

Answer to anonymous Referee #1

We thank the anonymous reviewer for her/his comments and suggestions on our manuscript. We agree with most comments and modified/updated the manuscript accordingly. Below is a point-by-point reply, [our answers appear in italics].

This is a creative and timely study on the solubility of dust-derived elements in the ocean. The authors find a range of behavior among different elements which will certainly help oceanographers interpret biogeochemical cycling of these elements. This is a high quality and careful study that should be published in Biogeosciences. I have a few substantive comments for the authors to address before final publication and several technical comments.

Referee question:

Substantive comments: Line 35, page 3. More needs to be said about how it was determined what is a “realistic” dust input. The 10 g/m² is realistic over what timescale? If considered over a 3 day time period, this would correspond to a flux of ~1200 g/m²/yr, which I believe is on the high end of model-based estimates for the Mediterranean. If the flux is considered only over an initial time period after dust deposition the effective flux would be even higher. Some of this information is brought up in the discussion, but it should be discussed in the introduction so we understand the experimental design.

***Reply:** It is now indicated in the introduction and sections 2.1. & 4. that dust enrichment occurred at the beginning of the experiment and that it corresponds to the simulation of a strong Saharan dust deposition event in the Mediterranean Sea, as illustrated by Ternon et al., (2010).*

*Ternon, E., Guieu, C., Loje-Pilot, M. D., Leblond, N., Bosc, E., Gasser, B., Miquel, J. C., Martin, J.: The impact of Saharan dust on the particulate export in the water column of the North Western Mediterranean Sea, *Biogeosci.*, 7, 809–826, <https://doi.org/10.5194/bg-7-809-2010>, 2010.*

Referee #1: Could you speculate on how the results might be different in a scenario where a smaller amount of dust was added at regular intervals over a longer time period?

***Reply:** Done. We now indicate in section 4.2 that very high dust content and adsorption rates were reached because all the dust was deposited instantaneously at the beginning of the experiment. Deposition of the same amount of dust over longer periods (weeks, months) as it occurs in less dusty environment than the Mediterranean Sea, would certainly result in less readsorption (but likely similar percentages of dissolution).*

Referee #1: It might also be worth mentioning that dust particles fall into low pH, high CO₂ and cold water in the thermocline which could affect element solubility.

***Reply:** It is complicated to extrapolate these surface water results to the thermocline, because it is unclear if the change in dissolution fraction is a direct effect of temperature or CO₂ change or an indirect effect through the biological activity. Therefore it is not possible to extrapolate to the thermocline where T, CO₂ and biological activity will not covariate like during the present experiment.*

Referee #1: L2, page 9. Fe ligands are almost always found in excess of observed Fe concentrations (e.g, Buck et al., 2015; <https://doi.org/10.1016/j.dsr2.2014.11.016>). This suggests Fe is not at a solubility limit if the availability of ligand is the effective solubility limit for seawater. Have Fe ligands been determined in the tanks or in the region?

Reply: *We agree with this comment of the reviewer. The effective solubility of iron is controlled by the availability of ligand in seawater and the role of Fe ligands in maintaining high concentration of DFe in seawater has been emphasized in many studies. Iron binding ligands have not been determined in the mesocosm. Only a limited amount of measurements have been done during the deployment of the RESPIRE traps during the cruise (Bressac et al 2020, Whitby et al 2020) indicating ligand excess in the subsurface. However, the limited existing studies on iron binding ligand concentration in the Med. Sea confirms the ligand excess in the surface Med. Sea. (e.g. Gerringa et al. 2017, Wuttig et al. 2013, Wagener et al 2008, Van den Berg, 1995) We mention the paper by Wagener et al. 2008 that demonstrates seasonal variation of organic speciation for iron and reports excess ligand concentration values for July of the same order of magnitude compared to a previous study by Van den Berg, 1995 (total iron binding ligands were in the range for both studies: 4.2-5.85 nM-eqFe).*

Bressac, M., Guieu, C., Ellwood, M. J., Tagliabue, A., Wagener, T., Laurenceau-Cornec, E. C., et al. (2019). Resupply of mesopelagic dissolved iron controlled by particulate iron composition. *Nature Geoscience*, 12(12), 995–1000. <https://doi.org/10.1038/s41561-019-0476-6>

Gerringa, L. J. A., Slagter, H. A., Bown, J., van Haren, H., Laan, P., de Baar, H. J. W., & Rijkenberg, M. J. A. (2017). Dissolved Fe and Fe-binding organic ligands in the Mediterranean Sea—GEOTRACES G04. *Marine Chemistry*, 194, 100–113. <https://doi.org/10.1016/j.marchem.2017.05.012>

van den Berg, C. M. G. (1995). Evidence for the organic complexation of iron in seawater. *Marine Chemistry*, 50(1–4), 139–157.

Whitby, H.; Bressac, M.; Sarthou, G.; Ellwood, M. J.; Guieu, C.; Boyd, P. W. Contribution of Electroactive Humic Substances to the Iron-Binding Ligands Released During Microbial Remineralization of Sinking Particles. *Geophysical Research Letters* 2020, 47 (7), e2019GL086685. <https://doi.org/10.1029/2019GL086685>.

Wagener, T., Pulido-Villena, E., & Guieu, C. (2008). Dust iron dissolution in seawater: Results from a one-year time-series in the Mediterranean Sea. *Geophysical Research Letters*, 35(16).

Wuttig, K., Wagener, T., Bressac, M., Dammshäuser, A., Streu, P., Guieu, C., & Croot, P. (2013). Impacts of dust deposition on dissolved trace metal concentrations (Mn, Al and Fe) during a mesocosm experiment. *Biogeosciences*, 4, 2583–2600.

Referee #1: Page 9. Is it possible to assess an uncertainty in the derived solubility fractions?

Reply: *The propagated uncertainties on the solubility fractions are now provided in table 1.*

Referee #1: Sections 4.4 and 5. There is an interesting juxtaposition of ideas here that should be clarified. First, there is a suggestion that previous high Th solubility fractions could be the result of lateral advective inputs. However, the observations of scavenging within the tank experiments as well as the conclusion that ²³²Th cannot accumulate along the Mediterranean deep circulation, would suggest relatively little lateral transport of ²³²Th. There may be certain

distance scales the authors have in mind to draw the distinction between these two cases, and if possible, they should be stated more explicitly.

Reply: *Scavenging within the tank is enhanced by the very high particle concentration. Although the dust quantity deposited in the tanks was realistic for the Mediterranean Sea (observed in situ, see for ex Ternon et al., 2010), it is not relevant for less “dusty” areas of the ocean and the high dust concentration observed in the amended tanks cannot be extended to all ocean surface waters. Therefore, the short Th residence time implied by the fast Th scavenging in the amended tanks is relevant for the Mediterranean Sea after strong dust deposition, but certainly not for most open ocean areas where the Th residence time is longer (as measured by ^{234}Th in surface waters for example or, as mentioned by Frank Pavia in his review “The ^{230}Th -based removal residence times in the upper 250m of the South Pacific Gyre (depths over which Th-based dust flux estimates are integrated) are 1-2 years (Pavia et al., 2020).”). Hence the good scale necessary to avoid lateral transport, as developed in the answer to the review of Frank Pavia, is, at best, the gyre scale. We agree that ^{232}Th does not accumulate along the deep Med sea circulation, but we stress that in the Med sea, ^{232}Th input and scavenging potentially occur all along this path through exchanges with the nearby margins.*

Detailed comments:

Referee #1: Line 14, page 3. Technically ^{230}Th is produced from the decay of ^{234}U .

Reply: *This is changed*

Referee #1: Line 18, page 3. Pa is defined in the first paragraph so the full name and extra open parenthesis can be removed here.

Reply: *This is done*

Referee #1: Lines 18-19, page 3. The meaning of the parenthetical is not clear “(absolute or at least relative) “. I think this could simply be deleted.

Reply: *This is deleted*

Referee #1: Line 28, page 3. Tells us which season is characterized by strong stratification (summer?)

Reply: *It is now indicated that the cruise occurred during the late spring.*

Referee #1: L15, page 4. The concentration given is unclear. Is the final concentration of formaldehyde a weight percentage, volume percentage or mole percentage?

Reply: *It is now indicated in the text that volume percentage are considered*

Referee #1: L16, page 5. Missing superscript 232

Reply: *This is corrected*

Referee #1: L23, page 6. Extra E, HREEE

Reply: This is corrected

Referee #1: L33, page 8. The equation involving “ $x m/V$ ” needs to be explained in words. It is not clear what is being calculated here.

Reply: It is now explained that m represents the mass of dust added to the tank and V represents the volume of seawater in the tank.

Referee #1: L10, page 9. DIP acronym not defined.

Reply: Dissolved Inorganic Phosphorus is now defined

Referee #1: Referee #1: L10, page 10. The number/range given is not clear “ $10-100 \mu\text{mol/mol}$ ”. Is this a range of ratios or a range of numerator and constant denominator?

Reply: Clarified by indicating that the Fe/C ratio ranges from $10 \mu\text{mol/mol}$ to $100 \mu\text{mol/mol}$