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Introduction to the project DUNE, a DUst experiment in a low Nutrient, low chlorophyll Ecosystem

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Abstract

The main goal of the project DUNE was to estimate the impact of atmospheric deposition on an oligotrophic ecosystem based on mesocosm experiments simulating strong atmospheric inputs of Aeolian dust. Atmospheric deposition is now recognized

- as a significant source of macro- and micro-nutrients for the surface ocean, but the quantification of its role on the biological carbon pump is still poorly determined. We proposed in DUNE to investigate the role of atmospheric inputs on the functioning of an oligotrophic system particularly well adapted to this kind of study: the Mediterranean Sea. The Mediterranean Sea – etymologically, sea surrounded by land – is
- ¹⁰ submitted to atmospheric inputs that are very variable both in frequency and intensity. During the thermal stratification period, only atmospheric deposition is prone to fertilize Mediterranean surface waters which has become very oligotrophic due to the nutrient depletion (after the spring bloom). This paper describes the objectives of DUNE and the implementation plan of a series of mesocosms experiments during which either wet
- or dry and a succession of two wet deposition fluxes of 10 gm⁻² of Saharan dust have been simulated. After the presentation of the main biogeochemical initial conditions of the site at the time of each experiment, a general overview of the papers published in this special issue is presented, including laboratory results on the solubility of trace elements in erodible soils in addition to results from the mesocosm experiments. Our
- 20 mesocosm experiments aimed at being representative of real atmospheric deposition events onto the surface of oligotrophic marine waters and were an original attempt to consider the vertical dimension in the study of the fate of atmospheric deposition within surface waters. Results obtained can be more easily extrapolated to quantify budgets and parameterize processes such as particle migration through a "captured"
- ²⁵ water column". The strong simulated dust deposition events were found to impact the dissolved concentrations of inorganic dissolved phosphorus, nitrogen, iron and other trace elements. In the case of Fe, adsorption on sinking particles yields a decrease in dissolved concentration unless binding ligands were produced following a former



deposition input and associated fertilization. For the first time, a quantification of the C export induced by the aerosol addition was possible. Description and parameterization of biotic (heterotrophs and autotrophs, including diazotrophs) and abiotic processes (ballast effect due to lithogenic particles) after dust addition in sea surface water, result
 ⁵ in a net particulate organic carbon export in part controlled by the "lithogenic carbon pump".

1 Context and objectives

Nutrient cycling controls in part the efficiency of the biological carbon pump through which CO₂ is consumed in ocean surface waters and transported to the deep sea as sinking particulate organic carbon (Moore et al., 2013). The interactions between atmo-10 sphere and ocean play a key role if we are to understand processes governing nutrient cycles in the ocean and to which extent climate change plays a role in this scheme. In this context, the scientific community pays special attention to atmospheric input of nutrients, which can alter community structure and nutrient cycling in the oceans and, therefore, modify the efficiency of the ocean to store atmospheric CO_2 (Law et al., 15 2013). Atmospheric deposition is now recognized as a significant source of iron (Fe) and other nutrients for surface waters of the global remote ocean (Duce et al., 1991; Jickells et al., 2005; Moore et al., 2013) as well as of the Mediterranean (e.g. Bergametti et al., 1992; Quétel et al., 1993; Ridame and Guieu, 2002; Bonnet and Guieu, 2006; Guieu et al., 2010a; Markaki et al., 2010). An unresolved issue in ocean and 20 climate sciences is whether changes of the input to the surface ocean can alter the flux of carbon to the deep ocean (Law et al., 2013). This question is of particular interest in the Mediterranean region, a regional hot spot of the global climate change (Giorgi et al., 2006) which, as the global ocean, is already experiencing a positive trend in both sea surface and deep-water temperatures (MERMEX Group, 2011). 25

While the impact of Fe on productivity has been recognized in high nutrient, low chlorophyll (HNLC) oceanic regions through both bioassay experiments



(Martin et al., 1994), mesoscale artificial iron fertilization experiments (see the syntheses by de Baar et al., 2005; Boyd et al., 2007) and natural iron fertilization (Blain et al., 2007), the ecological and biogeochemical effects of atmospheric dust-derived Fe and macronutrients (N, P) in oligotrophic, i.e. low nutrient, low chlorophyll (LNLC) environments are still poorly understood and resulting C export via fertilization and aggregation processes is not quantified. And yet, these LNLC oceanic 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 Mediterranean Sea is a typical LNLC region, particularly well adapted to tackle the question of the role of atmospheric input: it is an oligotrophic, quasi-enclosed basin, which receives a noticeable atmospheric flux of Aeolian soil dust, mainly derived from the Sahara desert, in the form of strong pulses (Loÿe-Pilot et al., 1986; Bergametti et al., 1989; Guerzoni et al., 1999). At the DYFAMED (DYnamique des Flux Atmosphériques en MEDiterranée) site in the remote northwestern Mediterranean,

- the sedimentation flux from surface waters of particulate manganese and aluminium corresponds to the atmospheric deposition flux on an annual time scale (Davies and Buat-Ménard, 1990), and most of the iron associated with organic matter sinking from surface waters is provided by atmospheric input (Quétel et al., 1993). Moreover, the Mediterranean basin continuously receives anthropogenic aerosols from industrial and
- ²⁰ domestic activities from populated areas around the basin and other parts of Europe, as well as seasonal inputs from biomass burning (Remoudaki et al., 1991a, b; Migon et al., 1991; Bergametti et al., 1992; Guieu et al., 2005, 2010a). As described earlier (MERMEX group) the mixing of those natural particles with anthropogenic aerosols might have a positive fertilization effect on biota by bringing both bioavailable-N and
- P to the Mediterranean surface waters, particularly during the stratification period. Indeed, when the water column is stratified and characterized by a low primary productivity, phosphate and nitrate concentrations are very low (under detection limits of standard methods, namely 20 and 50 nM respectively) and biological activity in the surface waters encounters nitrate limitation and nitrate-phosphate co-limitation



(Tanaka et al., 2011). Concerning dissolved iron, it becomes depleted after the spring bloom and could affect temporarily productivity with concentrations as low as 0.2 nM (Sarthou and Jeandel, 2001; Bonnet and Guieu, 2006). Throughout this stratified period, the atmosphere becomes the main external source for the marine mixed layer of both N and P (e.g., Loÿe-Pilot et al., 1990; Bergametti et al., 1992; Migon and Sandroni, 1999; Ridame and Guieu, 2002; Herut et al., 1999; Kouvarakis et al., 2001; Markaki et al., 2003; Krom et al., 2004) and iron (Guieu et al., 2002a; Bonnet and Guieu, 2006). Dulac et al. (1996) illustrate from Quétel's (1991) data the strong impact of a Saharan dust deposition event on the particulate iron concentration profile
in the surface layer at the DYFAMED station in the northwestern Mediterranean. Via

- theoretical calculation and/or bioassay experiments, it has been shown experimentally that those inputs can impact both heterotrophic (Thingstad et al., 2005; Pulido-Villena et al., 2008) and autotrophic (including diazotrophs) production in the Mediterranean Sea (Klein et al.,1997; Migon and Sandroni, 1999; Ridame and Guieu, 2002; Bonnet
- et al., 2005; Ridame et al., 2011; Ternon et al., 2011), underlining their capacity to relieve the macro- and micro-nutrient limitations encountered in this area. Other effects different from direct fertilization effect, such as POC fluxes mediated by organic-mineral aggregation have been shown to be very significant in the Mediterranean Sea: for example, the extreme Saharan event of February 2004 representing a dust flux of the or-
- der of 22 gm⁻² in some locations in Corsica and the French Riviera (Bonnet and Guieu, 2006; Ternon et al., 2010), allowed to export at 200 m in the Ligurian Sea (DYFAMED station) 45% of the total annual POC compared to an average of 25% for the whole bloom period that same year. This emphasizes the need to understand the role played by such high Saharan dust deposition in the carbon export efficiency both thanks to in-
- ²⁵ creased production induced by the input of new nutrients and through organic-mineral aggregation and ballast effect in a dust-rich area such as the NW Mediterranean Sea.

In this context, the main objective of DUNE was to follow the impacts of a well characterized atmospheric input on an oligotrophic ecosystem in order to answer the following questions:



- How the introduction of atmospheric particles does impact the cycle of chemical elements of biogeochemical interest: mainly C, P, N, Fe.
- What is the response of viruses, bacteria, phytoplankton, zooplankton in terms of abundance, activity and diversity?
- How the temporal pattern of the particulate export is modified by the introduction of atmospheric particles?
 - Does a same deposition flux reproduce or not the same effects and why?
 - Do dry and wet deposition act the same way?

In spite of previous efforts, answers to the question of the biological response of LNLC
 regions and, particularly, of the Mediterranean Sea to atmospheric inputs are still fragmented. To cover this gap, we proposed in DUNE to study the effect of atmospheric input on the oligotrophic Mediterranean ecosystem through artificial Aeolian aerosol additions over in situ large mesocosms representative of a significant body of the surface waters in perfectly controlled conditions: such an approach is indeed the strength of the project and was possible thanks to the strong and effective partnership between atmospheric and oceanic scientists partners of DUNE (Guieu et al., 2010b).

2 The mesocosm strategy

Our experiments rely on seeding of African dust in mesocosms in order to reproduce an intense deposition event and to follow its biogeochemical impact in marine surface water over about a week. Microcosm experiments are commonly used to study the impact of atmospheric deposition on the biogeochemistry of surface waters (Guieu et al., 2013b and ref. within). Because of the small volumes involved in the microcosm approach (usually a few liters at maximum), confinement issues can rapidly occur and thus the experimental duration is often a limit (usually 2 days and at max 6 days,



Guieu et al., 2013b). Such small volumes also limit the number of parameters that can be measured in stocks and fluxes. An important limit is also that microcosms rely on a fixed and homogeneously-distributed concentration of aerosols. Mesocosms present the advantage of enabling studies of processes both as a function of depth and time while the atmospheric particles are sinking. Indeed, the particularity of the atmospheric

- ⁵ while the atmospheric particles are sinking. Indeed, the particularity of the atmospheric input being that it is associated with a significant particulate flux after it has reached the sea surface (this is particularly true for desert dust deposition events), one can expect that there will be different processes such as adsorption/desorption, release of new nutrients, biological uptake, aggregation processes that will take place during the
- ¹⁰ course of the particles towards the depth. During DUNE, sampling in quick succession at different depths of the mesocosms allowed the measurement of a number of stocks (such as nutrients including trace metals, phytoplankton, zooplankton and heterotrophic bacteria biomass) and fluxes (primary production, respiration, N₂ fixation, export of particulate organic matter). Figure 1 summarizes all the measurements that
- ¹⁵ are possible in the mesocosms at different depth of the captured column water thanks to the large volume considered (52 m³ in this work). The derived parameterizations can constrain a biogeochemical model that takes into account the atmospheric deposition in oligotrophic conditions.

The mean annual dust deposition flux in Corsica during the period 1984–1994 was 12.5 gm⁻² and was shown to be attributed mainly to pulses > 1 gm⁻² (Loÿe-Pilot and Martin, 1996). Unusually strong events with short duration (few hours) have been recorded over the past decade with African dust deposition fluxes as high as 22 gm⁻² in November 2001 (Guieu et al., 2010a), and in February 2004 (Bonnet and Guieu, 2006; Ternon et al., 2010). During DUNE, we chose to mimic a high, but still realistic, Saharan dust deposition event of 10 gm⁻² into mesocosms. In order to reproducing a real Saharan dust deposition by the mean of a controlled seeding in seawater, it has first been necessary to produce hundreds of grams of particulate material identical to aerosols depositing at the surface of the ocean. This was achieved by experimental simulation of (i) the production of desert aerosols and (ii) the chemical aging mimicking



their transport and cloud processing in the atmosphere. Wet and dry depositions were mimicked during different experiments as there is so far no knowledge/evidence on potential differences in the impact of those different types of deposition on the biogeochemistry. The second part of the methodological development concerned the actual
 ⁵ conception of seeding experiments in large clean mesocosms. With a list of specifications (e.g. those systems should be transportable, totally made with plastic material

- to avoid any contamination considering the expected low concentrations of nutrients, etc.), the DUNE team worked on the concept of holding structures, enclosure (52 m³), sampling systems, anchoring. Both methodologies of dust aerosol and clean meso-
- cosm production have been successfully developed in the frame of the DUNE project. The detailed methodological approaches have been published in Guieu et al. (2010b). Artificial seeding over large mesososms have been realized during two campaigns in the preservation area of Scandola in Corsica: DUNE-1 experiment in 2008 and DUNE-2 experiment in 2010. The experimental site in the Preservation Area of Scandola –
 a typical low-nutrient low-chlorophyll area where the deployments took place is also described in Guieu et al. (2010b). The different steps of the DUNE approach are summarized on Fig. 2 and were followed during DUNE-1 and DUNE-2 (Fig. 3):

– During DUNE-1 in June 2008 we performed two distinct 8-day experiments: a first simulation of a Saharan wet deposition event (hereafter named "DUNE-P") and a second simulation of a Saharan dry deposition event (hereafter named "DUNE-Q"). Mesocosms were emptied and redeployed between the two experiments. DUNE-P and DUNE-Q consisted in the deployment of 3 mesocosms hereafter named "CONTROL" not subjected to any dust addition and used as a reference, and 3 mesocosms hereafter named "DUST" receiving a dust addition corresponding to a dust flux of 10 gm⁻². The wet deposition event during DUNE-P (11–18 June 2008) was mimicked by seeding evapocondensed (EC) dust over the DUST mesocosms, and the dry deposition event during "DUNE-Q" (20–27 June 2008) was mimicked by seeding non-evapocondensed (non-EC) dust (Guieu et al., 2010b).



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- During DUNE-2 in 2010 (26 June-9 July 2010): we performed a single 16-day experiment (hereafter named "DUNE-R") that consisted in 2 successive dust wet deposition simulations (using the same amount of EC-dust) with 7 days between each seeding (respectively "R1" and "R2"). DUNE-R consisted in the deployment of 3 mesocosms CONTROL (no dust addition) and 4 mesocosms DUST (each with dust addition corresponding to a dust flux of 10 gm^{-2}), 3 of them being devoted to biogeochemical studies like in DUNE-P and -Q, the additional mesocosm being dedicated to optical measurements. This strategy of two successive seedings was decided following DUNE-1 results. Indeed, Wagener et al. (2010) showed that dust addition during DUNE-P was followed by a decrease of dissolved iron (dFe) concentration likely due to dFe scavenging on settling dust particles giving evidence that large dust deposition events may be a sink for surface ocean dissolved iron. Such an effect has also been reported for dissolved thorium (Lambert et al., 1991). Combining the mesocosm experiment with a batch dissolution experiment. Wagener and collaborators have then shown that dissolved iron increased again after enhancement of biological activity following dust addition once Fe-binding ligands have been produced. The two successive seeding during DUNE-2 were planned in order to explore how dust deposition does impact biogeochemistry under different in situ biogeochemical initial conditions.

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 In addition, water outside of the mesocosms was sampled during the course of all the experiments for reference.

The bulk erodible soil material used to produce added dust was collected in a same dry river bed in the desert area of Tunisia during two sampling campaigns in 2007 and 2009 for the DUNE-P/Q and R experiments respectively. In consequence, the dust used during DUNE-1 and DUNE-2 had similar chemical composition for nutrients i.e. P, Fe and C, except for N (Table 1 and Desboeufs et al., 2013). Indeed, a difference of one order of magnitude in the N content between the composition of the non-EC dust (used as a proxy for dry deposition) and the EC-dust (used as a proxy for wet deposition) is



due to the addition of HNO_3 in the simulated cloud water used to process dust for wet deposition simulation (Guieu et al., 2010b). The ways the dust was seeded and the mesocosms sampled are fully described in Guieu et al. (2010b).

Seawater sampling was done every day at the same hours. The schedule for the 3 experiments is reported in Table 2. During DUNE-1, for the discrete sampling, 3 depths were sampled with the Teflon pump: sub-surface, 5 m and 10 m. Following the results obtained for dissolved iron (Wagener et al., 2010), it was decided during DUNE-2 to increase the number of sampled depths for 2 of the DUST-meso and 2.5 m, 7.5 m and 12.5 m depths were also sampled for trace elements. During DUNE-2, the sediment traps were changed every day instead of every two days during DUNE-1.

3 Main environmental initial conditions and evolution

Because of its weak cloud coverage, the Mediterranean Sea is subject to stronger solar radiation in comparison with oceanic areas of similar latitude (i.e. Vasilkov et al., 2001), with the consequence that the percentage of sunshine duration over the whole day light is close to 100% in particular in summer. The period to perform the experiments was chosen in order to have such summer condition with well stratified column water characterized by very poor concentration of nutrients. DUNE-1 and DUNE-2 were thus scheduled in June–July. The temperature was constantly monitored inside and outside the mesocosms and the temperature inside the mesocosm was not significantly dif-

- ferent from outside indicating that no confinement effect has occurred (Fig. 4a). The seawater temperature during the DUNE-P experiment was lower with smaller daily amplitude than during the other experiments. Seawater temperature along with the air temperature (recorded by MeteoFrance at the nearby station in Calvi) are represented in Fig. 4b. Also, the gradient between the temperature recorded at surface and 10 m is presented in Fig. 4c in order to represent in a first approximation the thermal stratifica-
- presented in Fig. 4c in order to represent in a first approximation the thermal stratification (Longhurst et al., 1995).



While the seawater temperature during the whole DUNE-P experiment was in the range 17–21.5 °C and the stratification of the column water inside the mesocosms was not marked, temperature significantly increased during the following DUNE-Q experiment with a range 18.5–26 °C. The air temperature during daytime was stable during 5 DUNE-P with an average value of 21.7 ± 1.8 °C while the percentage of sunshine duration could be as low as 5 % for some day (Fig. 4d); the air temperature increased rapidly at the beginning of the DUNE-Q experiment to reach an average value during the day of 26.3 ± 2.6 °C with light duration close to the maximum, typical of summer conditions in that area. This rapid increase in air temperature led to the establishment

¹⁰ of a thermal stratification of the surface waters. Those in situ data illustrate well that there was a shift from spring to summer conditions between P and Q experiments.

During DUNE-2, the seawater temperature during the whole experiment was in the range 20.0–27.3 °C, with a significant increase in surface temperature and well stratified waters toward the ends of both seeding periods. According to seawater temperature

- ¹⁵ data, the period after the R1 seeding experiment was representative of a transition between spring and summer conditions, while the second seeding was performed while the surface layer was well stratified. During the course of the second seeding period, a destratification followed by a restratification was observed with a strong increase in sea surface temperature typical of summer conditions. Photosynthetically available ra-
- diation (PAR) was measured both in the air and at the sub-surface of the mesocosms during DUNE-2 and even if some variation in the percentage of sunshine duration was noticed (Fig. 4d), the light flux was high during the whole experiment (Fig. 4d) with average daily flux at the sub-surface of the mesocosms of 20 and 19 mol photons m⁻² d⁻¹ respectively during R1 and R2 with maximums close to 900 µmol photons m⁻² s⁻¹. For
- P and R experiments, no strong and established stratification over the course of the experiment could be observed. For Q, the sharp thermocline at ~ 5 m depth could act as a physical barrier, with extremely low diffusion. Such features of the surface waters, well captured by the mesocosm are important for the interpretation of biogeochemical data acquired during the experiments.



According to satellite data, the same type of situation regarding chlorophyll (Chl a) concentrations was encountered in 2008 and 2010: the area where the experiments took place (red circles in Fig. 5) are typical of very "blue" waters because of the uplift of the Ligurian Current along the Corsican coast that isolates the coastal area from more productive waters of the center of the Ligurian Sea: during both experiments, the tested waters were typical of oligotrophic conditions with Chl a concentrations in the range $0.07-0.11 \,\mu g L^{-1}$ (Table 3). Because dust inputs to seawater have mainly been interpreted as a "fertilizer" for oligotrophic systems, Chl a is the parameter that has been targeted in most attempts to understand the impact of dust deposition on marine ecosystems, either considering satellite data (see for ex. Dulac et al., 1996, 10 2004; Volpe et al., 2009), or microcosm experiments with dust addition (see for ex. Bonnet et al., 2005). At least a doubling of Chl a was observed for DUNE-P and DUNE-R1 (Fig. 6). The second dust addition during DUNE-2 stimulated an additional Chl a increase (up to +160% compared to CONTROL-MESO). Interestingly, no significant Chl a increase was observed after deposition of non-EC dust (DUNE-Q). Despite the 15 significant changes observed for DUNE-P and DUNE-R, the Chl a concentrations remained very low (maximum values $0.22 \,\mu g L^{-1}$) maintaining the oligotrophic status of the tested waters.

Dissolved inorganic phosphorus and dissolved iron were measured along the 4 ex-²⁰ periments and data for DUNE-1-P have been published earlier (Pulido-Villena et al., 2010; Wagener et al., 2010). Initial conditions for DIP indicate very low and very similar concentrations for all P, Q and R experiments (averages 2 to 5 nM) (Table 3). DFe concentrations were of the same order of magnitude as DIP. Initial total dissolved iron concentration was similar for P and Q (respectively 2.4 ± 0.3 (Wagener et al., 2010) and

²⁵ 2.3 ± 0.3 nM (Wagener, personal communication, 2012) and higher for R (3.3 ± 0.8 nM) (Wuttig et al., 2013). Such nanomolar concentrations for DFe are quite typical of coastal area (Johnson et al., 1997); for example they are of the same order of magnitude as those recently measured at 5 m depth in the Villefranche Bay ($3.7, 3.6, and 3.8 \text{ nmol L}^{-1}$ in May, October, and February, respectively; Bressac and Guieu, 2013). Initial DIN



concentrations were not measured due to analytical issues for P and Q and were found below the detection limit (< 30 nM, Ridame et al., 2013a) for R. According to our analysis during R and to other recent measurements performed in a similar environment in summer at the Stareso station near Calvi in N-W Corsica, using nanomolar technique,

⁵ (MedSea EU project, $NO_3^- = 18 \pm 3 \text{ nM}$, J. Louis, personal communication, 2013), we can assume that the initial NO_3^- concentrations were below 30 nM also for P and Q, in agreement with the concentrations typical of oligotrophic Mediterranean waters measured at that time of the year (see for ex. Pujo-Pay et al., 2011).

These data overall indicate that similar in situ characteristics were encountered at the beginning of each experiment DUNE-1 P, DUNE-1 Q and DUNE-2-R. These characteristics do confirm the oligotrophic character of the experimental site. The perturbations induced by the simulated dust wet deposition (P exp.) or dry deposition (Q exp.) or two successive wet deposition (R1 and R2 exp.) can thus be (1) compared to each other and (2) taken as representative of actual changes that take place in the Mediter-

- ¹⁵ ranean Sea (MERMEX Group, 2011). Moreover, as previously emphasized in Guieu et al. (2010b), such nanomolar concentrations of nutrients imply a specific care at all the different steps (building, deployment, mooring, filling, seeding, sampling etc.) in order to avoid any type of contamination while using mesocosms. It is thus important to make clear that initial conditions (Table 3) outside the mesocosms and inside were not significantly different (*p* > 0.05 for P, Q (Guieu et al., 2010) and R). Our methodology
- indeed allowed working in those delicate conditions providing for the first time a large panel of solid/reliable biogeochemical data including trace metals.

4 Special issue presentation

Our "large clean mesocosm" approach allowed us to follow as a function of time and taking into account the vertical dimension, a number of key parameters involved when a strong atmospheric deposition impact the sea surface. Chemical and biological changes in the mesocosms and in the material exported below the surface layer,



along with modifications of the dynamic of particles following a wet or two successive wet or dry dust events have been followed thanks to the 3 experiments conducted. The multidisciplinary results described in this special issue are bringing new insights regarding the role of atmospheric deposition on oligotrophic biogeochemistry, ecosystem and carbon export.

Because DUNE was a transdisciplinary project between scientists from the atmosphere and the ocean, an important focus of the project was to study the adsorption/desorption/dissolution processes before and after the deposition. In their paper, Losno et al. (2013) report on the solubility of atmospheric nutrients from experiments conducted in laboratory using the fine fraction of soils including the one used for the artificial seeding during DUNE. The solubility of major and trace elements was shown to mainly depend on the chemical composition – and in particular the calcium carbonate content – and on pH.

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The temporal changes in the biogeochemistry of crustal metals in the water column
following dust events have been explored by Wuttig et al. (2013) during DUNE-R. The dissolved trace metals, Mn, Al and Fe, showed different behavior. In the case of Mn and Al, a clear increase in dissolved concentration was observed directly after both seedings R1 and R2 due to dissolution processes. On the contrary, the first seeding R1 resulted in a decrease of dissolved Fe due to scavenging by sinking dust particles, in agreement to what was observed and modeled during DUNE1-P (Wagener et al., 2010; Ye et al., 2011), whereas the second addition of dust R2 induced dissolution of Fe from the dust particles due to the excess Fe binding ligand concentrations present at that time.

A number of particulate tracers were followed in the water column and in the sed-²⁵ iments traps during the three experiments. When comparing the dust deposited with the particles in the mesocosms and those transiting below (sediment trap), Desboeufs et al. (2013) have shown that the dust composition evolves after seeding by dissolution of calcium minerals, although the crustal element as AI or Fe have a stable content all along the experiments, confirming their interest as dust proxy in sediment traps.



A clear difference of dust settling is observed for the experiment DUNE-Q in comparison to the experiments DUNE-P and -R, which seems to be associated to biota response to the seeding, supporting the ballast effect of organic matter proposed by Bressac et al. (2013). Wet deposition during DUNE-1-P yielded a transient increase

- of dissolved inorganic phosphorus (DIP) followed by a very rapid return to initial conditions Pulido-Villena et al. (2010). The same was observed during DUNE-2 after the first dust pulse. The second one also induced an increase in DIP concentration although no return to initial conditions was observed (Pulido-Villena et al., 2013) and EC-dust released significant amount of nitrate (Ridame et al., 2013b). By transiently increasing
- ¹⁰ DIP and DIN concentrations in P-N-starved surface waters of the Mediterranean Sea, wet deposition of Saharan dust can likely relieve the potential P and/or N limitation of biological activity.

Those results have all clearly shown the potential for Saharan wet deposition to modify the in situ concentrations of elements of biogeochemical interest such as P, Fe,

N. An important focus of DUNE was to quantify the impact in terms of biological response. Wet deposition of dust strongly stimulated primary production (PP) and during several days, indicating that dust was able to relieve the ambient nutrient limitation of primary production. Based on estimation of the new production (NP), a switch from a regenerated-production based system (NP ~ 15% PP) to a new-production based
 system (NP ~ 65% PP) is evidenced 24 h after P and R seedings (Ridame et al., 2013a).

This new production was supported by different size class organisms depending on conditions. Indeed, Giovagnetti et al. (2013) show that during DUNE-R1 small phytoplankton (<3 μ m) was better stimulated after the dust addition whereas the bigger

size class (> 3 µm) significantly increased after the second addition. The regulation of photobiological processes was distinctively affected by nutrient availability in both phytoplanktonic size classes. Picophytoplankton is the group responding the fastest to dust additions, in terms of both ecophysiological state of cells and community composition.



On the contrary, bigger-sized cells need further nutrient supply for being able to adjust their physiology and compete for resource acquisition and biomass increase.

Among microorganisms responding to the atmospheric inputs, a specific focus was made on abundance, diversity and N₂-fixing activity of diazotrophs following dry and ⁵ wet deposition. Ridame et al. (2013a) have shown that N₂ fixation, although only responsible for a few percent of the induced new production, is well stimulated by both wet and dry atmospheric deposition. The response of the picoplanktonic unicellular diazotrophic cyanbacteria (UCYN) which dominate the community of diazotrophic cyanbacteria in the Mediterranean Sea (Le Moal et al., 2011) was more contrasted.

- ¹⁰ Dry deposition (Q exp.) led to a strong increase in the UCYN abundance while simulated wet event induced a slight increase in UCYN during DUNE-R or no response over DUNE-P (Biegala, personal communication, 2013). Changes in bacteria community structure have been evidenced during DUNE-1 as Saharan dust was shown to induce changes in the active community of particle-attached bacteria as a higher contribution
- of Alteromonas macleodii to the active bacterial community was found at the end of the experiment (Laghdass et al., 2011). The authors hypothesized that small dust particles, by providing a source of iron, might favor the activity of specific heterotrophic bacteria like *A. macleodii* due to siderophores production. Similarly, during DUNE-2, dust deposition modified bacterial community structure by selectively stimulating and
- inhibiting certain members of the bacterial community. These modifications, however, did not translate into changes in bacterial diversity which remained constant over the duration of the experiment (Pulido-Villena et al., 2013). There was a strong impact of dust deposition on heterotrophic bacteria as shown by Pulido-Villena et al. (2013). Indeed, the first dust pulse in DUNE-2 did stimulate bacterial activity (i.e. respiration)
- processes) more than abundance and/or community structure. This pronounced stimulation of bacterial respiration seems to be bottom-up controlled after the first dust pulse R1. Bacterial activity, likely top-down controlled by viral lysis, was not further stimulated by the second dust pulse. The observed rapid C remineralization due to microbial food web processes may question the nature of the link between dust deposition and



carbon cycling which may not directly involve an increase in C sequestration by the ocean. Indeed, the tested waters during DUNE-1 and DUNE-2 were representative of the oligotrophic Mediterranean Sea i.e. characterized by a strong net heterotrophy (Guieu et al., 2013a) even after the dust deposition, indicating that bacteria are key players of ecosystem response to a dust deposition. This indicates that the so-called 5 fertilization effect induced by dust deposition is not in favor of a CO₂ sink: because of

- very high mineralization of organic matter, particulate carbon export should be lowered, resulting in a negative impact on carbon export. On the other hand, the importance of aggregation processes between organic material and dust was evidenced by a series of
- optical measurements performed inside one seeded mesocosm during DUNE-2 (Bres-10 sac et al., 2012). A clear link has been established between the lithogenic fluxes and the POC fluxes by Bressac et al. (2013, this issue) as (1) the lithogenic fluxes explained more than 80% of the variance in the particulate organic carbon (POC) fluxes and (2) 36-50 % of the POC flux was strictly associated with lithogenic particles through ag-
- gregation. These observations support the "ballast" hypothesis and suggest that this 15 "lithogenic carbon pump" could represent a major contribution of the global carbon export in areas receiving high rates of atmospheric deposition. Finally, the carbon budget indicates that the net effect between carbon fixation, respiration and the lithogenic ballast effect is a significant particulate organic carbon export outside of the surface layer

(Guieu et al., 2013a). 20

> We used a stoichiometric microbial food web model to investigate how the stoichiometry of consumer driven nutrient recycling (Sterner, 1990) may influence the ecosystem response to N and P dust addition. This model considers simultaneously the differential recycling of C, N and P in a food chain which includes 7 compartments (bacteria, au-

totrophic pico and nanoplankton, heterotrophic flagellates, ciliates and mesozooplank-25 ton) (Pondaven et al., 2013). Growth rates of bacteria and phytoplankton were controlled by the availability of dissolved inorganic and/or organic N. P and C. The model results showed that bacteria and zooplankton had substantial effects on the supply rate and the stoichiometry of N and P in the dissolved pools, thereby exerting a control



on the predicted primary production. These effects were more pronounced when the difference between the C:N:P ratio of producers and that of their consumers ("*sto-ichiometric mismatch*") increased. Although the model was kept simple, it highlights how *stoichiometric mismatch* between producers and consumers can influence the response of a planktonic food web to nutrient addition.

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Table 1. Composition of some key elements in the dust used for seeding during DUNE.

	non-processed representative of dry deposition used for DUNE-1-Q exp. ^a	evapocondensed representative of wet deposition used for DUNE-1-P exp. (1)	evapocondensed representative of wet deposition used for DUNE-2-R1 and DUNE-2-R2 exp. ^b
P (%)	0.044 ± 0.009	0.045 ± 0.015	0.055 ± 0.003
Fe (%)	2.28 ± 0.19	2.31 ± 0.04	2.26 ± 0.03
N (%)	0.11 ± 0.01	1.19 ± 0.05	1.36 ± 0.09
C %	6.75 ± 0.01	5.35 ± 0.06	5.08 ± 0.02

^a Guieu et al. (2010). ^b Desboeufs et al. (2013).

Table 2. Sampling sketch for all the DUNE dust seeding experiments. Sampling type OUT: all the mesocosms and a position at mid-distance between the 2 mesocosm clusters are sampled at the same detphs. Sampling type IN: all the mesocosms are sampled. Time indicated are the starting times of sampling; the average total sampling time was less than 2 h. Net sampling was performed at the beginning of experiments outside the mesocosms and both inside and outside after the last sampling of the experiment.

P experiment

Dates	sample label	time	type of work
10 Jun	p1	4 p.m.	sampling type OUT + install traps
11 Jun	_	9 a.m.	seeding D1 D2 D3
11 Jun	p2	4 p.m.	sampling type IN and net ^a
12 Jun	p3	9 a.m.	sampling type IN and traps
13 Jun	p4	9a.m.	sampling type OUT
14 Jun	-	10 a.m.	traps; no sampling of the mesocosms: bad weather conditions
15 Jun	p5	9a.m.	sampling type IN
16 Jun	p6	9a.m.	sampling type OUT and traps
17 Jun	р7	9a.m.	sampling type IN
18 Jun	p8	9a.m.	sampling type OUT, traps and nets ^b

^a only outside the mesocosms;

^b zooplankton sampling: "outside", C2, C3, D2, D3

Q experiment

Dates	sample label	time*	type of work
20 Jun	q1	9a.m.	sampling type OUT + install traps
20 Jun		12 a.m.	seeding D1 D2 D3
20 Jun	q2	6 p.m.	sampling type IN and net ^a
21 Jun	q3	9 a.m.	sampling type IN and traps
22 Jun	q4	9a.m.	sampling type OUT
23 Jun	q5	9a.m.	sampling type IN and traps
24 Jun	q6	9a.m.	sampling type IN
25 Jun	q7	9a.m.	sampling type OUT and traps
26 Jun	q8	9a.m.	sampling type IN
27 Jun	q9	9 a.m.	sampling type OUT, traps and nets ^b

^a only outside the mesocosms

^b zooplankton sampling: "outside", C1, C2, D1, D2



Table 2. Continued.

R experiment			
Dates	sample label	time*	type of work
26 Jun	r1	9a.m.	sampling type OUT and net ^a
26 Jun	-	11 a.m.	install traps + seeding D1 D2 D3 Optic
26 Jun	r2	9 p.m.	sampling type IN
27 Jun	r3	9a.m.	sampling type IN and traps
28 Jun	r4	9a.m.	sampling type OUT and traps
29 Jun	r5	9a.m.	sampling type IN and traps
30 Jun	r6	9a.m.	sampling type OUT and traps
1 Jul	r7	9a.m.	sampling type IN and traps
2 Jul	r8	9a.m.	sampling type OUT and traps
3 Jul	r9	9a.m.	traps
3 Jul	-	9a.m.	seeding D1 D2 D3 Optic
3 Jul	r9	2 p.m.	sampling type IN
3 Jul	r10	6 p.m.	sampling type IN
4 Jul	r11	9a.m.	sampling type OUT and traps
5 Jul	r12	9a.m.	sampling type IN and traps
6 Jul	r13	9a.m.	sampling type OUT and traps
7 Jul	r14	9a.m.	sampling type IN and traps
8 Jul	r15	9a.m.	sampling type IN and traps
9 Jul	r16	9a.m.	sampling type OUT, traps and nets ^b

^a only outside the mesocosms

^b zooplankton sampling: "outside", C1, C2, C3, D1, D2, D3



Table 3. Initial conditions of the tested waters during DUNE-P and DUNE-R.

	DUNE-1-P	DUNE-1-Q	DUNE-R
Chl a (μ g L ⁻¹)	0.11 ± 0.03	0.08 ± 0.02	0.07 ± 0.02
NO ₃ nM	na – – – ^b	na	< 01 ²
DIP, nM	$5\pm2^{\circ}$	$2\pm0^{\circ}$	$5\pm3^{\circ}$
DFe, nM	2.4 ± 0.3^{e}	2.3 ± 0.3^{9}	$3.3 \pm 0.8'$

^a Ridame et al. (2013).

^b Pulido-Villena et al. (2010). ^c Pulido-Villena, personal communication (2013).

^d Pulido-Villena et al. (2013).

^e Wagener et al. (2010).

^f from Wuttig et al. (2013).

^g Wagener et al., personal communication (2012).





Fig. 1. Stocks (green) and fluxes (blue) that were measured during the DUNE experiments. Stocks and fluxes can be measured in mesocosm after the simulation of a realistic atmospheric input in a water body large enough to be representative of natural processes. As the particles are naturally sinking, those changes are closer to the "real" processes occurring in the surface mixed layer of open ocean, compared to microcosm approaches where the particles are homogenized and not allowed to sink (modified from de Leeuw et al., 2013).





Fig. 2. The different steps of DUNE: (top panel, from left to right) sampling soils in a region of South Tunisia where the Saharan aerosol is produced and is typical of inputs to the north-western Mediterranean; soil collected on the three stages of the dry sieving column at the end of the fine particle production process; artificial aging of the atmospheric particles to mimic the wet deposition, in a clean room to reproduce the processes taking place during aerosols transport (bottom panel, from left to right), a view above the surface of a group of 3 mesocosms during the daily sampling, a group of 3 mesocosms from below (photos F. Dulac, CEA; K. Desboeufs, LISA; Luquet, OOV).





Fig. 3. Implementation of the two mesocosms experiments in the Scandola preservation area (Corsica).

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Fig. 4. Physical characteristics of the surface water, air temperature and light during the experiments. **(a)** Correlation between the temperature continuously monitored inside and outside the mesocosms; for DUNE-1, the dots represent the data acquired by temperature sensors at sub-surface, 5 m and 10 m; for DUNE-2, the dots represent the data acquired by temperature sensors at sub-surface, 3 m, 6 m and 10 m. **(b)** Temperature profiles inside the mesocosms during the 3 DUNE experiments. Air temperatures are superimposed (black line). **(c)** Evolution of the temperature difference between surface and 10 m illustrating both the diurnal variations and the evolution of the overall stratification. **(d)** Percentage of sunshine duration over the whole daytime hours (~ 15 h in June–July) (Météo-France); PAR measurements for R1 and R2 (*y* axis), the integration for the whole day acquisition are presented in molphotons m⁻² d⁻¹ at the subsurface of the mesocosms, the maximum flux values are reported.





Fig. 5. MODIS satellite images showing the distribution of the surface Chlorophyll *a* at the time of DUNE-1 (left) and DUNE-2 (right).





Fig. 6. Comparison of the changes in chlorophyll *a* (relative difference between average DUST and average CONTROL mesocosms, mean of concentrations at 0-5 and 10 m) observed in P, Q and R experiments as a function of time after seeding.

