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Phytoplanktonic response to contrasted Saharan dust deposition events during mesocosm experiments in LNLC environment

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Abstract

The response of the phytoplanktonic community (primary production and algal biomass) to contrasted Saharan dust events (wet and dry deposition) was studied in the framework of the DUNE "a DUst experiment in a low-Nutrient, low-chlorophyll Ecosystem" project. We simulated realistic dust deposition events (10 g m^{-2}) into large mesocosms (52 m^3). Three distinct experimental dust additions were conducted in June 2008 (DUNE-1-P: simulation of a wet deposition, DUNE-1-Q: simulation of a dry deposition) and 2010 (DUNE-2-R1, -R2: simulation of 2 successive wet depositions) in the northwestern oligotrophic Mediterranean Sea. No changes in primary production (PP) and chlorophyll *a* concentration (Chl *a*) were observed after a dry deposition event while a wet deposition event resulted in a rapid (24 h after dust additions), strong (up 2.4 fold) and long (at least a week duration) increase in PP and Chl *a*. We show that in addition to being a source of dissolved inorganic phosphorus (DIP), simulated wet deposition events were also a significant source of NO_3^- (net increases up to +9.8 µM NO_3^- at 0.1 m depth) to the nutrient depleted surface waters due to cloud processes and mixing with anthropogenic species such as HNO3. The dry deposition event was shown to be a negligible source of NO_3^-. By transiently increasing DIP and NO_3^- concentrations in P-N starved surface waters, wet deposition of Saharan dust was able to relieve the potential N or NP co-limitation of the phytoplanktonic activity. Due to the higher input of NO_3^- relative to DIP, a wet deposition event resulted in a strong increase in the NO_3^-/DIP ratio from initially < 6 to over 150 at the end of the DUNE-2-R1 experiment suggesting a switch from an initial N or NP co-limitation towards a severe P limitation. We also show that the contribution of new production to PP increased after wet dust deposition events from initially 15% to 60–70% 24 h after seeding, indicating a switch from a regenerated-production based system to a new-production based system. DUNE experiments show that wet and dry dust deposition events induce contrasted responses of the phytoplanktonic community due to differences in the atmospheric supply of bioavailable new nutrients. Our results from original mesocosm

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experiments demonstrate that atmospheric dust wet deposition greatly influences primary productivity and algal biomass in LNLC environments, changes nutrient stocks and alters the NO_3^-/DIP ratio leading to a switch in the nutrient limitation of the phytoplanktonic activity.

1 Introduction

The primary nutrients that limit marine phytoplankton productivity include nitrogen (N), phosphorus (P) and iron (Fe). Mineral dust deposition from arid regions provides a source for each of these nutrients to the open ocean surface waters and can have important effects on marine biogeochemical cycles and potentially affect the global climate (e.g., Baker et al., 2003; Jickells et al., 2005; Mahowald et al., 2008). Researchers have suggested that Fe delivered by dust could have stimulated oceanic productivity and may help explain the glacial-interglacial atmospheric pCO_2 transition (Martin, 1990; Falkowski, 1997; Ridgwell and Watson, 2002; Mahowald et al., 2006). While the impact of Fe on productivity has been recognized in high nutrient low chlorophyll (HNLC) oceanic areas (Boyd et al., 2000, 2007; de Baar et al., 2005; Blain et al., 2007), the ecological and biogeochemical effects of aeolian dust deposition of macro- (N, P) and micro-nutrients (Fe) in oligotrophic low nutrient low chlorophyll (LNLC) environments remain less explored and still poorly understood although LNLC regions represent 60 % of the global surface ocean (Longhurst et al., 1995) and over 50 % of the global oceanic carbon export (Emerson et al., 1997). The mineral dust can be transported thousands of miles across the Atlantic and Pacific oceans and deliver large quantities of nutrients to the LNLC Atlantic and Pacific gyres which in turn could play a significant role in stimulating primary production (Bishop et al., 2002; Johnson et al., 2003; Duarte et al., 2006; Maranon et al., 2010; Franchy et al., 2013) potentially increasing the efficiency of the biological pump in the sequestration of atmospheric CO_2.

The Mediterranean Sea is a typical LNLC region particularly well adapted to tackle the question of the planktonic response to atmospheric nutrient inputs. The Mediter-

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ranean Sea is an oligotrophic quasi-enclosed basin which receives a noticeable flux of dust, mainly derived from Sahara Desert, in the form of strong pulses (Guerzoni et al., 1999; Guieu et al., 2010a). After the seasonal phytoplanktonic bloom during spring, the surface mixed layer is isolated from deeper waters by a strong thermal stratification, and becomes N and P depleted (Marty et al., 2002; Pulido-Villena et al., 2010) leading to low primary productivity and phytoplanktonic biomass (Moutin and Raimbault, 2002; Marty and Chiaverini, 2002; Bosc et al., 2004; Lopez-Sandoval et al., 2011). During the whole stratification period, atmospheric inputs are therefore the main source of allochtonous nutrients to the oligotrophic surface waters. By bringing new macro-nutrients (e.g. Herut et al., 1999; Ridame and Guieu, 2002; Pulido-Villena et al., 2010; Markaki et al., 2010) and Fe (e.g. Bonnet and Guieu, 2006; Wuttig et al., 2013) to the Mediterranean surface waters, Saharan dust deposition is strongly suspected to play a key role in the control of primary production. During the BOUM (Biogeochemistry from the Oligotrophic to the Ultra-Oligotrophic Mediterranean) cruise in the summer of 2008, nutrient/dust additions in microcoms showed that primary production and chlorophyll *a* were N or NP co-limited in the western Mediterranean Sea (Tanaka et al., 2011) and that Saharan dust stimulated primary production (Ternon et al., 2011).

The goal of the DUNE project was to estimate the impact of Saharan dust events on a whole LNLC ecosystem like the Mediterranean Sea from virus to zooplankton over a period of one week and to evaluate the biogeochemical implications associated with this forcing (Guieu et al., 2010b, 2013a). The approach applied in DUNE was, for the first time, to perform realistic dust seedings onto large metal-free mesocosms. In this context, the present paper is focused on the response of the whole phytoplanktonic community to contrasted Saharan dust events (wet and dry deposition). Here we quantify the modifications of in situ concentrations of dissolved inorganic nitrogen as well as the changes in the primary production and algal biomass (chlorophyll *a*) after simulated dust deposition events. This study is complementary to the companion paper of Giovanetti et al. (2013) focusing on changes in the structure and composition of the

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phytoplanktonic community as well as in the ecophysiological state of the phytoplanktonic cells after two successive wet deposition events simulated during DUNE-2-R.

2 Material and methods

2.1 Experimental design and dust characterization

Three distinct experimental dust additions into large mesocosms were conducted in June 2008 and June–July 2010 in the northwestern Mediterranean Sea, in the frame of the DUNE ("a DUst experiment in a low-Nutrient, low-chlorophyll Ecosystem") project (http://www.obs-vlfr.fr/LOV/DUNE/index.html). More precisely, the experiments were realized in the Elbo Bay located in the Natural Preservation Area of Scandola (8.554° E, 42.374° N) which is representative of the LNLC conditions of the open western Mediterranean Sea (Guieu et al., 2010b, 2013a). The mesocosms experiment design and the accuracy of the strategy are fully described in Guieu et al. (2010b). Briefly, six mesocosms (height: 12.5 m, diameter: 2.3 m, surface area: 4.15 m^2, and volume: 52 m^3) entirely designed in plastic were deployed. The bags were made of polyethylene mixed with vinyl acetate and the holding structure of PVC and polyethylene. The anchors were screwed in the sea floor at 25–30 m depth. The mesocosms are closed systems, which prevent lateral advection and allow process studies in the vertical dimension over the time. Sediment traps (a 250 mL HDPE bottle) were screwed to the base of the mesocosms for the collection of the exported material and were changed every day (DUNE-2-R) or every two days (DUNE-1-P, -Q). Three replicate mesocosms (D1, D2 and D3 hereafter referred to as "Dust-meso") were amended with 41.5 g of mineral dust reproducing a high, but environmentally realistic atmospheric dust deposition of 10 g m^{-2} (Guieu et al., 2010b). The dust used in the three seeding experiments resulted from the fine fraction (< 20 µm) of Saharan soils collected in southern Tunisia, which is a Saharan aerosol source area (details in Guieu et al., 2010b; Desboeufs et al., 2013). Three other mesocosms served as controls (C1, C2 and C3 hereafter referred

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to as "Control-meso"). The sampling session took place every morning at the same time over the duration of the experiments. Each day, three different depths (0.1, 5 and 10 m depth) were sampled in the six mesocosms using a trace metal clean system of permanent PVC tubing placed at the center of the bags and connected to a Teflon pump. Additional depths (2.5 and 12.5 m) were sampled in two dust mesocosms during DUNE-2-R for nutrient determination. Every 48 h, seawater was also collected outside the mesocosms (hereafter referred to as "out") at the same depths in order to test the representativeness of data between the Control-meso and out.

June 2008 experiments: Two distinct seeding experiments were conducted with two types of dust. In the first one (10–18 June 2008 hereafter referred to as "DUNE-1-P"), the dust used was previously subjected to physico-chemical transformations through condensation/evaporation cycles that involved HNO_3 and H_2SO_4, mimicking atmospheric transport (Desboeufs et al., 2001; and details in Guieu et al., 2010b). Indeed, Saharan dust collected in the Mediterranean atmosphere is usually mixed with organic and inorganic material, as sulfate and nitrate due to cloud processing during atmospheric transport (e.g. Puteaud et al., 2004). This evapocondensed dust (hereafter referred to as "EC dust") contained on average 0.045 ± 0.015 % of P, 2.31 ± 0.04 % of Fe and 1.19 ± 0.05 % of N, in weight (Table 1). The amendment of the dust mesocosms in the DUNE-1-P experiment was performed with the EC dust mixed with 2 L of ultrapure water in order to mimic a wet deposition event.

In the second experiment (20–27 June 2008 hereafter referred to as "DUNE-1-Q"), non-processed dust was used (hereafter referred to as "non-EC dust") and contained on average 0.044 ± 0.009 % of P, 2.28 ± 0.19 % of Fe and 0.11 ± 0.01 % of N (Table 1). The amendment of the dust mesocosms in the DUNE-1-Q experiment was conducted with the non-EC dust mixed with 2 L of unfiltered surface seawater in order to mimic a dry deposition event.

June–July 2010 experiments: Two successive seeding experiments (26 June–2 July 2010 hereafter referred to as "DUNE-2-R1" and 3–9 July 2010 hereafter referred to as "DUNE-2-R2", see details in Guieu et al., 2013a) were performed with the same

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amount of EC dust and with the same deposition setup than in DUNE-1-P simulating a wet deposition event. The dust was characterized by an average content of 0.055 ± 0.003 % P, 2.26 ± 0.03 % Fe and 1.36 ± 0.09 % N (Table 1; see details in Desboeufs et al., 2013).

P, Fe, N contents of the dust: The particulate P and Fe contents of the EC dust (DUNE-1-P and DUNE-2-R) and non-EC dust (DUNE-1-Q) were similar ($p > 0.05$, Table 1). Due to the simulation of cloud water processes that involved HNO_3, the N content of EC dust was about 10 fold higher in comparison with the non-EC dust (Table 1; Guieu et al., 2010b, 2013a). Small differences of the N content in EC dust used in 2008 and 2010 experiments (1.19 ± 0.05 % and 1.36 ± 0.09 %) were observed (details in Guieu et al., 2013a).

2.2 Primary production

All materials were previously acid-washed (HCl Suprapur) following trace metal clean procedures. Before sampling, bottles were rinsed three times with the sampled seawater. One sample per depth of unfiltered seawater was collected in the morning at two depths (0.1 and 5 m depth) during DUNE-1-P and -Q and at 5 m depth during DUNE-2-R for determination of primary production (PP). Samples were collected in the six mesocosms and outside the mesocosms before and after dust seeding. During DUNE-1-P and -Q, 2.5 mL of $NaH^{13}CO_3$ (99 %, Eurisotop) were added to 4.5 L polycarbonate bottles for ^{13}C uptake determination while during DUNE-2-R, 1.5 mL of $NaH^{13}CO_3$ were added to 2.3 L polycarbonate bottles ($^{15}N_2$ tracer was also added in order to determine simultaneously N_2 fixation rates using the dual $^{13}C/^{15}N_2$ isotopic label technique, see Ridame et al., 2013). Prior to DUNE-2-R, intercomparison of PP rates measured in both 2.3 and 4.5 L incubated volumes showed coefficients of variation (CV) lower than 15 % (unpublished data) which was in the same order of magnitude than those found between triplicate mesocosms (CV < 20 %). Immediately after sampling, ^{13}C tracer was added (Fig. 1a) to obtain a final enrichment of about

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9 atom% excess and each bottle was well shaken. Then, the ^{13}C-amended bottles were incubated under in situ conditions on a mooring line, outside the mesocosms for 24 h at the corresponding sampling depths (0.1 m and 5 m depth, Fig. 1b). Incubations were terminated the following morning by filtration onto pre-combusted 25 mm GF/F filters (0.7 µm nominal porosity). Sample filters were stored at −20 °C and dried at 40 °C for 48 h before analysis. Concentrations of carbon and nitrogen in particulate matter and ^{13}C-enrichment were quantified with an isotope ratio mass spectrometer (IRMS, Delta plus, ThermoFisher Scientific, Bremen, Germany) coupled with a C/N analyzer (Flash EA, ThermoFisher Scientific) via a type III interface. Standard deviation was 0.009 µmol L^{-1} and 0.004 µmol L^{-1} for particulate carbon and nitrogen respectively, and 0.0002 atom% for ^{13}C enrichment. From these measurements, the C : N molar ratios in the particulate matter were calculated.

For DUNE-1-P and-Q, PP measured at 0.1 and 5 m depth were integrated over the mesocosm depth (12.5 m). We assume that PP at 5 m depth was similar to that at 12.5 m depth based on the similarity of the chlorophyll *a* concentrations (Chl *a*) measured at 5 and 10 m depth in the Control- and Dust-meso over the experiments ($p > 0.05$). For DUNE-2-R, PP measured at 5 m depth was integrated over the water column by assuming a homogenous distribution. Over DUNE-2-R1, Chl *a* was similar between the three sampling depths ($p > 0.05$). During DUNE-2-R2, Chl *a* at 5 and 10 m depth was similar ($p > 0.05$) while Chl *a* in surface waters was slightly higher (+ ∼ 30 %, $p < 0.05$) than deeper at 3 sampling times.

2.3 Estimation of new production

The increase in PP in the Dust-meso 24 h after seeding can be associated with new production ($NP_{seeding}$) as atmospheric deposition represents a source of external nutrients to the surface layer. We consider that after 24 h, the increase in PP could be partly supported by regenerated nutrients. NP was estimated before seeding and over the course of the experiments in the Control-meso considering that NP represents 15 %

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of PP during periods of stratification in the western Mediterranean Sea (Marty et al., 2002; Moutin and Raimbaut, 2002; L'Helguen et al., 2002). We estimated the NP in the Dust-meso (NP_{dust}) 24 h after seeding using the following equations (from Ridame et al., 2013):

$$NP_{dust} = NP_{control} + NP_{seeding}$$

$$NP_{seeding} = PP_{dust} - PP_{control}$$

2.4 Chlorophyll *a*

2 L of seawater from 0.1, 5 and 10 m depth were collected at daily scale and then filtered onto 25 mm GF/F filters. After 24 h extraction in 90 % acetone, at 4 °C and in the dark, the fluorescence of Chl *a* was measured on a Turner Trilogy Laboratory fluorometer (Strickland and Parsons, 1997). The Chl *a* concentrations measured at the three sampling depths were integrated over the mesocosm depth (12.5 m).

2.5 Nutrient concentrations

20 mL of filtered seawater (< 0.2 µm, Sartobran cartridge filters) from 0.1, 5 and 10 m depth were collected in acid-washed HDPE (high density polyethylene) bottles and frozen until analysis. Nitrate (NO_3^-) and nitrite (NO_2^-) were analyzed according to classical methods using the automated colorimetric technique as described in Grasshoff et al. (1999), on a QuAAtro Continuous Flow Analyzer (SEAL Analytical). The precision of measurements was ±30 nM and ±40 nM for NO_3^- and NO_2^-, respectively. The limits of detection, defined as three times the standard deviation of the blank were 30 nM and 10 nM for NO_3^- and NO_2^- respectively.

Dissolved inorganic phosphorus (DIP) concentrations presented in Pulido-Villena et al. (2010, 2014) were analyzed immediately after collection on 0.2 µm-filtered seawater by spectrophotometry using a long waveguide capillary cell (LWCC); the detection limit was 2 nM (details in Pulido-Villena et al., 2010, 2014). The concentration

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of dissolved Fe (DFe) (< 0.2 µm) was measured by flow injection analysis with online preconcentration and chemiluminescence detection (FIA-CL); the detection limit was 10 p.m. (details in Wagener et al., 2010). These data are presented in Wagener et al. (2010) and Wuttig et al. (2013).

2.6 Dissolution experiments under abiotic conditions

Both types of Saharan dust (EC and non-EC dust) used during the DUNE-1-P and -Q experiments were studied to quantify under abiotic conditions the amount of inorganic nitrogen (nitrate + nitrite: NO_x; ammonium: NH_4^+) potentially released in the Dust-meso after seeding. This work was complementary to the *in vitro* dissolution experiments performed for the quantification of DFe (Wagener et al., 2010) and DIP (Pulido-Villena et al., 2010) released from dust. All experiments were performed in a clean room under a laminar flow hood using trace metal clean techniques and all materials were previously acid-washed (5 % HCl).

Dissolution experiments were conducted in sterile artificial seawater (Chen et al., 1996). For each type of dust, a stock solution of 20 mg dust L^{-1} was made in artificial seawater and quickly (less than 2 min to minimize the instantaneous dissolution processes) after homogenization, increasing amounts of this stock solution were added to artificial seawater to reach a range of nine concentrations from 0 to 20 mg dust L^{-1}: 0, 0.01, 0.1, 0.5, 1, 3, 5, 10 and 20 mg dust L^{-1}. Each condition was performed in triplicate in 250 mL Nalgene® polycarbonate bottles. After dust inoculation, bottles were placed on an agitation table in the dark for 3 and 24 h. At each time point, 125 mL of each sample were filtered on washed (0.5 % HCl and rinsed three times with ultrapure water) Nucleopore® polycarbonate filters (0.2 µm porosity) under a 200 mbar vacuum. The filtration was performed to remove Saharan dust from liquid phase and to stop dissolution processes. The filtrate from each sample was subsampled to measure the concentration of NO_x and NH_4^+. Samples for NO_x were stored in Nalgene® HDPE vials in the dark at 4 °C for 2 days and NH_4^+ samples were immediately anal-

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ysed. Briefly, NO_x concentration was analysed with an AutoAnalyzer3 Digital Colorimeter (Bran Luebbe) according to the method described by Aminot and Kérouel (2007) (detection limit of 20 nmol L^{-1}). For NH_4^+ concentration, 40 mL of filtrate were recovered in a Schott flask (Duran) and analysed by the method described in Holmes et al. (1999) with a Turner Trilogy Laboratory fluorometer (detection limit of 11 nmol L^{-1}). Due to salt contamination, the initial concentrations of NO_x and NH_4^+ in the artificial seawater were 0.19 ± 0.02 µmol L^{-1} ($n = 12$) and 1.46 ± 0.12 µmol L^{-1} ($n = 7$), respectively.

2.7 Statistical analysis

Means of Chl *a*, PP, and NO_3^- concentrations in the Dust- and Control-meso as well as the NO_x concentrations in the dissolution experiments were compared using a repeated measure ANOVA and a Fisher Least Significant Difference (LSD) means comparison test ($\alpha = 0.05$). When assumptions for ANOVA were not respected, means were compared using a Kruskal–Wallis test and a post hoc Dunn's test in XLstat software.

3 Results

3.1 Characteristics of the seawater before seeding and evolution of the environmental conditions during the experiments

Statistical analysis of biological and chemical parameters in Table 2 showed no significant difference between Control-, Dust-meso and out before seeding for all experiments (Guieu et al., 2010b, 2013a). Chl *a* (0.07–0.11 µg L^{-1}) and PP (3.89–5.35 mg C m^{-3} d^{-1}) were initially low, and were slightly higher in DUNE-1-P relative to DUNE-1-Q and DUNE-2-R ($p < 0.05$, Table 2). The picophytoplanktonic biomass (Chl *a* < 3 µm) accounted for about 70 % of the total phytoplanktonic biomass before the Q and R seedings (C. Brunet, personal communication, 2013; Giovagnetti et al., 2013) (no data for DUNE-1-P). *Synechococcus* was the most abundant phytoplanktonic organism in the

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< 3 µm size fraction before all the dust additions (unpublished data; Giovagnetti et al., 2013). The initial C : N molar ratios in the particulate fraction, ranging from 7.3 to 7.8 were slightly higher than the Redfield ratio of 6.6 (106/16). The water column was initially DIP depleted (2–5 nM) with concentrations lower in the DUNE-1-Q experiment (2 nM) than in DUNE-1-P and DUNE-2-R (5 nM; $p < 0.05$, Pulido-Villena et al., 2010, 2014). The initial NO_3^- and NO_2^- concentrations were below detection limit (< 30 nM) in the DUNE-R experiment. By assuming a maximum initial NO_3^- concentration of 30 nM in DUNE-2-R, it should lead to a maximum NO_3^-/DIP ratio of 6. Due to analytical problems, NO_3^- concentrations were not available for P and Q experiments but are strongly suspected to be below detection limit before seeding as shown in experiment R and in surface waters of the northwestern Mediterranean Sea during stratification (Marty et al., 2002; Pujo-Pay et al., 2011). The initial DFe concentration was higher for DUNE-2-R (3.3 nM) relative to DUNE-1-P and -Q (~ 2.3 nM, $p < 0.05$, Table 2).

The initial seawater temperatures over the water column were warm (> 19 °C, Table 2). During DUNE-1-P, the temperature was lower than during the other ones (Guieu et al., 2013a). Over the duration of this experiment, the temperature in the water column of the mesocosms was homogenous and stable (mean $T = 19.8 \pm 0.5$ °C, Guieu et al., 2010b) while over the course of DUNE-1-Q, temperature increased rapidly (up to 26.0 °C at the surface) leading to a strongly marked thermal stratification typical of summer conditions (mean $\Delta T_{0-10\,m} = 3.6$ °C between 21 and 27 June, Guieu et al., 2013a). Over DUNE-2-R1, changes in temperature were representative of the transition period between spring (low stratification) and summer (strong stratification) conditions. While at the beginning of the second seeding (DUNE-2-R2) stratification was well established, a destratification followed by a restratification was then observed (details in Guieu et al., 2013a). The highest temperature was recorded during DUNE-2-R2 (up to 27.3 °C at the surface). Photosynthetically available radiation (PAR) was measured during DUNE-2-R (Guieu et al., 2013a; Giovagnetti et al., 2013). Over the 13 days of the experiment, PAR was high with maximum values of ~ 900 µmol photons $m^{-2}\,s^{-1}$ at the subsurface. During DUNE-1-P, the percentage of sunshine duration over the whole

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day light was lower than during DUNE-1-Q where the values were close to the maximum (Guieu et al., 2013a). So, it is likely that PAR values increased between P and Q experiments.

After the DUNE-2-R1 and -R2 seedings, strong increases in NO_3^- concentrations at the surface of the Dust-meso were observed relative to the Control-meso where NO_3^- concentrations were lower than 30 nM (Fig. 2a). Ten hours after the first seeding, NO_3^- concentrations in surface waters reached 3.3 ± 0.3 µM and then decreased to 0.5 ± 0.1 µM at the end of R1 (day 6). Five hours after the second seeding, an additional increase in NO_3^- concentrations (+9.8 µM relative to day 6 in the Dust-meso) was detected at the surface leading to NO_3^- concentrations of 10.3 ± 2.4 µM. At the end of R2, NO_3^- concentrations in the Dust-meso decreased up to detection limit. Lower increases in NO_3^- concentrations were recorded at 5 and 10 m depth in the Dust-meso (maximum of ~ 1 µM) (Fig. S1). NO_3^- and NO_2^- concentrations in the Control-meso were below detection limit at all the three depths over the 13-days duration of the experiment. No change in NO_2^- concentrations was observed in the Dust-meso after both seedings. Depth-integrated NO_3^- concentrations showed that the stock increased to 10.6 mmol NO_3^- m^{-2} few hours after the R1 seeding, and then decreased to value significantly higher relative to the Control-meso at the end of the R1 (6.3 mmol NO_3^- m^{-2}, day 6) (Fig. 2b). The second seeding led to an additional increase in the NO_3^- stock of about 10 mmol NO_3^- m^{-2} relative to day 6 in the Dust-meso. The NO_3^- stock was maximum few hours after the R2 seeding (~ 16 mmol $NO_3^- m^{-2}$) and dropped to undetectable value at the end of the experiment.

After the simulated wet deposition in DUNE-1-P and DUNE-2-R1, Pulido-Villena et al. (2010, 2014) showed transient increases in DIP concentrations few hours after seeding (up to +12 nM at the surface) leading to net increases in the DIP stock (from +21 to +24 µmol DIP m^{-2}) in the Dust-meso relative to the Control-meso. 24 h after these seedings, DIP stock dropped to initial concentrations and remained constant until the end of the experiments. Over DUNE-2-R2, the second dust addition resulted

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in a higher increase in the DIP stock (+54 µmol DIP m^{-2} in the Dust-meso relative to the Control-meso) which remained stable and significantly higher relative to the controls until the end of the experiment (Pulido-Villena et al., 2014). No change in the DIP concentration was recorded in the Dust-meso after the Q seeding simulating a dry deposition event (Pulido-Villena, personal communication, 2013).

3.2 Response of the phytoplanktonic community to dust seeding

Depth-integrated Chl *a* and PP showed no significant differences between OUT and the Control-meso during the whole P and Q experiments (Figs. 3 and 4). From day 4 in the DUNE-2-R experiment, integrated Chl *a* and PP increased in OUT relative to the Control-meso (Fig. 5) as bacterial abundance did (Pulido-Villena et al., 2014). Over the duration of the three experiments, integrated Chl *a* and PP were homogeneous and remained stable in the Control-meso as shown by the low coefficients of variation (CV < 20 %). The mean C : N molar ratio in the particulate matter did not change significantly ($p > 0.05$) in the Dust-meso after all the seedings (P, Q, R) relative to the Control-meso (Fig. S2). The average C : N ratios were 7.5 ± 0.4, 7.6 ± 0.6 and 7.8 ± 0.6 respectively over the duration of the P, Q and R experiments.

Response of the phytoplanktonic community to a wet deposition event. In the DUNE-1-P experiment, the EC dust addition led to a 1.6 fold increase ($p < 0.05$) in the integrated Chl *a* relative to the Control-meso 24 h after the seeding and to an average 1.9 fold increase over the duration of the experiment (Fig. 3). The increase in Chl *a* was observed during the entire duration of the experiment as shown by the 1.9 fold increase ($p < 0.05$) detected 7 days after the seeding. The increase in Chl *a* was observed at all the sampling depths (Fig. S3) with similar relative changes ($p > 0.05$). Like Chl *a*, integrated PP strongly increased after the seeding during 7 days (1.8 fold on average) compared to the unamended controls ($p < 0.05$, Fig. 3). 24 h after seeding, PP in the Dust-meso was about twice higher in comparison with the Control-meso. Relative increases in PP at the surface and at 5 m depth were similar ($p > 0.05$, Fig. S4).

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Response of the phytoplanktonic community to a dry deposition event. Over the duration of the DUNE-1-Q experiment, integrated Chl *a* and PP did not significantly increase after the addition of non-EC dust throughout the water column ($p > 0.05$, Figs. 4, S5, S6). A slight stimulation of PP (1.2 fold, $p < 0.05$) was detected in the Dust-meso at 5 m depth, 48 h after dust addition (Fig. S6).

Response of the phytoplanktonic community to 2 successive wet deposition events. In the DUNE-2-R experiment, the first and second seedings led to averaged 1.9 and 2.4 fold increases in integrated Chl *a* relative to the Control-meso over the duration of the experiments (Fig. 5). Six days after both seedings, Chl *a* in the Dust-meso was still twice higher ($p < 0.05$) than in the Control-meso. The increase in Chl *a* was observed all over the three depths (Fig. S7). Integrated PP in the Dust-meso was enhanced 2.3 fold on average after the first seeding from day 1 (2.5 fold) to day 6 (2.5 fold) (Figs. 5, S8). Relative to the PP in the Dust-meso at the end of R1 (day 6), the second seeding permitted an additional 1.8 fold stimulation of PP 24 h after R2 (day 8). At the end of the experiment (day 13), PP in the Dust-meso was still twice higher than that in the unamended controls.

Comparison of the relative changes between P, R1 and R2 experiments- The mean relative changes (RC, Dust/Control) in PP were similar over the duration of R1 and R2 (RC = 2.3 and 2.4, respectively, $p > 0.05$, Fig. 6) while the RC in Chl *a* was higher over R2 (RC = 2.4, $p < 0.05$) than over R1 (RC = 1.9) (Fig. 6). After a first wet deposition event simulated in the P and R1 experiments, the respective RC in Chl *a* were similar (RC = 1.9, $p > 0.05$) while the RC in PP was higher over DUNE-2-R1 (RC = 2.3) than over P (RC = 1.8) ($p < 0.05$). The RC in both PP and Chl *a* were significantly higher after the R2 seeding than after the P seeding. The relative increase in Chl *a* was similar ($p > 0.05$) to that in PP in the P and R2 experiments. In the R1 experiment, the relative increase in PP (RC = 2.3) was slightly higher than that in Chl *a* (RC = 1.9).

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3.3 Nitrogen solubility associated with dust under abiotic conditions

Additions of both EC and non-EC dust did not significantly change ($p > 0.05$) the NH_4^+ concentrations compared to the unamended artificial seawater (data not shown). Significant increases ($p < 0.05$) in the NO_x concentrations were observed concomitantly with increasing EC dust concentrations for both contact times (Fig. 7a). No significant differences in the NO_x concentrations ($p > 0.05$) released from EC dust have been found between 3 and 24 h contact times. NO_x concentrations increased linearly ($r^2 = 0.99$) with the increasing EC dust concentration up to 17 µM for 20 mg dust L^{-1}. The percentages of dissolution ($\Delta NO_x \times 100 / N_{tot}$) reached 100 % from EC dust concentrations higher than 0.5 mg L^{-1}, at both time points (Fig. 7b). Additions of non-EC dust did not significantly increase ($p > 0.05$) NO_x concentrations relative to the unamended artificial seawater (Fig. 7a).

4 Discussion

The initial characteristics of seawater in all experiments were typical of LNLC environments as depicted by low nutrient concentrations (DIP, NO_3^-), low primary production and low phytoplanktonic biomass. These values were consistent with previous measurements in the surface waters of the open western Mediterranean Sea during stratification periods (Moutin and Raimbault, 2002; Bosc et al., 2004; Lopez-Sandoval et al., 2011; Pujo-Pay et al., 2011) and at the DYFAMED (DYnamique des Flux Atmosphériques en MEDiterranée) times series station http://www.obs-vlfr.fr/sodyf/; Marty and Chiaverini, 2002: Marty et al., 2002, 2008; Pulido-Villena et al., 2010) located in the northwestern Mediterranean Sea (43°25′ N 07°52′ E).

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4.1 Contrasted responses of the phytoplanktonic community to dust deposition events

Wet and dry dust deposition events induced contrasted responses of the phytoplanktonic community. The dust seeding experiments mimicking a wet deposition event (P, R1 and R2) induced a significant stimulation of both Chl *a* and PP from 1.8 to 2.4 fold (Fig. 6) while no changes were observed after the seeding simulating a dry deposition event (Q). The response of the phytoplankton to a wet deposition event was (1) fast as shown by significant increases in PP and algal biomass 24 h after dust addition, (2) long as depicted by increases recorded during at least a week after seeding and (3) homogeneous over the water column as illustrated by the stimulation of Chl *a* throughout the mesocosms. Despite the strong increases in biomass and PP following the wet deposition events, the values remained typical of oligotrophic systems (maximum of 0.23 µg Chl *a* L^{-1} and 13.1 mgC $m^{-3}\,d^{-1}$ in the Dust-meso). PP and Chl *a* in the Dust-meso were higher over DUNE-2-R2 than over R1. The second dust addition (R2) led to a higher Chl *a* increase (R2, RC = 2.4) relative to the first one (R1, RC = 1.9). During the first dust addition, 65 % of the increase in total phytoplanktonic biomass was indeed due to picophytoplankton (< 3 µm) while during the second one, the nano and microphytoplankton (> 3 µm) contributed to about 50 % of the Chl *a* increase (Giovagnetti et al., 2013). Surprisingly, no increase in Chl *a* was observed after the Q seeding while the flux of POC export was higher in the Dust-meso (8 ± 3 mg $m^{-2}\,d^{-1}$) compared to the controls (2 ± 1 mg $m^{-2}\,d^{-1}$) 24 h after addition (Desboeufs et al., 2013; Guieu et al., 2013b). This could be explained by potential top-down effects through grazing pressure which could have regulated the phytoplanktonic biomass. Slight increases in PP (+20 % at 5 m depth; $p < 0.05$, Fig. S6) and in *Synechococcus* abundance (+30 % at 5 m depth, $p < 0.05$, unpublished data) were recorded after the Q amendment. The abundance of copepods representing the main contributor to zooplankton increased also in the Dust-meso at the end of the Q experiment (+45 %) relative to the Control-meso (L. Stemmann, unpublished data, 2013). The increase in POC export flux without

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an associated increase in the biogenic stock could be also explained by the lithogenic ballast effect through aggregation and/or DOC sorption processes onto the settling particles (Desboeufs et al., 2013; Bressac et al., 2013).

4.2 Atmospheric inputs of new nutrients and pathways of deposition

Differences in the atmospheric supply of bioavailable new nutrients depending on the pathway of deposition (wet or dry) and on the type of dust (EC and non-EC) may explain the variability in the response of the phytoplanktonic community (Table 3). In the P and R experiments, the amendment of the mesocosms was performed with the EC dust mixed with ultrapure water in order to mimic a wet deposition event (and thus cloud process including the mixing of particles with compounds from anthropogenic activities such as HNO_3, Desboeufs et al., 2013) whereas in DUNE-Q, non EC dust mixed with seawater was used to simulate a dry deposition event.

DIP – The total content of P was similar in EC and non-EC dust (Table 1). Through *in vitro* dissolution experiments, both types of dust have been evidenced as a source of DIP (Pulido-Villena unpublished data; Pulido-Villena et al., 2010; Losno et al., 2013). Nevertheless, the dissolution of P associated with Saharan dust has been shown to be about 2 to 5 fold higher, for an equivalent dust concentration, in ultrapure water (wet deposition) than in seawater (dry deposition) likely due to a lower pH in ultrapure water (Ridame and Guieu, 2002). Indeed, in situ measurements showed significant increases in the DIP stock after simulated wet deposition events and no changes after the simulated dry deposition event (Pulido-Villena, personal communication, 2013) while abiotic dissolution experiments in seawater have shown a release of DIP from non-EC dust (+3 nM for a particulate dust concentration of 1 mg L^{-1} after a contact time of 6 h, Pulido-Villena, unpublished data). The low amount of DIP released after the dry deposition event could have been quickly used by the biological activity explaining why no increases in DIP concentrations were recorded after the Q seeding.

NO_3^- – The N content of EC dust was about 10 fold higher compared to the non-EC dust, resulting in significant changes in the atmospheric supply of dissolved inorganic

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nitrogen (DIN) depending on the pathway of deposition. Abiotic dissolution experiments showed that EC dust – mimicking Saharan dust wet deposition – was a significant source of NO_x whereas non-EC dust – mimicking Saharan dust dry deposition (Q) – was a negligible source of NO_x. Due to the simulation of cloud water processes that involved HNO_3, dissolution of EC dust released NO_x as NO_3^-. The release of NO_x by EC dust under *in vitro* conditions was confirmed by in situ increases in NO_3^- concentrations throughout the Dust-meso after the DUNE-2-R1 and -R2 wet deposition events (Fig. 2, Table 3). Assuming a homogeneous distribution of the EC dust within the Dust-meso, it would lead to a particulate dust concentration of about 1 mg L^{-1} leading to a potential input of 0.9 µM NO_3^- (Fig. 7a). The vertical profiles of NO_3^- in the mesocosms indicate a maximum net increase of NO_3^- of +9.8 µM at the surface soon after the R2 seeding (Fig. 2a), and reveal a non-homogeneous vertical distribution of the mineral particles in the Dust-meso. This was confirmed by optical measurements by Bressac et al. (2012, 2013) and by vertical distribution of crustal elements as aluminum (Desboeufs et al., 2013). Based on the dissolution results (Fig. 7), a complete dissolution of the NO_3^- associated with the EC dust (P, R1 and R2 seedings) is expected within three hours after additions, resulting in theoretical inputs of atmospheric new N of 8.5, 9.7 and 9.7 mmol NO_3^- m^{-2}, respectively in the Dust-meso for DUNE-1-P and DUNE-2-R1 and -R2. Over DUNE-2-R, the successive net increases in the NO_3^- stock just after seeding (R1: +10.6 mmol NO_3^- m^{-2} and R2: +10 mmol NO_3^- m^{-2}, Fig. 2b) are in the same order of magnitude than the input of atmospheric NO_3^- based on a total dissolution of N in EC dust (9.7 mmol $NO_3^- m^{-2}$). In addition to the atmospheric input of new DIN, N_2 fixation could be a significant source of new N as strong stimulations of N_2 fixation (up to 5.3 fold, Table 3) were detected after all the DUNE seedings (Ridame et al., 2013). The increases in N_2 fixation rates led to net inputs of new N of +39, +15 and +22 µmol N m^{-2} respectively after P, R1 and R2 seedings over the duration of the experiments (Ridame et al., 2013) which are negligible compared to the estimated supply of new N from the wet deposition events (from 8.5 to 9.7 mmol $NO_3^- m^{-2}$). Thus, the increase

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in NO_3^- concentrations observed after simulated wet deposition events could not be explained by the increase in N_2 fixation activity. Over DUNE-1-Q, N_2 fixation could be the main source of external N as dry deposition of non-EC dust was shown to be a negligible source of NO_3^-. The slight stimulation of PP 48 h after the Q seeding at 5 m depth (+20 %) could not be explained by the strong stimulation of N_2 fixation recorded 24 h (5.2 fold increase) and 48 h (3.3 fold increase) after addition. The increase in PP converted to N with a C : N ratio of 7.6 (measured during DUNE-1-Q) required an input of $\sim 9\,\mathrm{nmol\,N\,L^{-1}\,d^{-1}}$ which was much more higher than the input from increased N_2 fixation ($\sim 1\,\mathrm{nmol\,N\,L^{-1}\,d^{-1}}$).

DFe – EC and non-EC dust have a similar Fe content (Table 1) and represent a source of DFe to surface waters (Wagener et al., 2010; Losno et al., 2013). Results of dissolution experiments of both types of dust in artificial rainwater did not show any significant differences in DFe concentrations released for equivalent dust concentration and contact time (Losno et al., 2013). Despite these similarities, unexpected changes in the DFe concentrations were observed after the DUNE seedings by Wagener et al. (2010) and Wuttig et al. (2013) (Table 3). DFe concentrations dropped to about 1.5–2 nM after the DUNE-1-P, -Q and DUNE-2-R1 seedings due to DFe scavenging on settling dust particles and aggregates in the Dust-meso (T. Wagener, personal communication, 2013; Wagener et al., 2010; Wuttig et al., 2013) while a transient increase in the DFe concentrations in surface waters of about 2 nM was observed after the DUNE-2-R2 seeding (Wuttig et al., 2013).

The simulated wet deposition events induced significant increases in the DIP and NO_3^- concentrations in the mesocosms as well as significant stimulations of PP and Chl *a* (Table 3). On the opposite, after the simulated dry deposition event, no significant changes in DIP (and likely in NO_3^- concentrations) as in Chl *a* and PP were detected in the mesocosms (Table 3). As a consequence, the pathway of deposition and the type of dust clearly control the response of the phytoplanktonic community (biomass and productivity). As wet deposition is the main pathway of Saharan dust deposition over the western Mediterranean Sea (e.g., Loÿe-Pilot and Martin, 1996), atmospheric

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deposition is probably the main source of new nitrogen (NO_3^-) (Markaki et al., 2010) and new phosphorus (DIP) to the open surface waters during stratification periods.

4.3 Nutrient factors controlling primary production and biogeochemical consequences

Iron – Before seeding, DFe concentrations ranged from 2.3 to 3.3 nM in the tested waters (Table 2). Despite the decreases in DFe concentrations after the P and R1 seedings, phytoplanktonic biomass and productivity increased (Table 3). This indicates that during the DUNE experiments, the bioavailability of Fe was not a controlling factor of the growth and CO_2 fixing activity of the phytoplanktonic community which is in good agreement with previous results of Bonnet et al. (2005) during summer in the northwestern Mediterranean Sea.

Nitrogen and Phosphorus – The non-stimulation of PP after the dry deposition event suggest that the phytoplanktonic community may be limited by N, P or NP co-limited. In the DUNE-2-R experiment, the estimated NO_3^-/DIP ratio before seeding (< 6) was lower than the Redfield ratio (16/1) suggesting a N limitation of the phytoplankton activity. As the DIP concentration was also initially extremely low (5 nM, Table 2), PP could be likely co-limited by both N and P as previously shown during summer in the northwestern Mediterranean Sea (Tanaka et al., 2011). By increasing DIP and NO_3^- concentrations in P- and N-depleted surface waters, simulated wet deposition events relieve the potential N or NP colimitation of the phytoplanktonic activity. The wet deposition events induced significant changes in the biogeochemical conditions of the tested waters by modifying the NO_3^- and DIP stock and by altering the ambient NO_3^-/DIP ratio (Fig. 8). Due to the higher input of NO_3^- relative to DIP, wet deposition events (P, R1, R2) are characterized by a NO_3^-/DIP ratio much more higher that the Redfield ratio resulting in increases in the NO_3^-/DIP ratio in the Dust-meso just after seeding. Studies have previously shown that the atmospheric input to the Mediterranean Sea displays

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a high N/P ratio for dissolved inorganic forms (Sandroni et al., 2007; Markaki et al., 2010).

DUNE-1-P and DUNE-2-R1 – Both experiments showed similar patterns in the response of the phytoplanktonic community to a wet event as well as in the evolution of the DIP concentration. As the NO_3^- stock in the Dust-meso was higher than in the controls at the end of R1 and as the DIP stock was similar to the controls, the NO_3^-/DIP ratio strongly increased from < 6 (initially) to > 150 at the end of the R1 experiment (Fig. 8) suggesting a switch from an initial N limitation or NP colimitation of the phytoplanktonic activity towards a severe P limitation until the end of R1. Net increase in integrated PP ($PP_{dust} - PP_{control}$) 24 h after seeding was converted to P using a C/P molar ratio of 245/1 determined in the particulate organic matter in surface waters of the northwestern Mediterranean Sea during stratification (Tanaka et al., 2011) and to N using a C : N ratio measured during DUNE. The phytoplanktonic P requirement (+25 and +18 µmol DIP m^{-2} for DUNE-1-P and DUNE-2-R1 at day 1) was in the same order of magnitude than the net increase in the DIP stock in the Dust-meso (+21 and +24 µmol DIP m^{-2} for DUNE-1-P and DUNE-2-R1) while the phytoplanktonic N requirement was much lower than the increase in the NO_3^- stock. The stimulation of PP 24 h after seeding can explain the rapid DIP depletion. Bacterial respiration was also stimulated after these seedings (Pulido-Villena, 2010, 2014) suggesting a potential competition for the DIP uptake between heterotrophic bacteria and autotrophic phytoplankton as already reported during microcosm experiments in the tropical Atlantic and Mediterranean Sea (Thingstad et al., 2005; Maranon et al., 2010). As the ~ 2 fold increase in PP and Chl *a* in the Dust-meso lasted during a week, it suggests a rapid remineralization of DIP by heterotrophic bacteria and/or the uptake of dissolved organic phosphorus (DOP) as a source of P through the alkaline phosphatase enzyme (Beardall et al., 2001). The PP integrated over the duration of the experiments and the estimated nutrient requirements showed that the atmospheric input of NO_3^- largely exceeds the phytoplanktonic N demand which may explain the NO_3^- accumulation at the end of the experiment.

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DUNE-2-R2 – Although the two seedings performed during DUNE-2-R were identical, the evolution of the NO_3^- and DIP stock after the second seeding was different relative to that observed after the first one (Fig. 8). The NO_3^- stock in the dust-meso decreased to undetectable values at the end of R2 while the DIP stock remained higher than in the controls. This leads to a NO_3^-/DIP ratio much lower than 16/1 indicating a potential N limitation of the phytoplanktonic activity at the end of the experiment. Over R2, the change in size structure of the phytoplanktonic community towards a larger dominance of big cells is consistent with a higher consumption of NO_3^- (Giovagnetti et al., 2013) as large cells have higher half-saturation constants (Aksnes and Egge, 1991; Hein et al., 1995) and higher minimum cellular metabolic requirements than small cells (Grover, 1991).

The DIP stock remained surprisingly stable and significantly higher relative to the controls until the end of R2 despite twice higher rates of PP in the Dust-meso than in the Control-meso. This does not imply that the input of new DIP was not used by phytoplankton but rather that the rate of DIP consumption through PP was compensated by an equivalent rate of DIP production and thus maintaining a quite constant DIP stock. It is unlikely that the increase in PP was sustained by DOP as the use of DOP by phytoplankton is more energy consuming through the synthesis of alkaline phosphatase enzyme. We thus hypothesize that the atmospheric DIP was assimilated by phytoplankton to sustain increases in biomass and PP, and that heterotrophic remineralization of organic phosphorus kept DIP stock quite stable through high rates of P recycling. This would imply that during R2, heterotrophic bacteria did not out-compete phytoplankton for P resource as is suspected for R1. As suggested by Pulido-Villena et al. (2014), even if carbon appeared not to be the limiting nutrient, bacterial respiration could have been stimulated by labile phytoplankton-derived dissolved organic matter (DOM). Besides carbon, phytoplankton DOM also provides P, offering an alternate source of the limiting element to bacteria. This shift in the P source could explain the non-utilization of DIP by heterotrophic bacteria during the second seeding.

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Consequences of dust deposition on new production – During the summer oligotrophic conditions in the western Mediterranean Sea, PP is mainly dominated by regenerated production as about 85 % of PP is sustained by internal recycling of organic matter within the euphotic zone through remineralization processes (Marty et al., 2002; Moutin and Raimbaut, 2002; L'Helguen et al., 2002). As atmospheric deposition constitutes a source of external nutrients to the surface mixed layer, it induces by definition new production (NP). Thus, the increase in PP in the Dust-meso 24 h after seeding observed in the DUNE-1-P and DUNE-2-R experiments can be associated with NP. We consider that after 24 h, the increase in PP could be mainly supported by regenerated nutrients. The contribution of NP to PP increased in the Dust-meso from initially 15 % to 60–70 % 24 h after seeding. Wet simulated dust deposition event induced a switch from a regenerated-production based system (NP/PP = 0.15) to a new-production based system (NP/PP = 0.60–0.70).

5 Summary and conclusions

Our results from original mesocosm experiments demonstrate that atmospheric dust deposition does greatly influence primary productivity and algal biomass in LNLC environments. The response of the phytoplanktonic community to dust events was quantified: primary production and biomass increased ~ 2 fold and the stimulation can be observed for at least one week. Differences in the atmospheric supply of bioavailable new nutrients depending on the pathway (wet or dry) and on the type of dust led to contrasted responses of the phytoplanktonic community. Primary production increased significantly only after wet deposition. In addition to being a source of DIP, wet deposition due to cloud processes and mixing with anthropogenic species such as HNO_3, represents also a significant source of NO_3^- inducing significant increase in the ambient NO_3^-/DIP ratio. As dry deposition was not a significant source of NO_3^-, primary production was likely N limited or co-limited by both N and P. By transiently increasing DIP and DIN concentrations in P-N starved surface waters of the Mediterranean Sea,

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wet deposition of Saharan dust relieves the potential N or NP co-limitation of the phytoplanktonic activity. This study underlines the importance of Saharan dust deposition on the phytoplankton dynamics in the Mediterranean Sea and potentially in all LNLC areas impacted by dust deposition such as the tropical Atlantic and Pacific Oceans. Despite the significant stimulation of the phytoplanktonic production and the increase in the POC export after wet events (Desboeufs et al., 2013; Bressac et al., 2013), Guieu et al. (2013b) have shown that simulated wet deposition did not result in a simple fertilization effect as the oligotrophic ecosystem keeps or reinforces its net heterotrophic character thanks to the high organic carbon remineralization due to microbial food web processes.

Modeling studies suggest that changes in climate and land use practices over recent decades may have altered dust fluxes and thus, aeolian Fe, P and N inputs to the oceans (e.g., Mahowald and Luo, 2003). In addition, in the future, a warming atmosphere and surface waters could potentially increase the stratification of the surface waters in the Mediterranean Sea (Somot et al., 2008) and other oceanic areas such as subtropical gyres (Bopp et al., 2001). In response to this, the biogeochemical impact of the Saharan deposition events on the primary production could be more pronounced in particular during stratification periods.

Supplementary material related to this article is available online at http://www.biogeosciences-discuss.net/11/753/2014/bgd-11-753-2014-supplement.pdf.

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Table 1. Particulate P, Fe and N (%, in weight) in the EC and non-EC dust used during DUNE (from Ridame et al., 2013). Means that were not significantly different for a given chemical element between the different experiments ($p > 0.05$) are labeled with the same letter (in parenthesis).

	DUNE-1-P 10–18 Jun 2008	DUNE-1-Q 20–27 Jun 2008	DUNE-2-R (R1, R2) 26 Jun–9 Jul 2010
Dust treatment	evapocondensed	non-processed	evapocondensed
Simulated deposition	wet	dry	wet
P (%)	0.045 ± 0.015^{a} (A)	0.044 ± 0.009^{a}(A)	0.055 ± 0.003^{b}(A)
Fe (%)	2.31 ± 0.04^{a} (B)	2.28 ± 0.19^{a}(B)	2.26 ± 0.03^{b}(B)
N (%)	1.19 ± 0.05^{a} (C)	0.11 ± 0.01^{a}(D)	1.36 ± 0.09^{b}(E)

[a] Guieu et al. (2010b).
[b] Desboeufs et al. (2013).

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Table 2. Initial temperature and biological and chemical properties of seawater before seeding in experiments DUNE-1-P, -Q and DUNE-2-R (average in the Dust-meso, Control-meso and Out). DIP: dissolved inorganic phosphorus; dl: detection limit (30 nM for NO_3^-); nd: no data. Data for temperature, chlorophyll *a*, DIP, NO_3^- and DFe are the means at 0.1, 5 and 10 m depth. Data for primary production and C : N ratio are the means at 0.1 and 5 m depth for P and Q experiments and at 5 m depth for R experiment. Means that were not significantly different for a given parameter between the different experiments ($p > 0.05$) are labeled with the same letter (in parenthesis).

	DUNE-1-P	DUNE-1-Q	DUNE-2-R
Temperature (°C)	19.6 ± 0.7 (A)	20.3 ± 0.5 (B)	21.2 ± 0.4 (C)
Chlorophyll *a* ($\mu g\,L^{-1}$)	0.11 ± 0.03 (A)	0.08 ± 0.02 (B)	0.07 ± 0.02 (B)
Primary production, mg C m^{-3} d^{-1}	5.35 ± 1.11 (A)	4.16 ± 0.38 (B)	3.89 ± 0.46 (B)
C : N (mol/mol)	7.8 ± 0.5 (A)	7.3 ± 0.5 (B)	7.5 ± 0.6 (A, B)
DIP, nM	5 ± 2[a] (A)	2 ± 0[b](B)	5 ± 3[c](A)
NO_3^-, nM	nd	nd	< dl
DFe, nM	2.4 ± 0.3[d](A)	2.3 ± 0.3[e](A)	3.3 ± 0.8[f](B)

[a] Pulido-Villena et al. (2010).
[b] E. Pulido-Villena, personal communication (2013).
[c] Pulido-Villena et al. (2014).
[d] Wagener et al. (2010).
[e] T. Wagener, personal communication, 2013.
[f] Wuttig et al. (2013).

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Table 3. Summary of the biological and chemical changes observed after the DUNE seedings in the Dust-meso relative to the Control-meso. Results from this study; Ridame et al. (2013); Pulido-Villena et al. (2010, 2014); Pulido-Villena, personal communication (2013); Wagener et al. (2010); T. Wagener, personal communication, 2013; Wuttig et al. (2013)

DUNE-1-P wet	DUNE-1-Q dry	DUNE-2-R1 wet	DUNE-2-R2 wet
PP ↑	PP ↔	PP ↑	PP ↑
Chl *a* ↑	Chl *a* ↔	Chl *a* ↑	Chl *a* ↑
N_2 Fixation ↑	N_2 Fixation ↑	N_2 Fixation ↑	N_2 Fixation ↑
Bacterial respiration ↑	Bacterial respiration↑	Bacterial respiration ↑	Bacterial respiration ↑
DIP ↑	DIP ↔	DIP ↑	DIP ↑
NO_x ↑ *	NO_x ↔ *	NO_x ↑	NO_x ↑
DFe ↓	DFe ↓	DFe ↓	DFe ↑

* From abiotic dissolution experiments, this study.

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Fig. 1. (A) Addition of ^{13}C tracer immediately after seawater sampling, **(B)** incubations of the ^{13}C-amended bottles under in situ conditions at the corresponding sampling depths.

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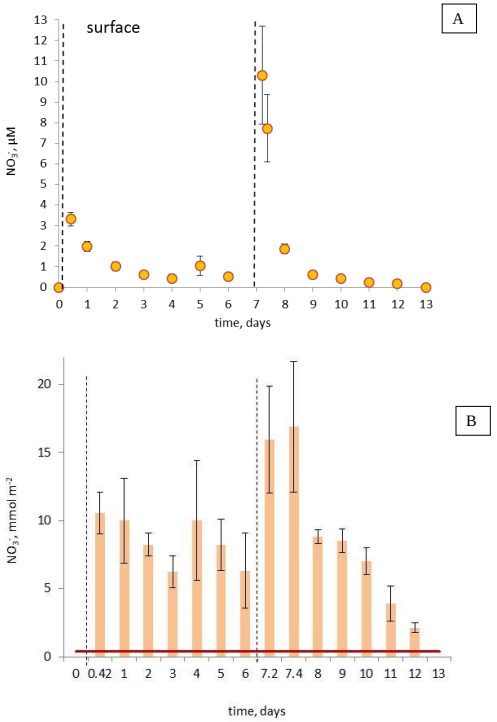

Fig. 2. (A) Mean NO_3^- concentration (µmol L^{-1}) during the DUNE-2-R experiment in the Dust-meso at the surface (0.1 m depth). The dotted lines represent the time of the dust seedings. Data represent the average and standard deviation of the three replicate mesocosms. NO_3^- concentrations were under detection limit (< 30 nM) in the Control-meso and outside over the duration of the experiment as well as in the Dust-meso before seeding (day 0) and at the end of the experiment (day 13), **(B)** integrated NO_3^- (mmol N m^{-2}) over the depth of the mesocosm (12.5 m) in the Dust-meso during the DUNE-2-R experiment. Assuming a maximum NO_3^- concentration of 30 nM in the Control-meso, the integrated NO_3^- should be lower than 0.4 mmol N m^{-2} (red line).

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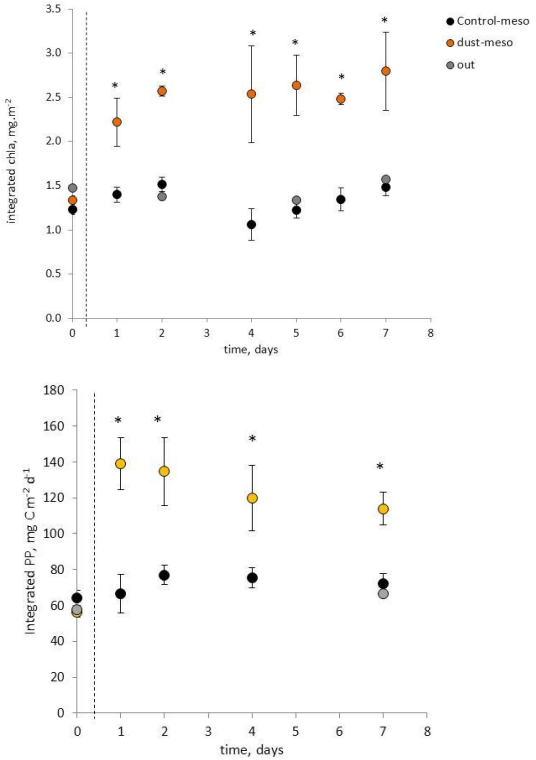

Fig. 3. Mean integrated chlorophyll *a* (Chl *a*) in $mg\,m^{-2}$ (top panel) and mean integrated primary production (PP) in $mg\,C\,m^{-2}\,d^{-1}$ (bottom panel) over mesocosms during the DUNE-1-P experiment in the Control-meso (black dot), Dust-meso (orange dot) and out (grey dot). The dotted line represents the time of the dust seeding. Data in the Control- and Dust-meso represent the average and standard deviation of the three replicate mesocosms. Means in the Dust-meso that were significantly different from the Control-meso ($p < 0.05$) are labeled with the * symbol.

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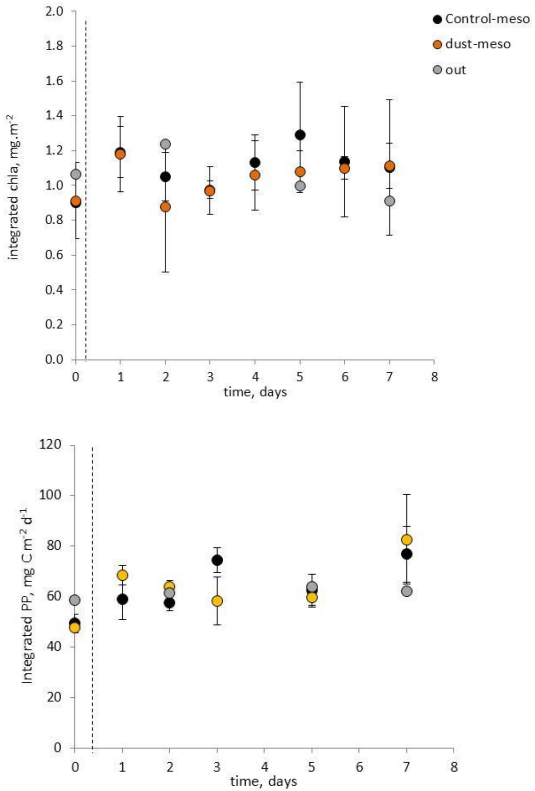

Fig. 4. Mean integrated chlorophyll *a* (Chl *a*) in mg m^{-2} (top panel) and mean integrated primary production (PP) in mg C m^{-2} d^{-1} (bottom panel) over mesocosms during the DUNE-1-Q experiment in the Control-meso (black dot), Dust-meso (orange dot) and out (grey dot). The dotted line represents the time of the dust seeding. Data in the Control- and Dust-meso represent the average and standard deviation of the three replicate mesocosms.

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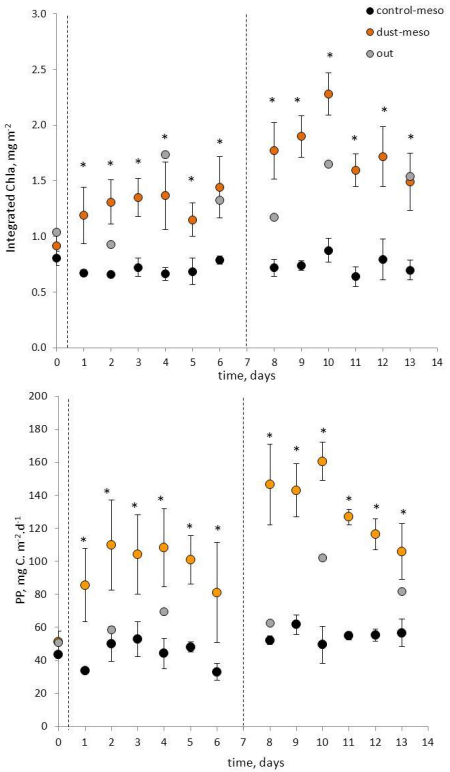

Fig. 5. Mean integrated chlorophyll *a* (Chl *a*) in mg m^{-2} (top panel) and mean integrated primary production (PP) in mg C m^{-2} d^{-1} (bottom panel) over mesocosms during the DUNE-2-R1 and -R2 experiments in the Control-meso (black dot), Dust-meso (orange dot) and out (grey dot). The dotted line represents the time of the dust seeding. Data in the Control- and Dust-meso represent the average and standard deviation of the three replicate mesocosms. Means in the Dust-meso that were significantly different from the Control-meso ($p < 0.05$) are labeled with the * symbol.

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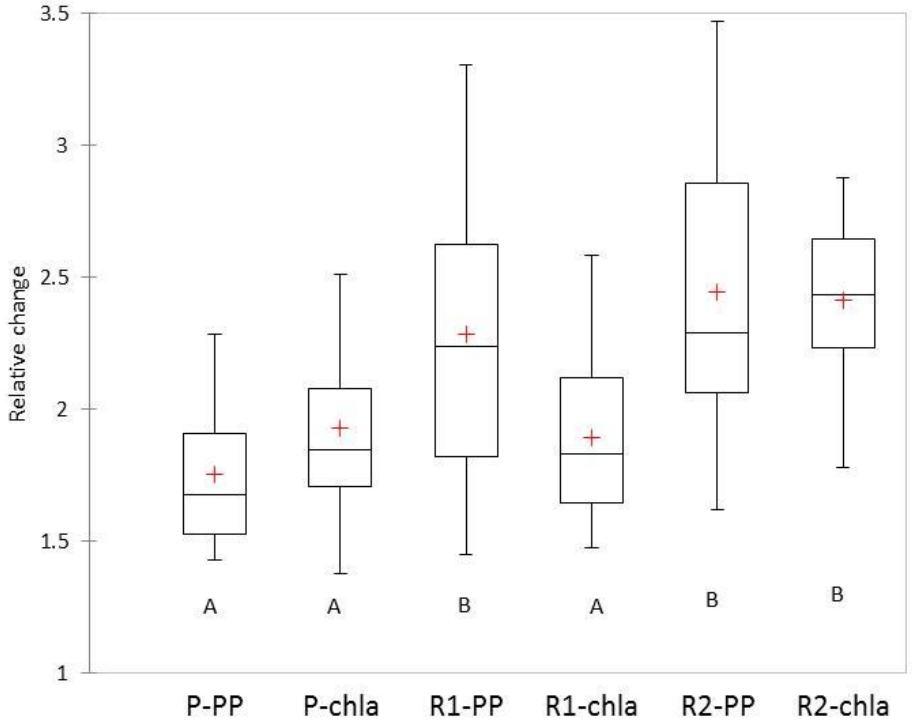

Fig. 6. Box plot of the relative changes in integrated Chl *a* (Chl a_{dust}/Chl $a_{control}$) and PP ($PP_{dust}/PP_{control}$) over the duration of the DUNE experiments. Means that were not significantly different between the different parameters and experiments ($p > 0.05$) are labeled with the same letter.

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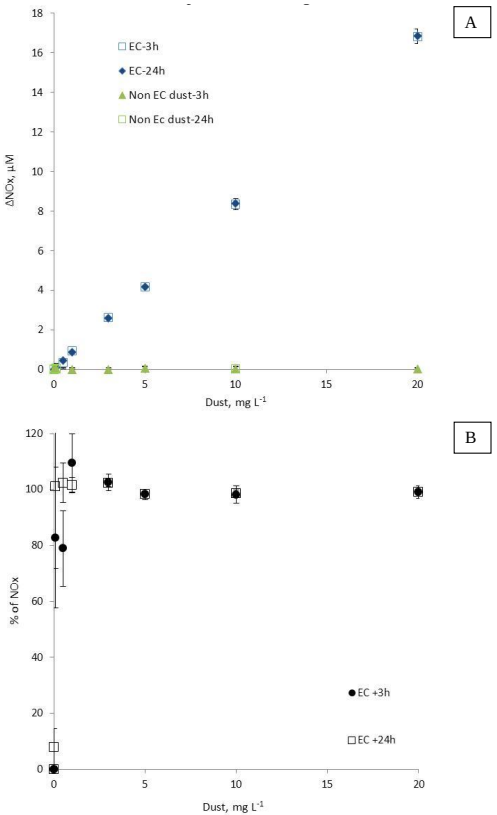

Fig. 7. Results of abiotic dissolution experiments: **(A)** ΔNO_x ($[NO_x]_{\text{after the introduction of dust}} - [NO_x]_{\text{initial}}$) in µM relative to the amount of dust (EC and non EC dust) introduced in seawater for contact times of 3 and 24 h, **(B)** percentage of NO_x released from EC dust (NO_x % = $\Delta NO_x \times 100 / N_{\text{total dust}}$) as a function of the dust amount introduced in seawater for contact times of 3 and 24 h. Data represent the average and standard deviation of the three replicates.

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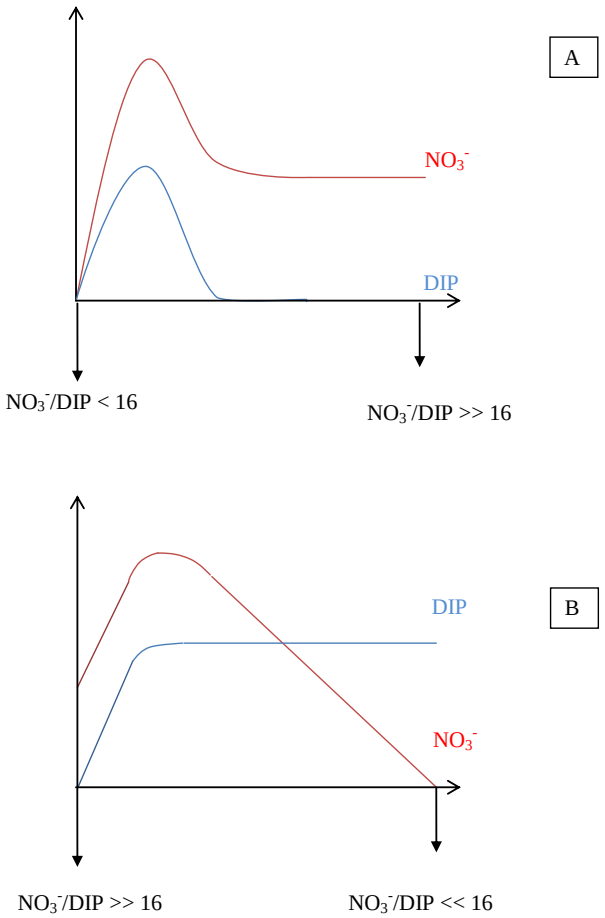

Fig. 8. Conceptual representation of the temporal changes in the variation (Dust – Control) of NO_3^- (red line) and DIP stock (blue line) **(A)** after the DUNE-2-R1 seeding and **(B)** after the DUNE-2-R2 seeding.

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