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Contrasted Saharan dust events in LNLC environments: impact on nutrient dynamics and primary production

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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 ($10 \, \mathrm{g \, m^{-2}}$) into large mesocosms ($52 \, \mathrm{m^3}$). Three distinct dust addition experiments 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 and DUNE-2-R2: simulation of two successive wet depositions) in the northwestern oligotrophic Mediterranean Sea. No changes in primary production (PP) and chlorophyll a concentrations (Chl a) were observed after a dry deposition event, while a wet deposition event resulted in a rapid (24 h after dust addition), strong (up to 2.4-fold) and long (at least a week in 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 nitrate (NO_3^-) (net increases up to $+9.8 \, \mu M \, NO_3^-$ at 0.1 m in depth) to the nutrient-depleted surface waters, due to cloud processes and mixing with anthropogenic species such as HNO_3. The dry deposition event was shown to be a negligible source of NO_3^-. By transiently increasing DIP and NO_3^- concentrations in N–P 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, and taking into account the stimulation of the biological activity, a wet deposition event resulted in a strong increase in the NO_3^-/DIP ratio, from initially less than 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 strongly 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 contrasting responses of the phytoplanktonic community due to differences in the atmospheric supply of bioavailable new nutrients. Our results from original mesocosm experiments demonstrate that atmospheric dust wet deposition

greatly influences primary productivity and algal biomass in LNLC environments through changes in the 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 of 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 mineral 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 regions (Boyd et al., 2000; de Baar et al., 2005, Boyd et al., 2007; 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 poorly understood, although LNLC regions represent 60 % of the global 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 tackling the question of the planktonic response to atmospheric nutrient inputs. The Mediterranean Sea is an oligotrophic quasi-enclosed basin that receives a noticeable flux of dust, mainly derived from the 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 of playing 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 microcosms 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 an 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, Guieu et al., 2014a). Due to the logistical difficulties of investigating in situ natural dust events, and due to the inherent limitations of microcosm experiments, new experimental approaches have been developed in the framework of the DUNE project. This allowed for the first time the performance of realistic dust seeding in in situ large metal-free mesocosms (52 m^3), and the following of the impact on chemistry, biology and particle dynamics, taking into account the vertical dimension. In this context, the present study is focused on the response of the phytoplanktonic community to contrasted Saharan dust events (wet and dry deposition). Here, we quantified the changes in in situ concentrations of dissolved inorganic nitrogen, as well as the changes in the primary production and algal biomass after simulated dust deposition events. This study is complementary to the companion paper of Giovagnetti et al. (2013), focusing on changes in the structure and composition of the phytoplanktonic community as well as in the ecophysiological state of the phytoplanktonic cells after two successive simulated wet deposition events (DUNE-2-R).

2 Material and methods

2.1 Experimental design and dust characterization

Three distinct experimental dust additions to large mesocosms were conducted in June 2008 and from June to July 2010 in the northwestern Mediterranean Sea, in the framework 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 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, 2014a). The mesocosm experiment design and the accuracy of the strategy are described fully 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 into the sea floor at 25–30 m in depth. The mesocosms are closed systems, which prevent lateral advection and allow process studies in the vertical dimension over time. Sediment traps (a 250 mL HDPE bottle) were screwed to the bases 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, 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 ($< 20\,\mu m$) of Saharan soils collected in southern Tunisia, which is a Saharan aerosol source area for the western Mediterranean Sea (details in Guieu et al., 2010b and Desboeufs, et al., 2014). To simulate a Saharan deposition event, we did not use collected rainwater or aerosols, but instead we used the fine fraction of soils as an analog to Saharan aerosols, in order to obtain enough quantity of the same material. The amount of dust per mesocosm required was 41.5 g, which resulted in a total of 125 g of dust for the three replicates for only one seeding experiment. Such a large amount of particles could not be collected from airborne dust in the vicinity of the experimental area. Moreover, Saharan dust events are sporadic, and collecting rainwater in the field at the time of the mesocosm experiment would have introduced large uncertainties into the feasibility of the project. Three other mesocosms served as controls (C1, C2 and C3, hereafter referred 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 in 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, as described in Guieu et al. (2010b). 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 the data between 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 reproducing the photochemistry, the gradients in pH, and ionic strength during cloud processing of dust particles (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, such as sulfate and nitrate, due to cloud processing during atmospheric transport (Putaud et al., 2004; Crumeyrolle et al., 2008). This evapocondensed dust (hereafter referred to as "EC dust") contained on average $0.045 \pm 0.015\,\%$ of P, $2.31 \pm 0.04\,\%$ of Fe and $1.19 \pm 0.05\,\%$ of N, in weight (Table 1). The seeding 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, as (i) the mixing of dust with anthropogenic components is a process that occurs mostly during cloud processes, and (ii) dissolution processes occur mainly in rainwater in the case of 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 $0.044 \pm 0.009\,\%$ of P, $2.28 \pm 0.19\,\%$ of Fe, and $0.11 \pm 0.01\,\%$ of N (Table 1). The seeding 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, as (i) the untreated dust is representative of dry-deposited dust in the Mediterranean Sea, and (ii) dissolution processes occur in seawater in the case of a dry-type deposition event. Indeed, the internal mixing between dust and polluted species is not systematically observed in the Mediterranean area (Marconi et al., 2014). Kandler et al. (2007) show that the mixing is below 2 % for the transported dust particles larger than $5\,\mu m$, and up to 5 % for particles larger than $1\,\mu m$, i.e., for the particles preferentially removed by dry deposition. The size distribution and the chemical composition of our untreated dust are consistent with the characteristics of long-range transported dust (Guieu et al., 2010b; Formenti et al., 2011; de Leeuw et al., 2014).

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., 2014a) were performed with the same amount of EC dust and with the same deposition setup as in DUNE-1-P, simulating a wet deposition event. The dust was characterized by an average content of $0.055 \pm 0.003\,\%$ P, $2.26 \pm 0.03\,\%$ Fe and $1.36 \pm 0.09\,\%$ N (Table 1; see details in Desboeufs et al., 2014). During the first seeding (DUNE-2-R1), the comparison of the data between the Control-meso and Dust-meso gives information on the effect of one wet deposition event (as for DUNE-1-P), while during DUNE-2-R2, the comparison between the Control-meso and Dust-meso gives information on the effect of the combination of two successive wet deposition events.

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 parentheses).

	DUNE-1-P 10–18 June 2008	DUNE-1-Q 20–27 June 2008	DUNE-2-R (R1, R2) 26 June–9 July 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. (2014).

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, 2014a). Small differences of the N content in EC dust used in the 2008 and 2010 experiments (1.19 ± 0.05 % and 1.36 ± 0.09 %) were observed (details in Guieu et al., 2014a).

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 using the trace metal clean sampling system, in the morning at two depths (0.1 and 5 m in depth) during DUNE-1-P and DUNE-1-Q, and at 5 m in depth during DUNE-2-R, for the determination of primary production (PP). Samples were collected in the six mesocosms and outside the mesocosms before and after dust seeding. CO_2 fixation rates were determined using the ^{13}C-tracer addition method (e.g., Slawyk et al., 1977). During DUNE-1-P and DUNE-1-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 labeling 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 on the same order of magnitude as those found between triplicate mesocosms (CV < 17 %). Immediately after sampling, ^{13}C tracer was added to obtain a final enrichment of about 9 atom % excess, and each bottle was shaken well. The atom % excess of the dissolved inorganic carbon (DIC) was calculated by using measured DIC concentrations at the LOCEAN laboratory (see the detailed protocol in Corbière et al., 2007). 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 in depth). Incubations were terminated the following morning by filtration onto precombusted 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 (C) and nitrogen (N) 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. The standard deviations were $0.009 \, \mu mol \, L^{-1}$ and $0.004 \, \mu mol \, L^{-1}$ 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 in depth was integrated over the mesocosm depth (12.5 m). We assume that the PP at 5 m in depth was similar to that at 12.5 m in depth, based on the similarity of the chlorophyll a concentrations (Chl a) measured at 5 and 10 m in depth in the Control-meso and Dust-meso over the experiments ($p > 0.05$). Besides, on some selected sampling days, PP was measured at the three depths (0.1, 5 and 10 m in depth). Depth-integrated PP calculated from measured data at the three depths was similar (-4 %) to that extrapolated from data measured at 0.1 and 5 m (assuming that the PP at 12.5 m in depth was similar to that measured at 5 m in depth).

For DUNE-2-R, PP measured at 5 m in depth was integrated assuming a uniform distribution over the entire mesocosm. 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 in depth was similar ($p > 0.05$), while Chl a in surface waters was slightly higher than deeper ($+ \sim 30$ %, $p < 0.05$) at the three sampling times. Also, measurements of PP at the three depths at some selected sampling times showed that depth-integrated PP calculated

from measured data at 0.1, 5 and 10 m in depth was not significantly different ($+4\%$) from that extrapolated from data measured at 5 m in depth (assuming a homogeneous distribution over the mesocosm depth).

2.3 Chlorophyll *a*

Two liters of seawater from 0.1, 5 and 10 m in depth were collected on a 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, 1972). The Chl *a* concentrations measured at the three sampling depths were integrated over the mesocosm depth (12.5 m).

2.4 Nutrient concentrations

Twenty milliliters of filtered seawater ($< 0.2\,\mu m$, Sartobran cartridge filters) from 0.1, 5 and 10 m in depth were collected in acid-washed high-density polyethylene (HDPE) 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 the 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 of dissolved Fe (DFe) ($< 0.2\,\mu m$) was measured by flow injection analysis with online preconcentration and chemiluminescence detection (FIA-CL); the detection limit was 10 pM (details in Wagener et al., 2010). These data are presented in Wagener et al. (2010) and Wuttig et al. (2013).

2.5 Dissolution experiments under abiotic conditions

Both types of Saharan dust (EC and non-EC dust) used during the DUNE-1-P and DUNE-1-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 seedings. 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 the liquid phase and to stop dissolution processes. The filtrate from each sample was subsampled to measure the concentrations 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 analyzed immediately. Briefly, the NO_x concentration was analyzed 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^+ concentrations, 40 mL of filtrate were recovered in a Schott flask (Duran) and analyzed 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 $0.19 \pm 0.02\,\mu mol\,L^{-1}$ ($n = 12$) and $1.46 \pm 0.12\,\mu mol\,L^{-1}$ ($n = 7$), respectively.

2.6 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 the 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 test in XLstat software.

3 Results

3.1 Characteristics of the seawater

Statistical analysis of biological and chemical parameters in Table 2 showed no significant differences between Control-meso, Dust-meso and out before seeding for all experiments (Guieu et al., 2010b; 2014a). 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

Table 2. Initial temperature and biological and chemical properties of seawater before seeding in experiments DUNE-1-P, DUNE-1-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 in depth. Data for primary production and the C : N ratio are the means at 0.1 and 5 m in depth for the DUNE-1-P and DUNE-1-Q experiments, and at 5 m in depth for the DUNE-2-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 parentheses).

	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 (μg 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).

biomass (Chl $a < 3\,\mu$m) accounted for about 70 % of the total phytoplanktonic biomass before the DUNE-1-Q and DUNE-2-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 $< 3\,\mu$m 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; $p < 0.05$). The initial NO_3^- and NO_2^- concentrations were below the 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 the DUNE-1-P and DUNE-1-Q experiments, but are strongly suspected to be below the detection limit before seeding, as shown in DUNE-2-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 DUNE-1-Q (~ 2.3 nM, $p < 0.05$, Table 2).

The initial seawater temperatures over the water column were greater than 19 °C (Table 2). During DUNE-1-P, the temperature was lower than during the other ones (Guieu et al., 2014a). 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, temperatures 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., 2014a). 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., 2014a). The highest temperature was recorded during DUNE-2-R2 (up to 27.3 °C at the surface). Photosynthetically available radiation (PAR) was measured at the subsurface of the mesocosms during DUNE-2-R (Giovagnetti et al., 2013; Guieu et al., 2014a). Over the 13 days of the experiment, PAR was high, with maximum values of $\sim 900\,\mu$mol photons m^{-2} s^{-1} at the subsurface. During DUNE-1-P, the percentage of sunshine duration over the whole daylight time was lower than during DUNE-1-Q, where the values were close to the maximum (Guieu et al., 2014a), so it is likely that PAR values increased between the P and Q experiments.

3.2 Changes in the nutrient concentrations after seeding

After the DUNE-2-R1 and DUNE-2-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. 1a). Ten hours after the first seeding, the NO_3^- concentrations in the surface waters reached $3.3 \pm 0.3\,\mu$M, and then decreased to $0.5 \pm 0.1\,\mu$M at the end of R1 (day 6). Five hours after the second seeding, an additional increase in the NO_3^- concentration ($+9.8\,\mu$M relative to day 6 in the Dust-meso) was
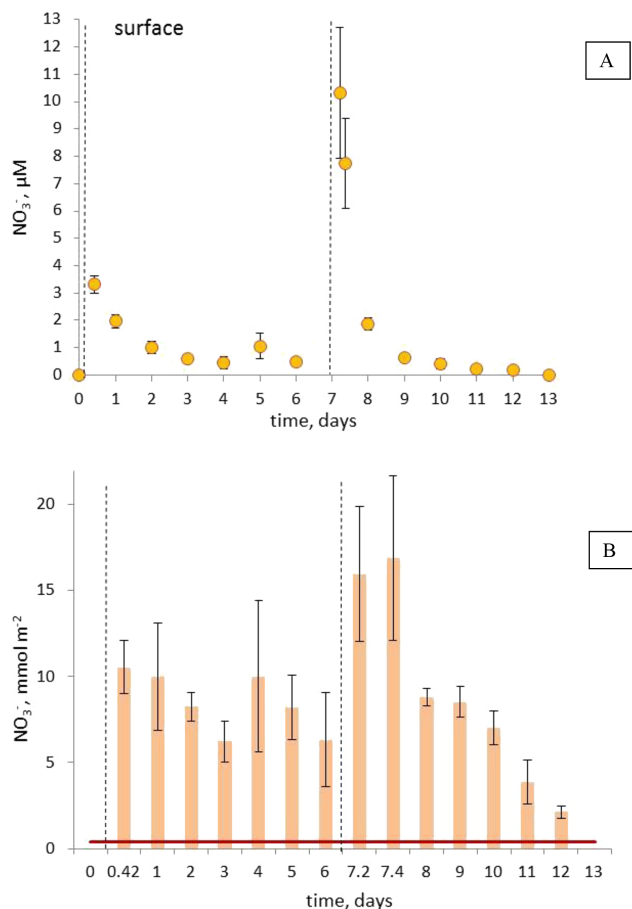
Figure 1. (A) Mean NO_3^- concentration (μmol L^{-1}) during the DUNE-2-R experiment in the Dust-meso at the surface (0.1 m in depth). The dotted lines represent the time of the dust seedings. Data represent the average and standard deviations of the three replicate mesocosms. NO_3^- concentrations were under the 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).

detected at the surface, leading to NO_3^- concentrations of $10.3 \pm 2.4\,\mu$M. At the end of R2, the NO_3^- concentration in the Dust-meso decreased down to the detection limit. Lower increases in the NO_3^- concentration were recorded at 5 and 10 m in depth in the Dust-meso (maximum of $\sim 1\,\mu$M) (Supplement Fig. S1). NO_3^- and NO_2^- concentrations in the Control-meso were below the detection limit at all three depths over the 13-day duration of the experiment. No change in the NO_2^- concentration was observed in the Dust-meso after both seedings. Depth-integrated NO_3^- concentrations showed that the stock increased to 10.2 mmol NO_3^- m^{-2}

a few hours after the R1 seeding, and then decreased to a value significantly higher relative to the Control-meso at the end of the R1 (6.3 mmol NO_3^- m^{-2}, day 6) (Fig. 1b). 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 at a maximum a few hours after the second seeding, ~ 16 mmol NO_3^- m^{-2}, representing the cumulated effect of the two seedings, and then 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 the DIP concentrations a few hours after seeding (up to $+12$ nM at the surface), resulting in net increases in the DIP stock (from $+21$ to $+24\,\mu$mol DIP m^{-2}) in the Dust-meso relative to the Control-meso. Twenty-four hours after these seedings, DIP stock dropped to initial concentrations (no significant difference compared to the controls), and remained constant until the end of the experiments. Over DUNE-2-R2, the dust addition led to a new increase in the DIP stock ($+54$–$104\,\mu$mol DIP m^{-2} in the Dust-meso relative to the Control-meso) that was higher than after the first one. The DIP stock 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 DUNE-1-Q seeding simulating a dry deposition event (Pulido-Villena, personal communication, 2013).

3.3 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 DUNE-1-P and DUNE-1-Q experiments (Figs. 2 and 3). From day 4 in the DUNE-2-R experiment, integrated Chl a and PP increased in out relative to the Control-meso (Fig. 4) 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 (Supplement 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 DUNE-1-P, DUNE-1-Q and DUNE-2-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. 2).
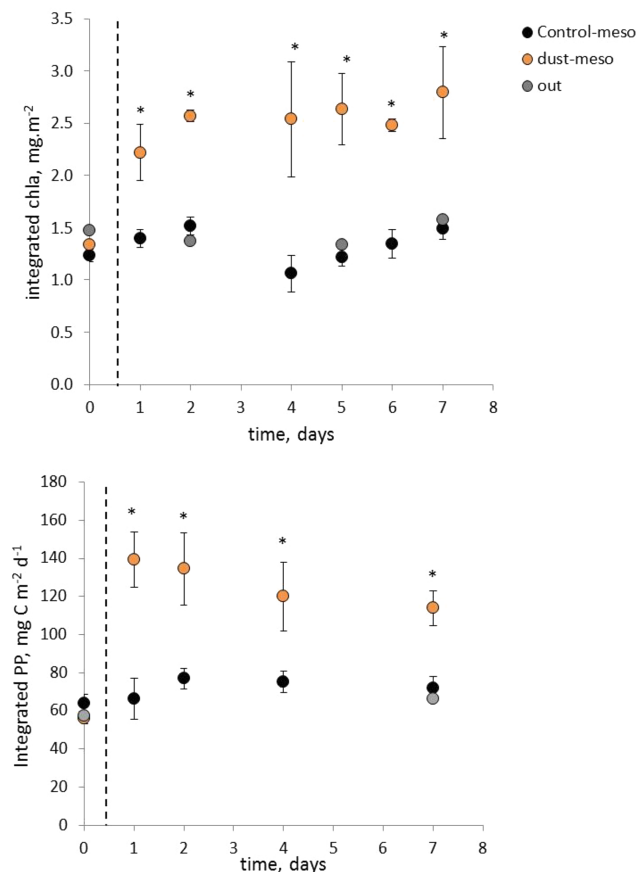
Figure 2. Mean integrated chlorophyll *a* (Chl *a*) in mg m^{-2} (top panel) and mean integrated primary production (PP) in mg C m^{-2} d^{-1} (bottom panel) over mesocosms during the DUNE-1-P experiment in the Control-meso (black dot), Dust-meso (orange dot) and out (grey dot). The dotted line represents the time of the dust seeding. Data in the Control-meso and Dust-meso represent the average and standard deviations 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.

The increase in Chl *a* was observed for 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 (Supplement 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. 2). Twenty-four hours after the 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 in depth were similar ($p > 0.05$, Supplement Fig. S4).
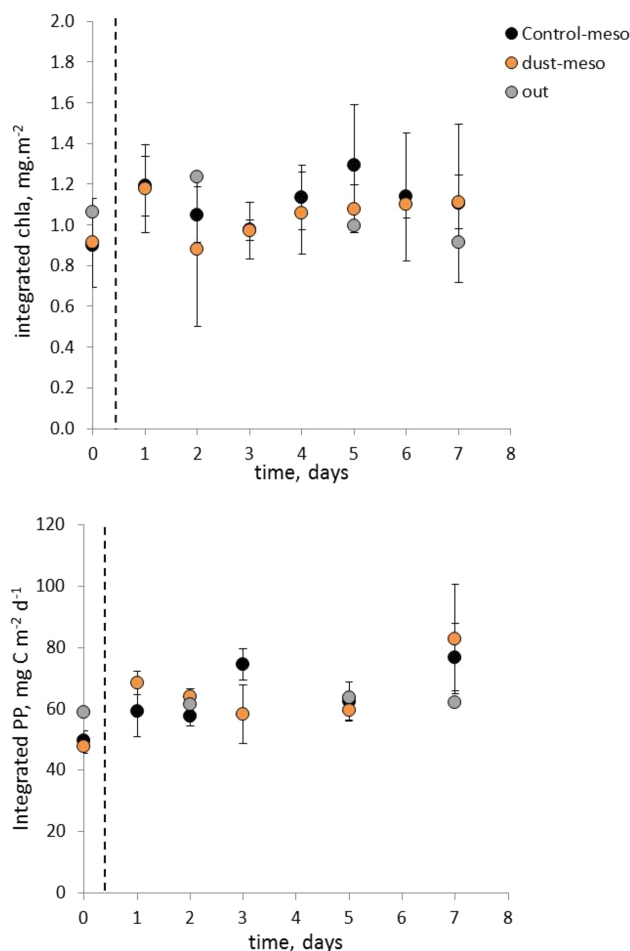
Figure 3. Mean integrated chlorophyll *a* (Chl *a*) in mg m^{-2} (top panel) and mean integrated primary production (PP) in mg C m^{-2} d^{-1} (bottom panel) over mesocosms during the DUNE-1-Q experiment in the Control-meso (black dot), Dust-meso (orange dot) and out (grey dot). The dotted line represents the time of the dust seeding. Data in the Control-meso and Dust-meso represent the average and standard deviations of the three replicate mesocosms.

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. 3, S5, and S6). A slight stimulation of PP (1.2-fold, $p < 0.05$) was detected in the Dust-meso at 5 m in depth, 48 h after dust addition (Supplement Fig. S6).

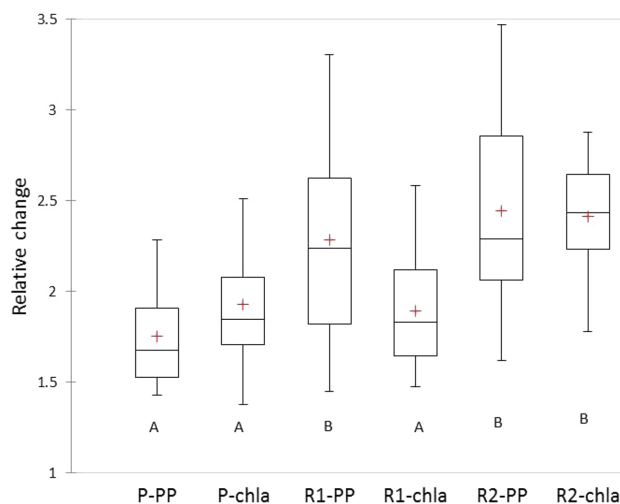## Response of the phytoplanktonic community to two successive wet deposition events

In the DUNE-2-R experiment, the first seeding (R1) led to averaged increases in integrated Chl *a* and PP of x1.9 and
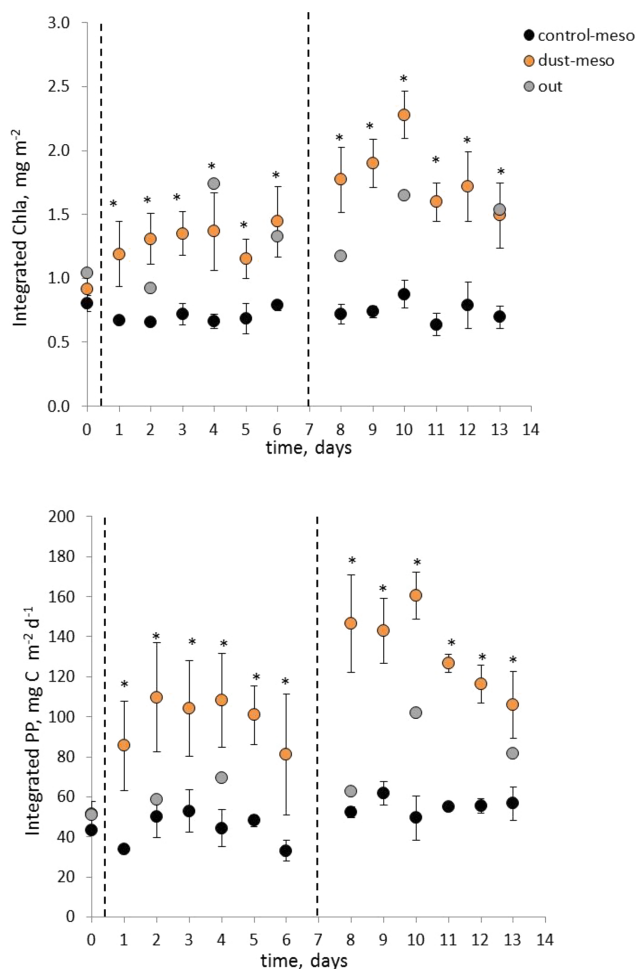
Figure 4. Mean integrated chlorophyll a (Chl a) in mg m^{-2} (top panel) and mean integrated primary production (PP) in mg C m^{-2} d^{-1} (bottom panel) over mesocosms during the DUNE-2-R1 and DUNE-2-R2 experiments in the Control-meso (black dot), Dust-meso (orange dot) and out (grey dot). The dotted lines represent the time of the dust seedings. Data in the Control-meso and Dust-meso represent the average and standard deviations 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.

Figure 5. 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-1-P, DUNE-2-R1 and DUNE-2-R2 experiments. Means that were not significantly different between the different parameters and experiments ($p > 0.05$) are labeled with the same letter.
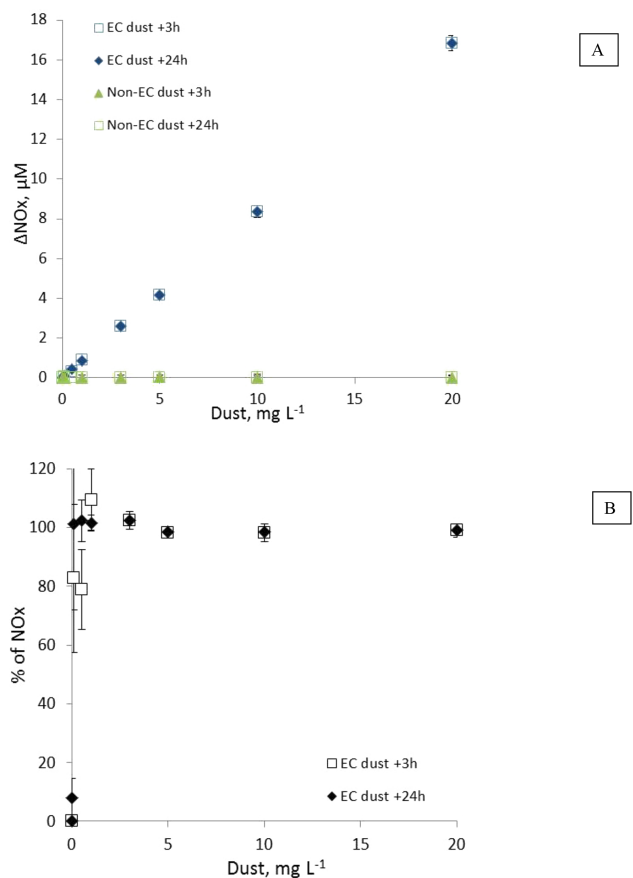
Chl a and PP in the Dust-meso were twice as high ($p < 0.05$) as that in the unamended controls.

Comparison of the relative changes between DUNE-1-P, DUNE-2-R1 and DUNE-2-R2 experiments

The mean relative Chl a change (RC, Dust/Control) was similar after the wet deposition event simulated in the DUNE-1-P and DUNE-2-R1 experiments (RC$_{Chla}$ = 1.9, $p > 0.05$, Fig. 5), while the RC in PP was higher during DUNE-2-R1 (RC$_{PP}$ = 2.3) than during DUNE-1-P (RC$_{PP}$ = 1.8, $p < 0.05$). The combination of two wet deposition events (DUNE-2-R2) resulted in a higher relative Chl a change (RC$_{Chla}$ = 2.4, $p < 0.05$) compared to that induced by the first seeding (R1, RC$_{Chla}$ = 1.9). For PP, the relative change was similar over DUNE-2-R1 (first seeding) and over DUNE-2-R2 (combination of two seedings) ($p > 0.05$; RC$_{PP}$ = 2.3 and 2.4, respectively; Fig. 5). The relative changes in Chl a and PP were similar in the DUNE-1-P experiment ($p > 0.05$; RC$_{Chla}$ = 1.9 and RC$_{PP}$ = 1.8), as well as over DUNE-2-R2 ($p > 0.05$; $RC_{Chla} = RC_{PP} = 2.4$), while over DUNE-2-R1, the relative increase in PP (RC$_{PP}$ = 2.3) was higher than that in Chl a (RC$_{Chla}$ = 1.9).

3.4 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

x2.3 relative to the Control-meso over the duration of DUNE-2-R1 (Fig. 4, Supplement Figs. S7 and S8). Six days after the first seeding, Chl a and PP in the Dust-meso were still at least twice as high ($p < 0.05$) as in the Control-meso. Twenty-four hours after the second seeding, an additional x1.8 stimulation of PP was observed. The combination of the two seedings induced a x2.4 increase in both integrated Chl a and PP relative to the Control-meso over the duration of DUNE-2-R2 (Fig. 4, Supplement Figs. S7 and S8). Over the 13-day duration of the DUNE-2-R experiment, the Chl a increase in the Dust-meso was observed over the entire mesocosm (Supplement Fig. S7). At the end of the experiment (day 13),

Figure 6. Results of abiotic dissolution experiments: **(A)** ΔNO_x ([NO_x]$_{after\ the\ introduction\ of\ dust}$ − [NO_x]$_{initial}$) in μM relative to the amount of dust (EC and non-EC dust) introduced in seawater for contact times of 3 and 24 h, and **(B)** percentage of NO_x released from EC dust (NO_x % = $\Delta NO_x \times 100/N_{total\ dust}$) as a function of the dust amount introduced in seawater for contact times of 3 and 24 h. Data represent the average and standard deviations of the three replicates.

were observed concomitantly with increasing EC dust concentrations for both contact times (Fig. 6a). 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. 6b). Additions of non-EC dust did not significantly increase ($p > 0.05$) NO_x concentrations relative to the unamended artificial seawater (Fig. 6a).

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).

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 wet deposition events (P, R) induced significant stimulation of both Chl a and PP, from 1.8- to 2.4-fold (Fig. 5), 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 (maxima of 0.23 μg Chl a L^{-1} and 13.1 mg C m^{-3} d^{-1} in the Dust-meso). PP and Chl a in the Dust-meso were higher over DUNE-2-R2 (combination of two seedings) than over DUNE-2-R1 (first seeding) (Fig. 4). The cumulated effect of the two seedings observed over DUNE-2-R2 led to a higher Chl a increase (R2, RC = 2.4) relative to the first one (R1, RC = 1.9, Fig. 5). During the first dust addition, 65 % of the increase in total phytoplanktonic biomass was indeed due to picophytoplankton (< 3 μm), while over DUNE-2-R2, the nanophytoplankton and microphytoplankton (> 3 μm) contributed to about 50 % of the Chl a increase (Giovagnetti et al., 2013). The rapid response of picophytoplankton after the R1 seeding fits their dominance and ability to outcompete bigger cells under LNLC and high light conditions, by having a higher efficiency in physiological processes in resource-limited habitats when compared to the larger ones (Raven et al., 2005). Surprisingly, no increase in Chl a was observed after the simulated dry deposition event (DUNE-1-Q experiment, non-EC dust). Previous results from bioassay experiments in the western Mediterranean Sea during stratification periods have also shown that the addition of the untreated Saharan dust analog (simulated dry deposition

event of 2.5–12.5 g m^{-2}) did not induce any significant Chl a change (Ridame 2001, Bonnet et al., 2005). Despite the lack of a detectable response of Chl a after the DUNE-1-Q seeding, the 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., 2014; Guieu et al., 2014b). 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 in depth, $p < 0.05$, Supplement Fig. S6) and in *Synechococcus* abundance ($+30\%$ at 5 m in depth, $p < 0.05$, unpublished data) were recorded after the DUNE-1-Q seeding. The abundance of copepods representing the main contributor to zooplankton also increased slightly in the Dust-meso at the end of the DUNE-1-Q experiment relative to the Control-meso (L. Stemmann, unpublished data, 2013). The increase in the POC export without an associated increase in the biogenic stock could also be explained by the lithogenic ballast effect through aggregation and/or DOC sorption processes on the settling particles (Desboeufs et al., 2014; Bressac et al., 2014).

4.2 Impact of the pathway of deposition on the nutrient dynamics

Differences in the atmospheric supply of bioavailable new nutrients depending on the pathway of deposition (wet or dry) may explain the variability in the response of the phytoplanktonic community (Table 3). In the DUNE-1-P and DUNE-2-R experiments, the seeding of the mesocosms was performed with the EC dust mixed with ultrapure water in order to mimic a wet deposition event (and thus cloud processes, including the mixing of particles with compounds from anthropogenic activities such as HNO$_3$; Desboeufs et al., 2014), whereas in DUNE-1-Q, non-EC dust mixed with seawater was used to simulate a dry-type 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; Desboeufs, personal communication, 2013; Aghnatios et al., 2014). Nevertheless, the dissolution of P associated with Saharan dust has been shown to be about two- to five-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 (Pulido-Villena et al., 2010, 2014) and no changes after the simulated dry deposition event (Pulido-Villena, personal communication, 2013), while abiotic dissolution exper-

iments 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 used quickly by the biological activity, explaining why no increase in DIP concentrations was recorded after the DUNE-1-Q seeding. Indeed, the dry deposition event in DUNE-1-Q induced a strong stimulation of N$_2$ fixation (Ridame et al., 2013) and bacterial respiration (Guieu et al., 2014b) (Table 3).

NO$_3^-$

The N content of the EC dust was about ten-fold higher compared to the non-EC dust, resulting in significant changes in the atmospheric supply of dissolved inorganic nitrogen (DIN), depending on the pathway of deposition. Abiotic dissolution experiments showed that EC dust – mimicking Saharan dust wet deposition (DUNE-1-P, DUNE-2-R) – was a significant source of NO$_x$, whereas non-EC dust – mimicking Saharan dust dry deposition (DUNE-1-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 DUNE-2-R2 wet deposition events (Fig. 1, Table 3). Assuming a homogeneous distribution of the EC dust within the Dust-meso, this would lead to a particulate dust concentration of about 1 mg L^{-1}, resulting in a potential input of 0.9 µM NO$_3^-$ (Fig. 6a). The vertical profiles of NO$_3^-$ in the mesocosms indicate a maximum net increase in NO$_3^-$ of $+9.8$ µM at the surface soon after the R2 seeding (relative to day 6 in the Dust-meso) (Fig. 1a), 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, 2014), and by the vertical distribution of crustal elements as aluminum (Desboeufs et al., 2014). Based on the dissolution results (Fig. 6), a complete dissolution of NO$_3^-$ associated with EC dust is expected within 3 hr after the additions, resulting in theoretical inputs of atmospheric new N of 8.5, 9.7 and 9.7 mmol NO$_3^-$ m^{-2} in the Dust-meso, respectively, for DUNE-1-P, DUNE-2-R1 and DUNE-2-R2. Over DUNE-2-R, the successive net increases in the NO$_3^-$ stock just after seedings (R1: $+10.2$ mmol NO$_3^-$ m^{-2} and R2: $+10$ mmol NO$_3^-$ m^{-2}; Fig. 1b) are on the same order of magnitude as the input of atmospheric NO$_3^-$ based on total dissolution of N associated with 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 in N-depleted waters, as strong stimulations of N$_2$ fixation (up to x5.3, Table 3) were detected after all the DUNE seedings (Ridame et al., 2013). The increases in the N$_2$ fixation rates led to net inputs of new N of $+39$, $+15$ and

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; and Wuttig et al. (2013).

| DUNE-1-P | DUNE-1-Q | DUNE-2-R1 | DUNE-2-R2 |
Wet	Dry	Wet	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.

$+22 \,\mu\text{mol N m}^{-2}$, respectively, after the DUNE-1-P, DUNE-2-R1 and DUNE-2-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 $9.7 \,\text{mmol NO}_3^- \,\text{m}^{-2}$). The increase in NO_3^- concentrations observed after simulated wet deposition events could thus 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 (x3.3–5.2 increases in N_2 fixation after dust addition, in Ridame et al., 2013), as dry deposition of non-EC dust was shown to be a negligible source of NO_3^-. The slight increase in PP 48 h after the DUNE-1-Q seeding (at 5 m in depth, $+20\%$), converted to N with a C : N ratio of 7.6 (measured during DUNE-1-Q), required an input of $\sim 8 \,\text{nmol N L}^{-1} \,\text{d}^{-1}$, which was higher than the input from increased N_2 fixation (~ 1–$2 \,\text{nmol N L}^{-1} \,\text{d}^{-1}$).

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; Desboeufs, personal communication, 2013; Aghnatios et al., 2014). 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 concentrations and contact times (Desboeufs, personal communication, 2013; Aghnatios et al., 2014). 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, DUNE-1-Q and DUNE-2-R1 seedings, due to DFe scavenging on settling dust particles 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^- stocks in the mesocosms, as well as significant stimulations of PP and Chl a (Table 3). On the opposite end, 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 atmospheric deposition clearly controls the response of the phytoplanktonic community (biomass and productivity) through the differences in the new nutrient supply. As wet deposition is the main pathway of Saharan dust deposition over the western Mediterranean Sea (e.g., Loÿe-Pilot and Martin 1996), atmospheric deposition is probably the main source of new nitrogen (NO_3^-) (Markaki et al., 2010) and new phosphorus (DIP) to the open Mediterranean surface waters during stratification periods.

4.3 Nutrient factors controlling primary production and biogeochemical consequences

Iron

Before seedings, DFe concentrations ranged from 2.3 to 3.3 nM in the tested waters (Table 2). After the DUNE-1-P, DUNE-1-Q and DUNE-2-R1 seedings, DFe concentrations dropped to about 1.5–2 nM, while primary production was either stimulated (DUNE-1-P, DUNE-2-R1) or unchanged (DUNE-1-Q) (Table 3). Wagener et al. (2010) showed that enhanced biological activity cannot explain by itself the observed decrease in DFe after the wet deposition event simulated during DUNE-1-P. Instead, the sink of DFe was likely due to scavenging on settling dust particles and aggregates in the Dust-meso. Despite opposite changes in DFe concentration after the DUNE-2-R1 (decrease) and DUNE-2-R2 (increase) seedings (Table 3), primary production increased after both additions with a similar magnitude (Fig. 5). This is evidence 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. This
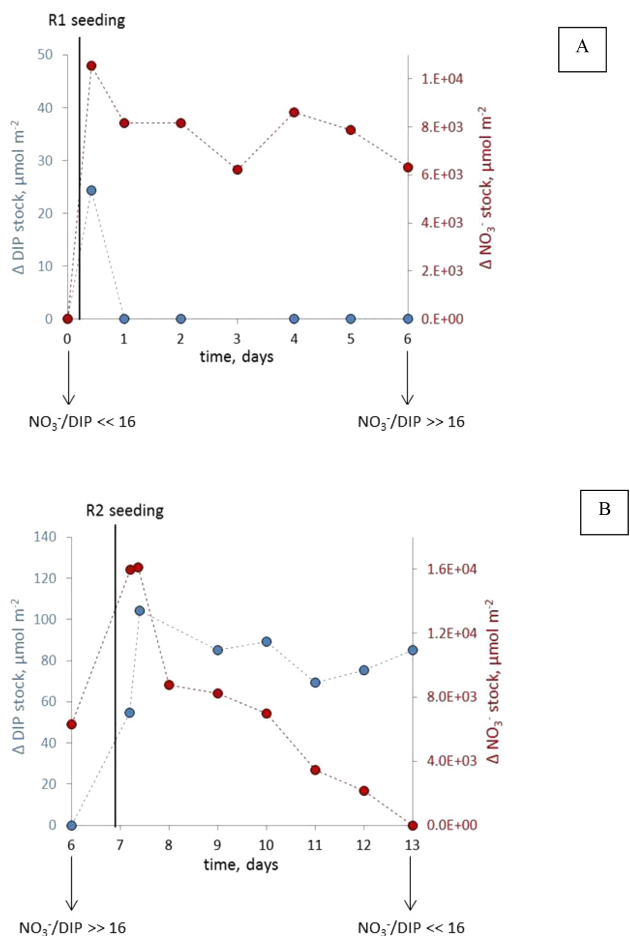
Figure 7. Averaged temporal changes in the variation (Dust-Control) in NO_3^- (red dots) and DIP stocks (blue dots) **(A)** after the DUNE-2 R1 seeding and **(B)** after the DUNE-2 R2 seeding.

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 suggests that the phytoplanktonic community may be limited by N and P, or be 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 phytoplanktonic activity. As the DIP concentration was also initially extremely low (5 nM, Table 2), PP could likely be 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 co-limitation 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 dynamics and by altering the ambient NO_3^-/DIP ratio (Fig. 7). 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 higher than 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 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 activity (PP) to a wet event, as well as in the evolution of the DIP concentration (and likely of the NO_3^- 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 less than 6 (initially) to greater than 150 at the end of the R1 experiment (Fig. 7a), suggesting a switch from an initial N limitation or NP co-limitation of the phytoplanktonic activity towards a severe P limitation until the end of R1. The 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 $+18 \,\mu mol\, DIP\, m^{-2}$ for DUNE-1-P and DUNE-2-R1 at day 1) was on the same order of magnitude as the net increase in the DIP stock in the Dust-meso ($+ 21$ and $+ 24 \,\mu mol\, DIP\, m^{-2}$ 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 the DUNE-1-P and DUNE-2-R1 seedings can explain the rapid DIP depletion (Fig. 7a). Bacterial respiration was also stimulated after these seedings (Pulido-Villena, 2010, 2014), suggesting a competition for the DIP uptake between heterotrophic bacteria and autotrophic phytoplankton, as already reported during microcosm experiments in the tropical Atlantic and the Mediterranean Sea (Thingstad et al., 2005; Maranon et al., 2010). As the $\sim$ 2-fold increase in PP and Chl a in the Dust-meso lasted for 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 for 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.

DUNE-2-R2

Over DUNE-2-R2, the temporal evolution of NO_3^- and DIP stocks was different relative to that observed over DUNE-2-R1 (Fig. 7). The second seeding (R2) permitted an additional supply of new nitrogen and phosphorus into the mesocosms (Fig. 7). Then, due to the biological activity, the NO_3^- stock in the Dust-meso decreased to undetectable values at the end of R2, while the DIP stock remained much higher than in the controls. This led 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 DUNE-2-R2, Giovagnetti et al. (2013) showed a change in the size structure of the phytoplanktonic community towards a larger dominance of big cells ($> 3\,\mu m$). Larger-sized cells need further nutrient supply in order to be able to adjust their physiology and compete for resource acquisition and biomass increase (Raven et al., 2005; Irwin et al., 2006). After the DUNE-2-R2 seeding, the nutrient (NO_3^-, DIP) concentrations were indeed higher than after the first seeding, which can explain why the response of larger cells was mainly relevant over DUNE-2-R2. This change in the size structure of the phytoplanktonic community is consistent with a higher NO_3^- consumption (Giovagnetti et al., 2013), as large cells have higher half-saturation constants (Aksnes and Egge, 1991; Hein et al., 1995) and higher cellular metabolic requirements than small cells (Grover, 1991).

The DIP stock surprisingly remained quite stable and significantly much higher relative to the controls until the end of R2, despite twice as high 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 for by an equivalent rate of DIP production, 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 the 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 DUNE-2-R2, heterotrophic bacteria did not compete with phytoplankton for the P resource, as is suspected for DUNE-2-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 DUNE-2-R2.

Consequences of dust deposition for 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 Raimbault, 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). The increase in PP in the Dust-meso 24 h after seeding observed in the DUNE-1-P and DUNE-2-R experiments can thus be associated with NP. We consider that, after 24 h, the increase in PP could be mainly supported by regenerated nutrients. Based on the estimates of the NP in the Dust-meso 24 h after the DUNE-1-P and DUNE-2-R seedings from Ridame et al. (2013), the contribution of NP to PP increased in the Dust-meso from initially 15 % to 60–70 % 24 h after seeding. Wet dust deposition events 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 wet atmospheric dust wet deposition does greatly influence primary productivity and algal biomass in LNLC environments through the atmospheric supply of new nutrients. 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 input of bioavailable new nutrients depending on the pathway of dust deposition (wet or dry) led to contrasting responses of the phytoplanktonic community. Primary production increased significantly only after wet deposition events. In addition to being a source of DIP, wet deposition due to cloud processes and mixing with anthropogenic species such as HNO_3 also represent a significant source of NO_3^-, inducing a significant increase in the ambient NO_3^-/DIP ratio soon after deposition. 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, 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., 2014; Bressac et al., 2014), Guieu et al. (2014b) 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 in 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.

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