockmann, P., Cadule, P., Cheruy, F., Codron, F., Cozic, A., Cugnet, D., Noblet, N. d., Duvel, J.-P., Ethé, C., Fairhead, L., Fichefet, T., Flavoni, S., Friedlingstein, P., Grandpeix, J.-Y., Guez, L., Guilyardi, E., Hauglustaine, D., Hourdin, F., Idelkadi, A., Ghattas, J., Joussaume, S., Kageyama, M., Krinner, G., Labetoulle, S., Lahellec, A., Lefebvre, M.-P., Lefevre, F.,
- 1205 Levy, C., Li, Z. X., Lloyd, J., Lott, F., Madec, G., Mancip, M., Marchand, M., Masson, S., Meurdesoif, Y., Mignot, J., Musat, I., Parouty, S., Polcher, J., Rio, C., Schulz, M., Swingedouw, D., Szopa, S., Talandier,

C., Terray, P., Viovy, N., and Vuichard, N.: Climate change projections using the IPSL-CM5 Earth System Model: from CMIP3 to CMIP5, Climate Dynamics, 40, 2123–2165, doi:10.1007/s00382-012-1636-1, https://link.springer.com/article/10.1007/s00382-012-1636-1, 2013.

1210 Dulac, F., BUAT-MÉNARD, P., Ezat, U., Melki, S., and Bergametti, G.: Atmospheric input of trace metals to the western Mediterranean: uncertainties in modelling dry deposition from cascade impactor data, Tellus B, 41, 362–378, http://onlinelibrary.wiley.com/doi/10.1111/j.1600-0889.1989.tb00315.x/full, 1989.

1215

- Dunić, N., Vilibić, I., Šepić, J., Somot, S., and Sevault, F.: Dense water formation and BiOS-induced variability in the Adriatic Sea simulated using an ocean regional circulation model, Climate Dynamics, doi:10.1007/s00382-016-3310-5, http://link.springer.com/10.1007/s00382-016-3310-5, 2016.
- Faugeras, B., Lévy, M., Mémery, L., Verron, J., Blum, J., and Charpentier, I.: Can biogeochemical fluxes be recovered from nitrate and chlorophyll data? A case study assimilating data in the Northwestern Mediterranean
  Sea at the JGOFS-DYFAMED station, Journal of Marine Systems, 40-41, 99–125, doi:10.1016/S0924-7963(03)00015-0, http://www.sciencedirect.com/science/article/pii/S0924796303000150, 2003.
- Fichaut, M., Garcia, M. J., Giorgetti, A., Iona, A., Kuznetsov, A., Rixen, M., and Group, M.: MEDAR/MEDATLAS 2002: A Mediterranean and Black Sea database for operational oceanography, in: Elsevier Oceanography Series, edited by H. Dahlin, K. Nittis and S. E. Petersson, N. C. F., vol. 69 of *Building the European Capacity in Operational OceanographyProceedings of the Third International Conference on EuroGOOS*, pp. 645–648, Elsevier, http://www.sciencedirect.com/science/article/pii/S0422989403801071, 2003.
- Gallisai, R., Peters, F., Volpe, G., Basart, S., and Baldasano, J. M.: Saharan Dust Deposition May Affect Phytoplankton Growth in the Mediterranean Sea at Ecological Time Scales, PLoS ONE, 9, e110762, doi:10.1371/journal.pone.0110762, http://dx.plos.org/10.1371/journal.pone.0110762, 2014.
- Gibelin, A.-L. and Déqué, M.: Anthropogenic climate change over the Mediterranean region simulated by a
- 1230 global variable resolution model, Climate Dynamics, 20, 327–339, doi 10.1007/s00382-002-0277-1, https: //link.springer.com/article/10.1007/s00382-002-0277-1, 2003.
  - Giorgi, F.: Climate change hot-spots, Geophysical Research Letters, 33, L08 707, doi:10.1029/2006GL025734, http://onlinelibrary.wiley.com/doi/10.1029/2006GL025734/abstract, 2006.
  - Giorgi, F. and Lionello, P.: Climate change projections for the Mediterranean region, Global and Planetary
- 1235 Change, 63, 90–104, doi:10.1016/j.gloplacha.2007.09.005, http://www.sciencedirect.com/science/article/pii/ S0921818107001750, 2008.
  - Gómez, F.: The role of the exchanges through the Strait of Gibraltar on the budget of elements in the Western Mediterranean Sea: consequences of human-induced modifications, Marine Pollution Bul-
- letin, 46, 685–694, doi 10.1016/S0025-326X(03)00123-1, http://www.sciencedirect.com/science/article/pii/
   S0025326X03001231, 2003.
  - Guieu, C., Ridame, C., Pulido-Villena, E., Bressac, M., Desboeufs, K., and Dulac, F.: Dust deposition in an oligotrophic marine environment: impact on the carbon budget, Biogeosciences Discussions, 11, 1707–1738, doi:[10.5194/bgd-11-1707-2014, http://www.biogeosciences-discuss.net/11/1707/2014/, 2014.

Harley, C. D. G., Randall Hughes, A., Hultgren, K. M., Miner, B. G., Sorte, C. J. B., Thornber, C. S., Ro-

1245 driguez, L. F., Tomanek, L., and Williams, S. L.: The impacts of climate change in coastal marine systems,

- Ecology Letters, 9, 228–241, doi 10.1111/j.1461-0248.2005.00871.x, http://onlinelibrary.wiley.com/doi/10. 1111/j.1461-0248.2005.00871.x/abstract, 2006.
- Hattab, T., Albouy, C., Lasram, F. B. R., Somot, S., Le Loc'h, F., and Leprieur, F.: Towards a better understanding of potential impacts of climate change on marine species distribution: a multiscale modelling approach,
- 1250 Global Ecology and Biogeography, 23, 1417–1429, doi:10.1111/geb.12217, http://onlinelibrary.wiley.com/ doi/10.1111/geb.12217/abstract, 2014.

Hauglustaine, D. A., Balkanski, Y., and Schulz, M.: A global model simulation of present and future nitrate aerosols and their direct radiative forcing of climate, Atmos. Chem. Phys., 14, 11031–11063, doi:10.5194/acp-14-11031-2014, http://www.atmos-chem-phys.net/14/11031/2014/, 2014.

- Herrmann, M., Sevault, F., Beuvier, J., and Somot, S.: What induced the exceptional 2005 convection event in the northwestern Mediterranean basin? Answers from a modeling study, Journal of Geophysical Research: Oceans, 115, C12 051, doi:10.1029/2010JC006162, http://onlinelibrary.wiley.com/doi/10.1029/2010JC006162/abstract, 2010.
  - Herrmann, M., Diaz, F., Estournel, C., Marsaleix, P., and Ulses, C.: Impact of atmospheric and oceanic in-
- 1260 terannual variability on the Northwestern Mediterranean Sea pelagic planktonic ecosystem and associated carbon cycle, Journal of Geophysical Research: Oceans, 118, 5792–5813, doi:10.1002/jgrc.20405/ http://onlinelibrary.wiley.com/doi/10.1002/jgrc.20405/abstract, 2013.
  - Herrmann, M., Estournel, C., Adloff, F., and Diaz, F.: Impact of climate change on the northwestern Mediterranean Sea pelagic planktonic ecosystem and associated carbon cycle, Journal of Geophysical Re-
- 1265 search: Oceans, 119, 5815–5836, doi:10.1002/2014JC010016, http://onlinelibrary.wiley.com/doi/10.1002/ 2014JC010016/abstract, 2014.
  - Huertas, I. E., Ríos, A. F., García-Lafuente, J., Navarro, G., Makaoui, A., Sánchez-Román, A., Rodriguez-Galvez, S., Orbi, A., Ruíz, J., and Pérez, F. F.: Atlantic forcing of the Mediterranean oligotrophy, Global Biogeochemical Cycles, 26, GB2022, doi:10.1029/2011GB004167, http://onlinelibrary.wiley.com/doi/10.1029/
- 1270 2011GB004167/abstract, 2012.
  - Ibello, V., Cantoni, C., Cozzi, S., and Civitarese, G.: First basin-wide experimental results on N2 fixation in the open Mediterranean Sea, Geophysical Research Letters, 37, L03 608, doi:10.1029/2009GL041635, http: //onlinelibrary.wiley.com/doi/10.1029/2009GL041635/abstract, 2010.

IPCC: Managing the Risks of Extreme Events and Disasters to Advance Climate Change Adaptation. A Special

1275 Report of Working Groups I and II of the Intergovernmental Panel on Climate Change, Cambridge University Press, Cambridge, United Kingdom, and New York, NY, USA, 2012.

IPCC and Working Group III: Emissions scenarios. a special report of IPCC Working Group III, Intergovernmental Panel on Climate Change, Geneva, http://catalog.hathitrust.org/api/volumes/oclc/59436339.html, oCLC: 891560334, 2000.

 Jordà, G., Marbà, N., and Duarte, C. M.: Mediterranean seagrass vulnerable to regional climate warming, Nature Climate Change, 2, 821–824, doi:10.1038/nclimate1533, http://www.nature.com/nclimate/journal/v2/ n11/abs/nclimate1533.html?foxtrotcallback=true, 2012.

Kanakidou, M., Myriokefalitakis, S., Daskalakis, N., Fanourgakis, G., Nenes, A., Baker, A. R., Tsigaridis, K., and Mihalopoulos, N.: Past, Present, and Future Atmospheric Nitrogen Deposition, Journal of the Atmo-

- 1285 spheric Sciences, 73, 2039–2047, doi:10.1175/JAS-D-15-0278.1, http://journals.ametsoc.org/doi/10.1175/ JAS-D-15-0278.1, 2016.
  - Klein, B., Roether, W., Kress, N., Manca, B. B., Ribera d'Alcala, M., Souvermezoglou, E., Theocharis, A., Civitarese, G., and Luchetta, A.: Accelerated oxygen consumption in eastern Mediterranean deep waters following the recent changes in thermohaline circulation, Journal of Geophysical Research: Oceans, 108, 8107,

1290 doi:10.1029/2002JC001454, http://onlinelibrary.wiley.com/doi/10.1029/2002JC001454/abstract, 2003.

- Krom, M. D., Herut, B., and Mantoura, R. F. C.: Nutrient budget for the Eastern Mediterranean: Implications for phosphorus limitation, Limnology and Oceanography, 49, 1582–1592, doi:10.4319/lo.2004.49.5.1582, https://aslopubs.onlinelibrary.wiley.com/doi/abs/10.4319/lo.2004.49.5.1582, 2004.
- Krom, M. D., Emeis, K.-C., and Van Cappellen, P.: Why is the Eastern Mediterranean phosphorus limited?,
- 1295 Progress in Oceanography, 85, 236–244, doi:10.1016/j.pocean.2010.03.003, http://www.sciencedirect.com/ science/article/pii/S0079661110000340, 2010.

Lascaratos, A., Roether, W., Nittis, K., and Klein, B.: Recent changes in deep water formation and spread-

- ing in the eastern Mediterranean Sea: a review, Progress in Oceanography, 44, 5–36, doi:10.1016/S0079-6611(99)00019-1, http://www.sciencedirect.com/science/article/pii/S0079661199000191, 1999.
- 1300 Lazzari, P., Mattia, G., Solidoro, C., Salon, S., Crise, A., Zavatarelli, M., Oddo, P., and Vichi, M.: The impacts of climate change and environmental management policies on the trophic regimes in the Mediterranean
  - Sea: Scenario analyses, Journal of Marine Systems, 135, 137–149, doi:10.1016/j.jmarsys.2013.06.005, http: //linkinghub.elsevier.com/retrieve/pii/S0924796313001425, 2014.
  - Lefort, S., Aumont, O., Bopp, L., Arsouze, T., Gehlen, M., and Maury, O.: Spatial and body-size dependent
- response of marine pelagic communities to projected global climate change, Global Change Biology, 21, 154–164, doi:10.1111/gcb.12679, http://onlinelibrary.wiley.com/doi/10.1111/gcb.12679/abstract, 2015.

Locarnini, R. A., Garcia, H. E., Boyer, T. P., Antonov, J. I., and Levitus, S.: World Ocean Atlas 2005, Volume 3: Dissolved Oxygen, Apparent Oxygen Utilization, and Oxygen Saturation [+DVD], NOAA Atlas NESDIS, http://www.vliz.be/nl/imis?module=ref&refid=117383&printversion=1&dropIMIStitle=1, 2006.

- 1310 Ludwig, W., Dumont, E., Meybeck, M., and Heussner, S.: River discharges of water and nutrients to the Mediterranean and Black Sea: Major drivers for ecosystem changes during past and future decades?, Progress in
  - Oceanography, 80, 199–217, doi:10.1016/j.pocean.2009.02.001, http://linkinghub.elsevier.com/retrieve/pii/ S0079661109000020, 2009.

- Biogeochemical Cycles, 24, GB0A13, doi:10.1029/2009GB003594, http://onlinelibrary.wiley.com/doi/10. 1029/2009GB003594/abstract, 2010.
- Luna, G. M., Bianchelli, S., Decembrini, F., Domenico, E. D., Danovaro, R., and Dell'Anno, A.: The dark portion of the Mediterranean Sea is a bioreactor of organic matter cycling, Global Biogeochemical Cycles,
- 1320 26, doi:10.1029/2011GB004168, https://agupubs.onlinelibrary.wiley.com/doi/abs/10.1029/2011GB004168, 2012.
  - Macias, D. M., Garcia-Gorriz, E., and Stips, A.: Productivity changes in the Mediterranean Sea for the twentyfirst century in response to changes in the regional atmospheric forcing, Frontiers in Marine Science, 2, doi:10.3389/fmars.2015.00079, https://www.frontiersin.org/articles/10.3389/fmars.2015.00079/full, 2015.

Ludwig, W., Bouwman, A. F., Dumont, E., and Lespinas, F.: Water and nutrient fluxes from major Mediterranean and Black Sea rivers: Past and future trends and their implications for the basin-scale budgets, Global

1325 Madec, G.: NEMO ocean engine, http://eprints.soton.ac.uk/64324/, 2008.

- Marty, J.-C., Chiavérini, J., Pizay, M. D., and Avril, B.: Seasonal and interannual dynamics of nutrients and phytoplankton pigments in the western Mediterranean Sea at the DYFAMED time-series station (1991-
- 1999), Deep-Sea Research, Part II. Topical Studies in Oceanography, http://www.vliz.be/en/imis?module= ref&refid=39389&printversion=1&dropIMIStitle=1, 2002.

1330 Monod, J.: Recherches sur la croissance des cultures bactériennes, Hermann, 1958.

Morel, A. and Gentili, B.: Yellow substance and the shades of blue in the Mediterranean Sea, Biogeosciences Discussions, 6, 8503–8530, http://adsabs.harvard.edu/abs/2009BGD.....6.8503M, 2009.

Moutin, T., Van Wambeke, F., and Prieur, L.: Introduction to the Biogeochemistry from the Oligotrophic to

- the Ultraoligotrophic Mediterranean (BOUM) experiment, Biogeosciences, 9, 3817–3825, doi:10.5194/bg 9-3817-2012, http://www.biogeosciences.net/9/3817/2012/, 2012.
  - Nabat, P., Somot, S., Mallet, M., Michou, M., Sevault, F., Driouech, F., Meloni, D., di Sarra, A., Di Biagio,
    C., Formenti, P., Sicard, M., Léon, J.-F., and Bouin, M.-N.: Dust aerosol radiative effects during summer
    2012 simulated with a coupled regional aerosol-atmosphere-ocean model over the Mediterranean, Atmos.

Chem. Phys., 15, 3303–3326, doi:10.5194/acp-15-3303-2015, http://www.atmos-chem-phys.net/15/3303/ 2015/, 2015a.

- Nabat, P., Somot, S., Mallet, M., Sevault, F., Chiacchio, M., and Wild, M.: Direct and semi-direct aerosol radiative effect on the Mediterranean climate variability using a coupled regional climate system model, Cli-
- mate Dynamics, 44, 1127–1155, doi:10.1007/s00382-014-2205-6, https://link.springer.com/article/10.1007/s00382-014-2205-6, 2015b.
- 1345 Nittis, K., Lascaratos, A., and Theocharis, A.: Dense water formation in the Aegean Sea: Numerical simulations during the Eastern Mediterranean Transient, Journal of Geophysical Research: Oceans, 108, 8120, doi:10.1029/2002JC001352, http://onlinelibrary.wiley.com/doi/10.1029/2002JC001352/abstract, 2003.

Palmieri, J., Orr, J. C., Dutay, J.-C., Béranger, K., Schneider, A., Beuvier, J., and Somot, S.: Simulated anthropogenic CO2 storage and acidification of the Mediterranean Sea, Biogeosciences, 12, 781–802, doi:10.5194/bg-12-781-2015. http://www.biogeosciences.net/12/781/2015/, 2015.

- Powley, H. R., Dürr, H. H., Lima, A. T., Krom, M. D., and Van Cappellen, P.: Direct Discharges of Domestic
  Wastewater are a Major Source of Phosphorus and Nitrogen to the Mediterranean Sea, <a href="https://core.ac.uk/display/74234820">https://core.ac.uk/display/74234820</a>, 2016.
- Powley, H. R., Krom, M. D., and Cappellen, P. V.: Understanding the unique biogeochemistry of theMediterranean Sea: Insights from a coupled phosphorus and nitrogen model, Global Biogeochemical Cy
  - cles, 31, 1010–1031, doi:10.1002/2017GB005648, https://agupubs.onlinelibrary.wiley.com/doi/abs/10.1002/2017GB005648, 2017.
  - Pujo-Pay, M., Conan, P., Oriol, L., Cornet-Barthaux, V., Falco, C., Ghiglione, J.-F., Goyet, C., Moutin, T., and Prieur, L.: Integrated survey of elemental stoichiometry (C, N, P) from the western to eastern Mediterranean
- Sea, Biogeosciences, 8, 883–899, doi:10.5194/bg-8-883-2011, http://www.biogeosciences.net/8/883/2011/, 2011.

Redfield, A. C., Ketchum, B. H., and Richards, F. A.: The influence of organisms on the composition of sea-

water, The sea: ideas and observations on progress in the study of the seas, <a href="http://www.vliz.be/en/imis?module=ref&refid=28944&printversion=1&dropIMIStitle=1">http://www.vliz.be/en/imis?module=ref&refid=28944&printversion=1&dropIMIStitle=1</a>, 1963.

1340

1350

- 1365 Richon, C., Dutay, J.-C., Dulac, F., Wang, R., Balkanski, Y., Nabat, P., Aumont, O., Desboeufs, K., Laurent, B., Guieu, C., Raimbault, P., and Beuvier, J.: Modeling the impacts of atmospheric deposition of nitrogen and desert dust-derived phosphorus on nutrients and biological budgets of the Mediterranean Sea,
  Progress in Oceanography, doi 10.1016/j.pocean.2017.04.009, http://www.sciencedirect.com/science/article/pii/S0079661116301811, 2017.
- 1370 Richon, C., Dutay, J.-C., Dulac, F., Wang, R., and Balkanski, Y.: Modeling the biogeochemical impact of atmospheric phosphate deposition from desert dust and combustion sources to the Mediterranean Sea, Biogeosciences, 15, 2499–2524, doi:10.5194/bg-15-2499-2018, https://www.biogeosciences.net/15/2499/2018/, 2018.
- Robinson, A. R., Leslie, W. G., Theocharis, A., and Lascaratos, A.: Mediterranean Sea Circulation, in:
  1375 Encyclopedia of Ocean Sciences (Second Edition), edited by Steele, J. H., pp. 710–725, Academic
  - Press, Oxford, doi:10.1016/B978-012374473-9.00376-3, http://www.sciencedirect.com/science/article/pii/B9780123744739003763, 2001.
  - Rodellas, V., Garcia-Orellana, J., Masqué, P., Feldman, M., and Weinstein, Y.: Submarine groundwater discharge as a major source of nutrients to the Mediterranean Sea, Proceedings of the National Academy of
- Sciences, 112, 3926–3930, doi:10.1073/pnas.1419049112, http://www.pnas.org/content/112/13/3926, 2015.
   Roether, W., Klein, B., and Hainbucher, D.: The Eastern Mediterranean Transient, in: The Mediterranean Sea, edited by Borzelli, G. L. E., Gačić, M., Lionello, P., and Paolalanotte-Rizzoli, pp. 75–83, John Wiley & Sons, Inc., http://onlinelibrary.wiley.com/doi/10.1002/9781118847572.ch6/summary, 2014.

Rohling, E. J.: Shoaling of the Eastern Mediterranean Pycnocline due to reduction of excess evaporation: Impli-

cations for sapropel formation, Paleoceanography, 6, 747–753, doi:10.1029/91PA02455, http://onlinelibrary.
 wiley.com/doi/10.1029/91PA02455/abstract, 1991.

Rohling, E. J.: Review and new aspects concerning the formation of eastern Mediterranean sapropels, Marine

- Geology, 122, 1–28, doi:10.1016/0025-3227(94)90202-X, http://www.sciencedirect.com/science/article/pii/ 002532279490202X, 1994.
- 1390 Rossignol-Strick, M., Nesteroff, W., Olive, P., and Vergnaud-Grazzini, C.: After the deluge: Mediterranean stagnation and sapropel formation, Nature, 295, 105–110, doi:10.1038/295105a0, http://www.nature.com/nature/journal/v295/n5845/abs/295105a0.html, 1982.
  - Royer, J.-F., Cariolle, D., Chauvin, F., Déqué, M., Douville, H., Hu, R.-M., Planton, S., Rascol, A., Ricard, J.-L., Salas Y Melia, D., Sevault, F., Simon, P., Somot, S., Tyteca, S., Terray, L., and Valcke, S.: Simulation des
- 1395 changements climatiques au cours du XXIe siècle incluant l'ozone stratosphérique, Comptes Rendus Geo
  - science, 334, 147–154, doi:10.1016/S1631-0713(02)01728-5, http://www.sciencedirect.com/science/article/pii/S1631071302017285, 2002.
  - Ruti, P. M., Somot, S., Giorgi, F., Dubois, C., Flaounas, E., Obermann, A., Dell'Aquila, A., Pisacane, G., Harzallah, A., Lombardi, E., Ahrens, B., Akhtar, N., Alias, A., Arsouze, T., Aznar, R., Bastin, S., Bartholy,
- 1400 J., Béranger, K., Beuvier, J., Bouffies-Cloché, S., Brauch, J., Cabos, W., Calmanti, S., Calvet, J.-C., Carillo, A., Conte, D., Coppola, E., Djurdjevic, V., Drobinski, P., Elizalde-Arellano, A., Gaertner, M., Galàn, P., Gallardo, C., Gualdi, S., Goncalves, M., Jorba, O., Jordà, G., L'Heveder, B., Lebeaupin-Brossier, C., Li, L., Liguori, G., Lionello, P., Maciàs, D., Nabat, P., Önol, B., Raikovic, B., Ramage, K., Sevault, F., Sannino, G., Struglia, M. V., Sanna, A., Torma, C., and Vervatis, V.: Med-CORDEX Initiative for Mediterranean

1405 Climate Studies, Bulletin of the American Meteorological Society, 97, 1187-1208, doi:10.1175/BAMS-D-14-00176.1, http://journals.ametsoc.org/doi/10.1175/BAMS-D-14-00176.1, 2016.

Sanchez-Gomez, E., Somot, S., and Mariotti, A.: Future changes in the Mediterranean water budget projected by an ensemble of regional climate models, Geophysical Research Letters, 36, L21401, doi:10.1029/2009GL040120, http://onlinelibrary.wiley.com/doi/10.1029/2009GL040120/abstract, 2009.

- 1410 Santinelli, C., Ibello, V., Lavezza, R., Civitarese, G., and Seritti, A.: New insights into C, N and P stoichiometry in the Mediterranean Sea: The Adriatic Sea case, Continental Shelf Research, 44, 83-93, doi:10.1016/j.csr.2012.02.015, http://www.sciencedirect.com/science/article/pii/S0278434312000519, 2012.
- Schaap, D. M. and Lowry, R. K.: SeaDataNet Pan-European infrastructure for marine and ocean data 1415 management: unified access to distributed data sets, International Journal of Digital Earth, 3, 50-69, doi:10.1080/17538941003660974, http://www.tandfonline.com/doi/abs/10.1080/17538941003660974, 2010.
  - Sevault, F., Somot, S., Alias, A., Dubois, C., Lebeaupin-Brossier, C., Nabat, P., Adloff, F., Déqué, M., and Decharme, B.: A fully coupled Mediterranean regional climate system model: design and evaluation of the
- ocean component for the 1980-2012 period, Tellus A, 66, doi 10.3402/tellusa.v66.23967, http://www.tellusa. 1420 net/index.php/tellusa/article/view/23967, 2014.
  - Somot, S., Sevault, F., and Déqué, M.: Transient climate change scenario simulation of the Mediterranean Sea for the twenty-first century using a high-resolution ocean circulation model, Climate Dynamics, 27, 851-879, doi:10.1007/s00382-006-0167-z, http://link.springer.com/10.1007/s00382-006-0167-z, 2006.
- 1425 Soto-Navarro, J., Somot, S., Sevault, F., Beuvier, J., Criado-Aldeanueva, F., García-Lafuente, J., and Béranger, K.: Evaluation of regional ocean circulation models for the Mediterranean Sea at the Strait of Gibraltar: volume transport and thermohaline properties of the outflow, Climate Dynamics, 44, 1277-1292, doi:10.1007/s00382-014-2179-4, https://link.springer.com/article/10.1007/s00382-014-2179-4, 2015.
- Spillman, C. M., Imberger, J., Hamilton, D. P., Hipsey, M. R., and Romero, J. R.: Modelling the effects of Po 1430 River discharge, internal nutrient cycling and hydrodynamics on biogeochemistry of the Northern Adriatic
  - Sea, Journal of Marine Systems, 68, 167–200, doi:10.1016/j.jmarsys.2006.11.006, http://www.sciencedirect. com/science/article/pii/S0924796306003459, 2007.
  - Takahashi, T., Broecker, W. S., and Langer, S.: Redfield ratio based on chemical data from isopycnal surfaces, Journal of Geophysical Research: Oceans (1978-2012), 90, 6907-6924, doi:10.1029/JC090iC04p06907 http://onlinelibrary.wiley.com/doi/10.1029/JC090iC04p06907/abstract, 1985.
  - Tanaka, T., Thingstad, T. F., Christaki, U., Colombet, J., Cornet-Barthaux, V., Courties, C., Grattepanche, J.-D., Lagaria, A., Nedoma, J., Oriol, L., Psarra, S., Pujo-Pay, M., and Wambeke, F. V.: Lack of P-limitation of phytoplankton and heterotrophic prokaryotes in surface waters of three anticyclonic eddies in the stratified Mediterranean Sea, https://core.ac.uk/display/93249213, 2011.
- 1440 Taylor, K. E., Stouffer, R. J., and Meehl, G. A.: An Overview of CMIP5 and the Experiment Design, Bulletin of the American Meteorological Society, 93, 485-498, doi 10.1175/BAMS-D-11-00094.1, http://journals. ametsoc.org/doi/abs/10.1175/BAMS-D-11-00094.1, 2012.
  - Theocharis, A., Nittis, K., Kontoyiannis, H., Papageorgiou, E., and Balopoulos, E.: Climatic changes in the Aegean Sea influence the eastern Mediterranean thermohaline circulation (1986-1997), Geophysical

1435

- 1445 Research Letters, 26, 1617–1620, doi:10.1029/1999GL900320, http://onlinelibrary.wiley.com/doi/10.1029/ 1999GL900320/abstract, 1999.
  - Thingstad, T. F., Krom, M. D., Mantoura, R. F. C., Flaten, G. a. F., Groom, S., Herut, B., Kress, N., Law, C. S., Pasternak, A., Pitta, P., Psarra, S., Rassoulzadegan, F., Tanaka, T., Tselepides, A., Wassmann, P., Woodward, E. M. S., Riser, C. W., Zodiatis, G., and Zohary, T.: Nature of Phosphorus Limitation in the Ultraoligotrophic
- 1450 Eastern Mediterranean, Science, 309, 1068–1071, doi:10.1126/science.1112632, http://science.sciencemag. org/content/309/5737/1068, 2005.
  - Vadsaria, T., Ramstein, G., Li, L., and Dutay, J.-C.: Sensibilité d'un modèle océan-atmosphère (LMDz-NEMOMED8) à un flux d'eau douce : cas du dernier épisode de sapropèle en mer Méditerranée, Quaternaire, 28, 2017.
- 1455 Velaoras, D. and Lascaratos, A.: North-Central Aegean Sea surface and intermediate water masses and their role in triggering the Eastern Mediterranean Transient, Journal of Marine Systems, 83, 58–66, doi:10.1016/j.jmarsys.2010.07.001, wOS:000282548300005, 2010.
- Wang, R., Balkanski, Y., Boucher, O., Ciais, P., Peñuelas, J., and Tao, S.: Significant contribution of combustion-related emissions to the atmospheric phosphorus budget, Nature Geoscience, 8, 48–54, doi;10.1038/ngeo2324, http://www.nature.com/doifinder/10.1038/ngeo2324, 2014.
  - Yogev, T., Rahav, E., Bar-Zeev, E., Man-Aharonovich, D., Stambler, N., Kress, N., Béjà, O., Mulholland, M. R., Herut, B., and Berman-Frank, I.: Is dinitrogen fixation significant in the Levantine Basin, East Mediterranean Sea?, Environmental Microbiology, 13, 854–871, doi:10.1111/j.1462-2920.2010.02402.x, http://onlinelibrary.wiley.com/doi/10.1111/j.1462-2920.2010.02402.x/abstract, 2011.

heightName	Dynamics (NEMO years)	Buffer zone concentrations	<b>River inputs</b>	N deposition	<b>P</b> deposition
CTRL	1966–1981	1966–1981	1966–1981	No	No
CTRL_R	1966–1981	1966–1981	1980–2099	No	No
CTRL_RG	1966–1981	1980–2099	1980–2099	No	No
HIS/A2	1980–2099	1980–2099	1980–2099	No	No
HIS/A2_N	1980-2099	1980-2099	1980-2099	1997-2012	No
HIS/A2_NALADIN	1980-2099	1980-2099	1980-2099	1997-2012	1980-2012

**Table 1.** Description of the simulations. The years indicate the forcing years throughout the 120 years of simulation. The cycles are repeated in the CTRL simulations.

Simulation	Period	Whole Med.	Western	Eastern	Ionian-Levantine	Adriatic	Aegean	Atlantic buffer zone
	1980-1999	551	241	310	305	1.5	4.0	535
HIS/A2	2030-2049	570 (+3.4)	240 (0)	329 (+6.1)	324 (+6.2)	1.4(0)	3.6 (-10)	543 (+1.5)
	2080–2099	598 (+8.5)	251 (+4.1)	346 (+11.6)	341 (+11.8)	1.5(0)	3.5 (-12.5)	562 (+5.0)
	1980–1999	545	238	307	302	1.6	4.0	532
CTRL	2030–2049	553 (+1.5)	238 (0)	314 (+2.3)	309 (+2.3)	1.6(0)	4.2 (+5.0)	532 (0)
	2080-2099	560 (+2.6)	241 (+1.3)	319 (+3.9)	313 (+3.6)	1.7 (+6.3)	4.2 (+5.0)	532 (0)
	1980–1999	547	239	309	303	1.5	4.2	534
CTRL_R	2030-2049	536 (-2.0)	232 (-2.9)	304 (-1.6)	299 (-1.3)	1.4 (-6.7)	3.5 (-17)	534 (0)
	2080-2099	538 (-1.6)	230 (-3.8)	309 (0)	303 (0)	1.5 (0)	3.7 (-12)	534 (0)
	1980–1999	548	239	309	303	1.5	4.2	535
CTRL_RG	2030-2049	536 (-2.2)	233 (-2.5)	303 (-1.9)	298 (-1.7)	1.4 (-6.7)	3.5 (-17)	544 (+1.7)
	2080–2099	540 (-1.5)	235 (-1.7)	306 (-1.0)	301 (-0.7)	1.4 (-6.7)	3.6 (-14)	562 (+5.0)

Table 2. Simulated integrated phosphate content (10<sup>9</sup> mol) over 20. years 20. years periods in the Mediterranean sub-basins in the different simulations. Basins are the same as defined in Fig.2 of Adloff et al. (2015), with the eastern basin including the Ionian, Levantine, Adriatic and Aegean subbasins. Values in parenthesis indicate the percentage difference from the 1980-1999 period.

Simulation	Period	Whole Med.	Western	Eastern	Ionian-Levantine	Adriatic	Aegean	Atlantic buffer zone
	1980–1999	13400	5520	7890	7690	6.99	132	8091
HIS/A2	2030-2049	13800 (+3.0)	5450 (-1.3)	8350 (+5.8)	8100 (+5.3)	88.5 (+32)	163 (+23)	8230 (+1.7)
	2080–2099	14700 (+9.7)	5750 (+4.2)	8920 (+13)	8650 (+12)	98.3 (+47)	164 (+24)	8510 (+5.2)
	1980–1999	13500	5530	7970	7760	66.2	144	8050
CTRL	2030-2049	12900 (-4.4)	5320 (-3.8)	7610 (-4.5)	7420 (-4.4)	61.7 (-6.8)	131 (-9.0)	8050 (0)
	2080-2099	12500 (-7.4)	5170 (-6.5)	7330 (-8.0)	7150 (-7.9)	58.7 (-11)	123 (-15)	8050 (0)
	1980–1999	13300	5470	7870	7760	66.8	138	8070
CTRL_R	2030-2049	13300(0)	5250 (-4.0)	8020 (+1.9)	7770 (+0.1)	88.0 (+32)	162 (+17)	8070 (0)
	2080-2099	13700 (+3.0)	5300 (-3.1)	8440 (+7.2)	8170 (+5.3)	94.2 (+41)	177 (+28)	8080 (+0.1)
	1980–1999	13300	5480	7870	7760	66.8	138	8090
CTRL_RG	2030–2049	13300(0)	5270 (-3.8)	8010 (+1.8)	7760 (0)	88.0 (+32)	162 (+17)	8080 (-0.1)
	2080–2099	13800 (+3.8)	5390 (-1.6)	8430 (+7.1)	8160 (+5.2)	94.3 (+41)	177 (+28)	8080 (-0.1)

Table 3. Simulated integrated nitrate content (10<sup>9</sup> mol) over 20 years 20. years periods in the Mediterranean sub-basins in the different simulations. Basins are the same as defined in Fig.2 of Adloff et al. (2015), with the eastern basin including the Ionian, Levantine, Adriatic and Aegean subbasins. Values in parenthesis indicate the percentage difference from the 1980-1999 period.

Simulation	Period	Whole Med.	Western	Eastern	Ionian-Levantine	Adriatic	Aegean	Atlantic buffer zone
	1980-1999	25700	9680	16000	13500	830	1720	3210
HIS/A2	2030-2049	23800 (-7.4)	8980 (-7.2)	14800 (-7.5)	12700 (-5.9)	720 (-13)	1440 (-16)	3280 (+2.2)
	2080–2099	23400 (-8.9)	9180 (-5.1)	14300 (-11)	12200 (-9.6)	690 (-17)	1390 (-19)	3570 (+11)
	1980-1999	27000	10200	16700	14200	880	1670	3180
CTRL	2030-2049	27000 (0)	10200 (0)	16900 (+1.2)	14300 (+0.7)	890 (+1.1)	1710 (+2.4)	3180 (0)
	2080-2099	26600 (-1.5)	9980 (-2.2)	16600 (-0.1)	14000 (-1.4)	880 (0)	1690 (+1.2)	3180(0)
	1980–1999	27000	10300	16700	14100	875	1720	3210
CTRL_R	2030-2049	26800 (-0.7)	10100 (-1.9)	16700 (0)	14300 (+1.4)	780 (-12)	1610 (-6.4)	3210 (0)
	2080-2099	26400 (-2.2)	9940 (-3.5)	16500 (-1.2)	14100(0)	760 (-13)	1600 (-7.0)	3220 (0.3)
	1980–1999	27000	10300	16700	14100	875	1720	3230
CTRL_RG	2030-2049	26900 (-0.4)	10200 (-1.0)	16700(0)	14300 (+1.4)	780 (-12)	1600 (-7.0)	3260 (+0.9)
	2080–2099	26700 (-1.1)	10200 (-1.0)	16500 (-1.2)	14100(0)	750 (-14)	1600 (-7.0)	3420 (+5.9)

Table 4. Simulated integrated chlorophyll production chl-a (10<sup>9</sup> mol) over 20 years 20 years 20 years periods in the Mediterranean sub-basins in the different simulations. Basins are the same as defined in Fig.2 of Adloff et al. (2015), with the eastern basin including the Ionian, Levantine, Adriatic and Aegean subbasins. Values in parenthesis indicate the percentage difference from the 1980-1999 period.



**Figure 1.** Average surface <u>chlorophyll\_chl-a</u> concentration from the CTRL (top, left: 1980–1999 right: 2080–2099) and HIS/A2 (bottom left) simulations, and from satellite estimations (MyOcean Dataset 1997–2012, bottom right).



**Figure 2.** Average surface <u>chlorophyll chl-a</u> concentration from the HIS/A2 simulation in blue and from the SeaWiFs satellite data (Bosc et al.) [2004) in red over the period 1997–2005. Shaded colors represent the standard deviations. Values are normalized by dividing by the average cholorphyll concentration over the period.



**Figure 3.** Evolution of average Mediterranean SST (left) and SSS (right) in the CTRL (greyline) and HIS/A2 (blueline) simulations.



(c) Total outgoing nitrate



**Figure 4.** Evolution of total incoming (top) and outgoing (bottom) fluxes of nitrate and phosphate  $(10^9 \text{ mol month}^{-1})$  through the Strait of Gibraltar in the CTRL (greyline) and HIS/A2 (redline) simulations. Negative values indicate outgoing fluxes of nutrients.



(a) Total river discharge of nitrate

(b) Total river discharge of phosphate

**Figure 5.** Evolution of total river discharge fluxes of nitrate and phosphate  $(10^9 \text{ mol month}^{-1})$  to the Mediterranean Sea in the CTRL (greyline) and HIS/A2 (redline) simulations.



(a) Total nitrogen sedimentation

(b) Total phosphorus sedimentation

**Figure 6.** Evolution of total sedimentation fluxes of N and P ( $10^9$  mol month<sup>-1</sup>) in the Mediterranean Sea in the CTRL (greyline) and HIS/A2 (redline) <u>simulations</u>. Negative fluxes indicate that the nutrients are exiting the Mediterranean waters.



**Figure 7.** Evolution of yearly average phosphate concentration  $(10^{-3} \text{ mol m}^{-3})$  in the surface (left), intermediate (middle) and bottom (right) layers in the western (top) and eastern (bottom) basin. Red lines represent the HIS/A2 simulation, black lines represent the CTRL (with standard deviation), blue and light blue lines represent the CTRL\_R and CTRL\_RG simulations respectively. Colored numbers in the highlighted areas represent the average concentrations in the corresponding simulations for the highlighted time periods.


**Figure 8.** Evolution of yearly average nitrate concentration  $(10^{-3} \text{ mol m}^{-3})$  in the surface (left), intermediate (middle) and bottom (right) layers in the western (top) and eastern (bottom) basins. Red lines represent the HIS/A2 simulation, black lines represent the CTRL (with standard deviation), blue and light blue lines represent the CTRL\_R and CTRL\_RG simulations respectively. Colored numbers in the highlighted areas represent the average concentrations in the corresponding simulations for the highlighted time periods.



**Figure 9.** Present (1980–1999, top) interannual average surface (0–200 m) concentrations of nitrate  $(10^{-3} \text{ mol m}^{-3})$  in the CTRL (left) HIS/A2 (right) simulations. The bottom maps show the <u>percent</u> relative difference (in %) between the 2080–2099 and the 1980–1999 periods in the CTRL (left) and HIS/A2 (right) simulations.



**Figure 10.** Present (1980–1999, top) interannual average surface (0–200m) concentrations of phosphate  $(10^{-3} \text{ mol m}^{-3})$  in the CTRL (left) and HIS/A2 (right) simulations. The bottom maps show the <u>percent</u> relative difference (in %) in-primary production between the 2080–2099 and the 1980–1999 periods in the CTRL (left) and HIS/A2 (right) <u>simulations</u>.



**Figure 11.** Present (1980–1999, top) interannual average surface (0–200 m) integrated primary production (gC m<sup>-2</sup>) in the CTRL (left) and HIS/A2 (right) simulations. The bottom maps show the <u>percent</u> relative difference (in %) between the 2080–2099 and the 1980–1999 periods in the CTRL (left) and HIS/A2 (right) simulations.



**Figure 12.** Present (1980–1999) and future (2080-2099) interannual average surface (0–200 m) limiting nutrient in the HIS/A2 simulation. N and P colimitation is considered when limitation factors for N and P differ by less than 1 %. Green zones are P–limited, Orange zones are N–Limited–N–limited and purple zones are N and P co–limited.



**Figure 13.** Present (1980–1999, top) interannual average DCM (m) in the CTRL (left) and HIS/A2 (right) simulations. The bottom maps show the <u>percent</u> relative difference (in %) in DCM between the 2080–2099 and the 1980–1999 periods in the CTRL (left) and HIS/A2 (right) simulations.



**Figure 14.** Present (1980–1999) and future (2080–2099) interannual average vertical profiles of total <u>ehlorophyll\_chl-a</u> a (<del>10e<sup>-9</sup>gng</del> L<sup>-1</sup>) at the DYFAMED station and averaged profiles over the western and eastern (including Aegean and Adriatic) basins.</del>



(c) Diatoms western basin



**Figure 15.** Evolution of yearly average nanophytoplankton and diatoms concentration  $(10^{-3} \text{ mol m}^{-3})$  in the surface layer of the western and eastern basin. Red\_The red line represent the HIS/A2 simulation, the black lines represent the CTRL simulation (with standard deviation), blue and light blue lines represent the CTRL\_R and CTRL\_RG simulations respectively. Colored numbers in the highlighted areas represent the average concentrations in the corresponding simulations for the highlighted time periods.



(c) Mesozooplankton western basin



Figure 16. Evolution of yearly average microzooplankton and mesozooplankton concentrations  $(10^{-3} \text{ mol m}^{-3})$  in the surface layer of the western and eastern basins. Red lines represent the HIS/A2 simulation, black lines represent the CTRL <u>simulation</u> (with standard deviation), blue and light blue lines represent the CTRL\_RG simulations respectively. Colored numbers in the highlighted areas represent the average concentrations in the corresponding simulations for the corresponding time periods.



(a) Relative effects of total nitrogen deposition



(b) Relative effects of natural dust deposition

**Figure 17.** Present (1980–1999) and future (2080–2099) relative effects of total nitrogen (top) and natural dust (bottom) deposition on surface (0–10 m) total primary production.



(a) Phosphate fluxes



## (b) Nitrate fluxes

**Figure 18.** Schematic diagrams illustrating the Mediterranean budgets of phosphate and nitrate. For each component, the 3 lines represent the average fluxes (in Gmol year<sup>-1</sup>) over the periods 1980–1999, 2030-2049 and 2080-2099, numbers in parenthesis indicate the percentage difference from the 1980–1999 values.



**Figure A1.** Average <u>chlorophyll-a chl-a-a</u> profiles in February (left) and May (right) for the years 1991 to 2005 at the DYFAMED station in the Ligurian Sea <u>Sea</u>-(43.4277°N, 7.2522°E). Dots represent data points (<u>Marty</u> et al., 2002; Faugeras et al., 2003). Lines represent the HIS/A2 simulation. Colors represent individual years.



**Figure A2.** Average phosphate (left) and nitrate (right) profiles in May for the years 1991 to 2005 at the DY-FAMED station in the Ligurian Sea Sea (43.4277°N, 7.2522°E). Dots represent data points (Marty et al.) 2002; Faugeras et al.) (2003). Lines represent the HIS/A2 simulation. Colors represent individual years.



(a) nitrate concentration



## (b) PO<sub>4</sub> concentration

**Figure A3.** Average concentrations of nitrate (top) and phosphate (bottom) for the 20 first years of the control simulation (CTRL). The dots represent data along a transect from Marseille to Finokalia from the BOUM campaign (distances in km Moutin et al., 2012). The framed areas represent a vertical zoom of the top 500 m along the whole transect.