Dear Dr Zhang,

Below are the point-by-point replies to the reviewer's comments and corrections. The reviewer's comments are in plain text, the author's answers in blue and italic. We provide with this reply a revised version of the manuscript.

Thank you,

Karine Desboeufs

Refere 1: Rachel Shelley

The revised version of the manuscript, 'Wet deposition in the remote western and central Mediterranean as a source of trace metals to surface seawater' by Desboeufs et al. is much improved. By removing the sections where the SML was discussed, the manuscript has become more focused. The updates made to the figures also greatly improve this manuscript. I am happy that the changes made adequately address my earlier comments and appreciate the author's detailed response to those comments. I have a few minor suggestions detailed below.

All editorial and technical corrections requested by Dr Shelley have been made in the revised manuscript.

Referee2:

This discussion related to mass balance of N, P, Al and Fe on the water column and contributions of atmospheric inputs relative to other inputs was not presented in the paper. I suggest a paragraph summarizing the mass balance analyses and the uncertainties associated with these calculations.

The mass balance of N, P, Al and Fe on the water column are the topic of three other publications of the SI as mentioned in the manuscript. These papers are summarized in this manuscript: p668-684: "The impact of the dust wet deposition on nutrients stocks in the Mediterranean surface waters is discussed in details in van Wanbeke et al. (2020) and Pulido-Villena et al. (2021). To briefly summarise, both nitrate and DIP increased in the ML following the rain. Although the closure of the N and P budgets had to necessarily take into account post-deposition processes such as new nutrient transfer through the microbial food web (uptake, remineralisation, and adsorption/desorption processes on sinking particles), it was shown that wet deposition was a significant source of nutrients for the ML during the cruise. For example, atmospheric supply of phosphate could contribute to 90% of new production at FAST (Pulido-Villena et al., 2021). Bressac et al. (2021) studied the response to Al and Fe cycles to dust deposition during the cruise. They showed that total Fe and Al stocks were increased by dust wet deposition, and that the dissolved Fe atmospheric inputs were transient in the ML and were accumulated in the subsurface waters (100-1000m). The low depth-resolution of marine TM concentration samplings prevent the possibility to make TM inventories on the water column. We focus here on the role of dust wet deposition events as a source of TMs to the surface mixed-layer, except Mo due its marine origin in the rain samples."

This kind of calculations is subject different uncertainties inherent in the assumptions that are used to estimate the various fluxes (atmospheric inputs, advection, diapycnal fluxes ..) and biological uptake of these elements in the mass budgets, it will be too long to discuss the choice made for these assumptions in this paper which the topic is not the mass balance of these elements. However, nutrients and Fe

being the main elements governing marine phytoplankton growth, the purpose of this part of the manuscript is to inform that the discussion on these topics is available in other papers.

While the authors stated that the purpose of this study was not to conduct longer term monitoring, the results of the two rain samples is quite limiting and needs to be stated. Sample size is an important indicator of the representativeness of the findings and reliability of the statistical summaries. Data comparability is another issue. Given that most wet deposition monitoring studies were conducted over a longer time period (e.g. seasonal or annual scales), is it possible to compare the fluxes obtained from this study with other studies?

We agree that the comparability of fluxes at the scale of event with long-term measurements is an issue. This is why we compared and discussed our data with concentrations in rain samples collected in network measurements (L491-505), rather than flux values (daily or yearly period). Indeed, the fluxes obtained in this study were only wet deposition fluxes on a period of 3 weeks in 2 different locations, we do not think it is relevant to compare them with long-term measurements. As suggested by the reviewer, since the number of samples was limited, there would be no consistency in extrapolating the values of these two rain events to the scale of the year for comparison with long-term fluxes or even to use long-term daily fluxes when it is known that wet atmospheric deposition is very sporadic in the Mediterranean climate.

The study also measured metals and metalloids that are toxic to biota; therefore, there should be some discussions on the potential negative effects of toxic metals on aquatic ecosystems. If there are no negative effects on aquatic organisms, that should be stated. If there is uncertainty regarding the negative effects or requires further study, that should be stated as well.

We do not really understand this suggestion and what it is expected as "statement" by the referee. We mentioned in the introduction of the manuscript the previous literature showing that the TM effect could be positive or negative (L75-85). However, we cannot discuss these effects since we did not follow the biological response after rain events during the cruise. We measured atmospheric inputs of metals to the surface sea water. To our knowledge, it is not possible to assess simply by their marine concentrations whether metals have an effect on phytoplankton. It would have been necessary to do seeding experiments in culture for this (e.g. Mackey et al., 2012) or else to make estimates by considering atmospheric inputs in a biogeochemical model (e.g. Richon et al., 2018 for P). The data that we provide are dedicated to be used by this type of models (at least to constrain them) but at the stage of this study this estimate cannot be made.

The authors mentioned, "...Moreover, even if annual wet and dry deposition are equivalent in Mediterranean (Theodosi et al., 2010), wet deposition is known to provide soluble, and potentially bioavailable forms of TMs (Jickells et al., 2016)." Dry deposition also provides soluble forms of TMs; see Muezzinoglu and Cizmecioglu (2006) for an example. According to that study, the mean soluble fraction of TMs in dry deposition were similar to those in wet deposition. Doesn't this suggest that dry deposition can also provide soluble forms of TMs? The revised sentence does not seem to justify the importance of wet over dry deposition of TMs. I suggest a brief discussion of literature comparing dry and wet deposition fluxes in the Mediterranean region. Dry and wet deposition of TMs is likely equally important in regions with high dust loadings.

Muezzinoglu, A., and Cizmecioglu, S. C. (2006). Deposition of heavy metals in a Mediterranean climate area. Atmospheric Research, 81(1), 1-16.

We agree that dry deposition is also a source of soluble metals, as shown by numerous studies including the work of Muezzinoglu and Cizmecioglu (2006). However, in this paper, measurements have been done in urban area, where the continental aerosol sources are continuous and hence dry deposition

fluxes are higher than in open Sea. Moreover, contrary to wet deposition which provides immediately dissolved metals for marine surface after deposition, the dissolution of metals after dry deposition could be on several days, e.g. as mentioned in L729: "Mackey et al. (2015) show that in case of dry deposition, aerosol Co and Fe dissolution in seawater can be gradual and continue up to 7 days after contacting seawater.". To clarify this difference between these two kinds of inputs and justify the interest to focus our work on wet deposition, we have changed "soluble" by "dissolved" in the introduction L 99: "Moreover, even if annual wet and dry deposition are equivalent in Mediterranean (Theodosi et al., 2010), wet deposition is known to provide ***dissolved****, and potentially bioavailable forms of TMs (Jickells et al., 2016).".*

Concerning the requested brief discussion to compare dry and wet deposition in Med., we don't think that this comparison would not support or enrich the rationale and the conclusion of our manuscript. We think that our work on wet deposition fluxes is of interest by itself, even without comparison to dry deposition. The purpose of this paper is not to estimate the atmospheric fluxes of TMs but to characterize the TM composition of rain water in open sea and to study the impact of wet deposition on TM marine pool. Even if the annual dry deposition is higher, the wet deposition is limited in the time and intense, so the impact on marine TM pool will be different. It is not antagonistic, the study of wet and dry deposition are complementary. In this idea, we suggested the need to do new measurements for dry deposition in the abstract (L51) and in the conclusion (L821-826).