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Interactive comment on "Quantification of the lithogenic carbon pump following a dust deposition event" *by* M. Bressac et al.

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Please find below our reply after the review of our paper initially entitled "Quantification of the lithogenic carbon pump following a dust deposition event". We would like to thank the first reviewer for their relevant comments on the manuscript. We have taken the suggestions of the reviewers into account and some sections have been re-written based on these comments.

General Comments: The manuscript by Bressac et al. describes results from a mesocosms experiment, investigating the effect of dust addition on the sedimentation rate of POC. Previous studies suggest that minerals like lithogenic particles act as ballast for the downward transport of organic matter in the ocean. However, as the authors also emphasize an accurate understanding and quantification of the POC-dust asso-

ciation in the upper ocean is currently lacking. This study indicates that POC fluxes are strongly increased after dust addition yielding a linear relationship between dust flux and POC. The authors attempt to explain the observed enhancement of POC sedimentation by two main processes: a) the direct ballasting of POC that increase settling speed of particles, and b) a fertilization effect that results in higher biomass production und thus in higher export fluxes of dust amended mesocosms.

Specific comments: An enhancement of POC fluxes after dust addition meets well with our expectations, based on previous studies that showed strong co-occurrence of POC and minerals in deep water sediment traps, or higher settling velocities of mineral-organic aggregates. In this manuscript, however, there are several issues that need to be addressed and better explained, to convince that the presented results are novel and give deeper insight. Even if the manuscript is part of a Biogeoscience special issue with more information likely given in accompanying papers, enough information should be given also in this paper to support interpretation of data.

1) The method of dust addition and the dust size frequency distribution could have large impact on the interaction of dust with POC and need to be described in more detail. The authors wrote that addition of 10g dust m-2 (i.e. 41.5g in total) mimics realistic wet deposition; but over what time? How fast was the dust added and spread? Is the amount still realistic when deposited all at once?

RESPONSE: A new paragraph (2.2. The dust analogue and the seeding) has been included in the methodology section, as follows: "The finest dust fraction (<20 μm) was separated from the bulk soil samples, dominated by quartz (40%), calcite (30%) and clays (25%), by grinding and dry-sieving. Then the dust analogs were processed to simulate cloud evapocondensation cycles. The physicochemical characteristics of the dust analog are reported in Desboeufs et al. (2013). The resulting dust population presented a volume median diameter around 6.5 μm and a peak at $\sim$10 μm, while the particle number size distribution peaked at 0.1 μm (Guieu et al., 2010a). The DUNE-2 experiment lasted 14 days, from 26 June to 09 July 2010. Two artificial seedings were successively conducted seven days apart within the same mesocosm and consisted of mimicking realistic wet deposition events with a dust flux of 10 g m-2. Such a flux corresponds to 41.5 g of evapocondensed dust diluted in 2 L of ultrapure water and sprayed onto the surface of each of the mesocosms for a total duration of ∼40 minutes. In the Mediterranean basin, dust is 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). Between 1984 and 1994, Loÿe-Pilot and Martin (1996) reported a mean annual flux in Corsica of 12.5 g m-2 yr-1, mainly attributed to pulses > 1 g m-2. According to the same authors, this deposition is mainly wet deposition and may occur only with few drops of rain meaning that high amount of dust can be deposited in time scales of minutes. Similar strong and sudden (few hours) events have been recorded over the past decade with African dust deposition fluxes as high as 22 g m−2 (Bonnet and Guieu, 2006; Guieu et al., 2010b; Ternon et al., 2010). Our simulation, which allows to seed on a quasi-synoptic way all the +dust mesocosms, is thus realistic in term of flux and duration (Guieu et al., 2013a)".

2) Since the authors speculate about a fertilization effect of dust, it is indispensable to show nutrient data, or better explain how the dust addition increased biomass production.

RESPONSE: This comment has been taken into account. The evolution of nutrient concentrations (DIN, DIP, and DFe) following the seedings is now detailed in the section 3.1 (Evolution of the physical and biogeochemical parameters). The evolution of nutrient concentrations is used to explain the increase in primary production and chlorophyll-a concentrations observed in the +dust mesocosms: "Initial conditions indicate: (i) very low dissolved inorganic phosphorus concentrations ([DIP]) (5±3 nM; Pulido-Villena et al., 2013), (ii) dissolved inorganic nitrogen concentrations ([DIN]) below the detection limit (<30 nM; Ridame et al., 2013b), and (iii) dissolved iron concentrations ([DFe]) typical of coastal area (3.3±0.8 nM; Wuttig et al., 2013). Following the first seeding, a decrease in [DFe] due to scavenging by sinking dust (Wuttig et al.,

2013) and a transient increase in [DIP] (Pulido-Villena et al., 2013) were observed. The second addition of dust induced a significant increase in [DFe] (Wuttig et al., 2013) and [DIP] (Pulido-Villena et al., 2013). Furthermore, significant increases in [DIN] were observed following both seedings (Ridame et al., 2013b). By increasing [DIP] and [DIN], the seedings relieved the ambient nutrient limitation and strongly stimulated primary production (see section 4.1). Based on estimation of the new production, Ridame et al. (2013b) observed a switch from a regenerated-production based system to a new-production based system 24 h after the seeding".

3) Likewise, it is essential to include POC concentration from the water column. As the authors also note, PP cannot accurately predict POC fluxes. Thus, knowing POC concentration of the water column would give a much better insight into the partitioning of PP into particles and sedimentation processes.

RESPONSE: We agree that POC concentration from the water column is a relevant parameter for our study. However, primary production values presented in this paper are directly derived from POC measurements performed at 5 m depth (13C uptake method; see details in Ridame et al., 2013). Therefore, we think that POC concentrations cannot be used to better predict POC fluxes. However, the evolution of Δcp(670) (figure 3) demonstrates the ballasting of the suspended matter, and the sediment trap data provides a global view of the particulate export (organic and lithogenic fractions) throughout the experiment. Therefore, this data set enables us to address our initial question (i.e. the lithogenic ballasting following dust deposition pulses), but does not allowed to further investigate the partitioning of PP into particles and sedimentation processes.

4) PP data were integrated over the water column (0-12.5m) assuming a homogenous profile. Given the determined changes in light attenuation after dust seeding, the assumption of a homogenous profile seems to be wrong for light. What was the light intensity during the experiment? Could photo-inhibition explain lower PP in the non-seeded mesocosms?

RESPONSE: This is a relevant comment as the phytoplankton community undergoes quite extreme regimes of high light in the surface waters during summer. As highlighted by the reviewer, the light attenuation profile was not homogeneous in the +dust mesocosms, especially during the first 24 h following both seedings (figure 3 of the manuscript). Photosynthetically available radiation (PAR) was measured both in the air and at the sub-surface of the mesocosms during the DUNE-2 experiment. The light flux was high during the whole experiment with average daily flux at the sub-surface of the mesocosms of 20 mol photons m^{-2} d^{-1} (Guieu et al., 2013). Giovagnetti et al. (2013) discussed the effect of the dust additions on the physiological state of the phytoplankton community. The authors showed that increasing nutrient concentrations caused a rapid enhancement of the pigment content per cell, reflecting nutrient-dependent processes within photosystems. At the photophysiological level, Giovagnetti and colleagues (2013) observed a rapid photoprotective response (i.e. NPQ development) soon after the dust additions. The photoprotective response remained quite high during few days, before decreasing during nutrient concentration lowering. By considering the extreme regime of high light in the surface water, photo-inhibition could explain the low PP observed in the non-seeded mesocosms. However, the increase in PP within the seeded mesocosms has been likely favored by physiological acclimation induced by the input of new nutrients, rather than light attenuation induced by the high dust concentration within the water column. Therefore, we could assume that difference in PP is directly related to the input of new nutrients. This aspect is discussed in the section 4.1, as follows: "The strong stimulation of PP went along with a rapid enhancement of the pigment content per cell and a rapid photoprotective response (Giovagnetti et al., 2013). Therefore, the increase in PP seems to have been favored by such physiological acclimation in relation to the high light environment (Guieu et al., 2013a)".

5) The authors should add an error estimate for POC fluxes that are given as average.

RESPONSE: The standard deviation of the replicate mesocosms has been added (figure 1 of the manuscript).

6) Was PP comparable between mesocosms before dust addition? The data shown in Fig. 5 suggest a large variability in PP of replicate dust mesocosms. Please indicate replicate mesocosms.

RESPONSE: Primary production data and the figure presented below will be submitted in a companion paper to Biogeosciences Discussion in December (Ridame et al., in prep.). Although variability in PP of replicate dust mesocosms is large (especially after the first seeding), PP values measured in the +dust mesocosms are significantly different from values measured in the control mesocosms (see figure 1 of the author comment). The increase in PP following both seedings, and the difference between both data sets (control and +dust values) are now detailed in the text, as follows: "Since optical data was limited to the first 48 hours of experiments, sediment trap data and primary production (PP) were used to determine the biological and lithogenic contributions in POC export over experiments. Similar increases in PP (by a factor of 2.3-2.4) were observed within +dust mesocosms following both seedings, while PP remained constant within control mesocosms (Ridame et al., 2013a). Both data sets remained significantly different (p-values < 0.05) throughout the experiment".

Caption of the figure 1 (from Ridame et al., in prep.) – Mean integrated primary production (PP) in mg C m-2 d-1 over mesocosms during the DUNE-2 experiment in Control-meso (black dot), Dust-meso (orange dot) and Out (grey dot). The dotted line represents the time of the dust seeding. Data in the Control- and Dust-meso represent the average and standard deviation of the three replicate mesocosms. Means in Dust-meso that were significantly different from Control-meso ($p<0.05$) were labeled with the * symbol.

7) POC flux in dust-amended mesocosms was quite comparable after both seedings, while POC control flux was different. Moreover, OM carrying capacity of dust was lower than determined for minerals during previous studies. This could indicate that DOM sorption onto dust, rather than dust-particle interaction was responsible for observed POC fluxes. As the trap material was collected for analyses during this study, there

must be more information about the quality of sinking particles. Were there visible aggregates or fecal pellets? Which were the dominant phytoplankton species/size in the traps? What was the C:N ratio of sinking particles?

RESPONSE: Binocular observation revealed that aggregates were present in both control and +dust samples. However, samples from the +dust mesocosms were composed of a denser and highly aggregated matter (see figure 2 of the author comment), in agreement with the formation of organic-mineral aggregates (Bressac et al., 2012). Because of their too small size, it was not possible to determine the phytoplankton species and their abundance through the use of the binocular microscope. Furthermore, phytoplankton cells were included in this sticky and highly aggregated matter. The abundance of fecal pellets and swimmers was not significantly different between control and +dust samples (N. Leblond, personal communication). The C:N ratio of sinking particles was well correlated with the lithogenic fluxes. The relationship between lithogenic fluxes and C:N ratio was linear and highly correlated for both seeding experiments (R2 = 0.70 and 0.78 for the first and second seeding experiments, respectively). We decided to not present this data set, as it does not provide additional information on the evolution of the quality of the exported material.

8) Sorption of DOM onto minerals is a well-known phenomenon and should be discussed in more detail here, referring to the work of Arnarson and Keil. Measurements of DOC, if available, should be included in this discussion. Perhaps, if the same type of dust is still available, side experiments with filtered seawater can still be conducted to determine the amount of carbon adsorbed onto the dust surface.

RESPONSE: This kind of abiotic experiment has already been performed with the same dust analog and flux in 0.2 μm filtered seawater and the results have been published (Bressac and Guieu, 2013). In this study, the amount of dissolved organic carbon adsorbed onto the dust surface was estimated by measuring the organic carbon content of the material collected in the sediment trap of the “minicosm” (a 300 L tank). Sorption of DOM onto dust particles is discussed in more detail in the re-

vised manuscript (but also with more care, as DOC concentrations were not measured (please see the last comment of the reviewer #2)) (1) by comparing results from the abiotic experiment with the POC fluxes measured during the DUNE-2 experiment, and (2) by referring to the work of Arnarson and Keil. “Sorption of dissolved organic matter (DOM) from solution onto mineral surfaces could reach equilibrium very rapidly (within $\sim$1 hour; Arnarson and Keil, 2000, 2005). This process likely contributed to the downward POC export in the first 24 h following the seedings. By using the same dust analog and flux in 0.2 μm filtered seawater, Bressac and Guieu (2013) quantified this abiotically driven organic carbon export and its variation as a function of the composition and abundance of DOM. During this abiotic experiment, the amount of organic carbon adsorbed onto dust particles and exported downward was of the same order of magnitude as the POC fluxes observed in the +dust mesocosms. Furthermore, sticky particles such as transparent exopolymer particles (TEP), known for promoting the aggregation process (Chin et al., 1998; Passow, 2002; Engel et al., 2004; Verdugo et al., 2004), could also have indirectly influenced the particulate export. Although DOM and TEP concentrations were not measured during this experiment, the abundance of such organic material prior to the second seeding was likely higher within the +dust than within the control mesocosms, contributing to the larger difference in POC fluxes observed following the second seeding. Although lithogenic ballasting cannot be strictly considered as a pump (as surface primary production is required to sustain it), Bressac and Guieu (2013) introduced the concept of the “lithogenic carbon pump” in order to highlight the fact that aggregation and adsorption processes could trigger a dust-induced POC export event. As the fertilization effect (e.g. Marañón et al., 2010), POC export abiotically triggered by dust deposition could vary according to the biogeochemical state of sea surface waters at the time of deposition (Bressac and Guieu, 2013)”.

Technical comments: The manuscript should be carefully revised and spell-checked by a native speaker.

RESPONSE: A revision of the English language has been performed by a native speaker prior to the initial submission.

References cited: Bressac, M., Guieu, C., Doxaran, D., Bourrin, F., Obolensky, G., and Grisoni J.-M.: A mesocosm experiment coupled with optical measurements to assess the fate and sinking of atmospheric particles in clear oligotrophic waters, Geo-Mar. Lett., 32, 153-164, doi:10.1007/s00367-011-0269-4, 2012. Bressac, M., and Guieu, C.: Post-depositional processes: What really happens to new atmospheric iron in the ocean's surface? Global Biogeochem. Cycles, 27, 859-870, doi:10.1002/gbc.20076, 2013. Giovagnetti, V., Brunet, C., Conversano, F., Tramontano, F., Obernosterer, I., Ridame, C., and Guieu, C.: Assessing the role of dust deposition on phytoplankton ecophysiology and succession in a low-nutrient low-chlorophyll ecosystem: a mesocosm experiment in the Mediterranean Sea, Biogeosciences, 10, 2973-2991, doi:10.5194/bg-10-2973-2013, 2013. Guieu, C., Dulac, F., Ridame, C., and Pondaven, P.: Introduction to the project DUNE, a DUst experiment in a low Nutrient, low chlorophyll Ecosystem, Biogeosciences Discuss., 10, 12491-12527, doi:10.5194/bgd-10-12491-2013, 2013. Ridame, C., Dekaezemacker, J., Guieu, C., Bonnet, S., and L'Helguen, S.: Phytoplanktonic responses to contrasted Saharan dust events: results from dust seeding in large mesocosms, Biogeosciences, in preparation, 2013.

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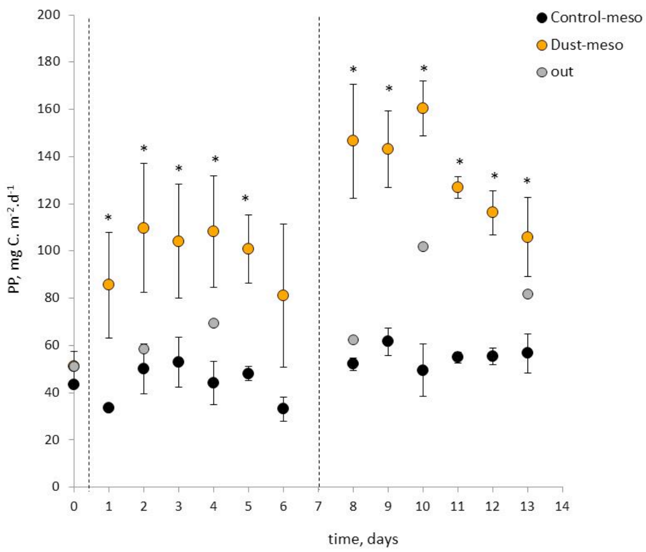

Fig. 1. From Ridame et al. (in prep.) – Mean integrated primary production in Control-meso (black), dust-meso (orange) and Out (grey).

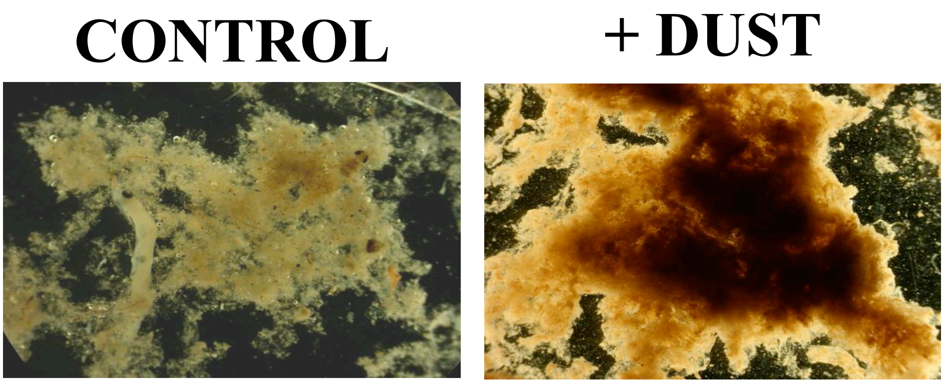

Fig. 2. Binocular observation of the matter collected in the sediment traps of the control and +dust mesocosms.