Referee2:

We thank the reviewer for his/her review of our manuscript. Below are our detailed responses to questions and comments. The reviewer's comments are in plain text, the author's answers in italic, quotes from the manuscript are in quotation marks.

This is an interesting study that examined the wet deposition fluxes of trace metals with co-located measurements in surface seawater and marine stocks in the Mediterranean Sea prior to and after the rain events. The study showed that wet deposition contributed to trace metals in surface seawater and marine stocks. However, there are some scientific questions that have not been addressed in the paper. The role of dry deposition of particles is undermined considering that dry deposition of trace metals is often equivalent to or greater than that of wet deposition based on a literature review of worldwide measurements (Cheng et al. 2021). The duration of the wet deposition monitoring is too short and limited to two rain events. The paper reported one event representing regional wet deposition and another event representing wet deposition from a dust episode. One question is whether these single event wet deposition fluxes can be extrapolated to seasonal or annual fluxes, which is typically what is measured in other wet deposition monitoring studies. Indeed, wet deposition does contribute to trace metals in surface seawater and marine stocks, but it is highly uncertain to what degree when compared with post deposition processes, effluent discharge to the sea, shipping pollution, etc. To better understand the relative importance of these processes, a mass balance analysis on the water chemistry is something to consider.

We agree that dry deposition can be an important input for marine environment. However, the purpose of this paper is to study the role of wet deposition and strategy has been elaborated to limit the role of dry deposition since our conclusions are based on a comparison between wet deposition fluxes and marine stocks in short period before and after rain. Indeed, the Mediterranean climate with short periods of intense rains favours the role of wet deposition. It is the reason why the paper is here focused on wet deposition. We have added a specific comment in the conclusion about dry deposition which should be also studied in open Med Sea: "As the wet deposition fluxes decreased since the 90's due to mitigation, it is highly probable that the dry deposition fluxes were also changed. Further measurements on dry deposition in open Med Sea are needed in order to estimate its contribution in TM atmospheric inputs."

We are aware that the conclusions of this study are subject to limitations, due to the fact that only two rains were studied and due to the role of post-depositional processes. We have mentioned these limitations (e.g. p27, L 691: " However, we cannot exclude that the pCu and pNi inputs were masked by the uncertainties of stock calculations.") and taken precautions about our conclusions by using "suggest" or "could.." and by mentioning always "our results". For example p29, L734: " Finally, our results show that the studied atmospheric dust event was a net source of particulate TMs and dissolved Fe and Co for ML at FAST. Even if the wet deposition delivered TMs already as soluble forms (Fig. 8), our results showed that the wet deposition constitutes only a source of some of dissolved TMs for surface waters. Due to various marine post-deposition processes, it is more complicated to observe the effect of wet deposition on dissolved stocks. The post-deposition dissolution of particulate rain inputs could represent an additional pathway of dissolved TMs supply for the surface ocean, notably for low soluble TMs in wet deposition. Thus, the dissolved atmospheric inputs could be underestimate from the only measurements of atmospheric fluxes."

We agree that post-deposition processes could be critical for mari,e TMs stocks, besides these processes are discussed in the revised manuscript (section 4.2.2.). However, our results showed that the stocks were clearly increased after rain and that even if it is a lateral transfer, these input were linked to

surrounding precipitations. It is also the reason why we estimated the atmospheric fluxes on a radius of 25 km around the R/V position. Moreover, about effluent discharge and shipping pollution, the measurements were carried in open sea with specific material and procedures to limit all contamination by other sources than atmospheric deposition. It is not intended to extrapolate our findings to coastal areas where river inputs or coastal pollution need to be considered and where atmospheric inputs are known to be often negligible.

The mass balance of N, P, Al and Fe on the water column was made for estimating the contribution of atmospheric inputs relative to other inputs (advection, diapycnal fluxes ..) in these elements budgets, in other publications issued from PEACETIME cruise, as mentioned from p2, L652 to 671 (van Wanbeke et al. (2020) and Pulido-Villena et al. (2021), Bressac et al., 2021). This kind of calculations was subject too much uncertainties in the case of TMs due to both the depth-resolution and time-resolution sampling of ML and water column during the cruise.

Regarding the extrapolation of our data to the seasonal or annual scale, we have refrained from talking about it since we determined two rain samples during the stratified period of spring, p31, L792: " The marine TM concentrations measured during the cruise being typical of Mediterranean surface seawater concentrations, we can conclude that wet deposition events were an external supply of dissolved Fe, Co and Zn for the Med Sea, and more generally for all TMs in case of intense wet dust deposition, during the period of thermal stratification." Indeed, even if we extrapolate to the scale of the measurement area, we cannot go any further in our conclusions on the role on the annual or even seasonal scale, for that we would need a longer term monitoring and also have the dry deposition values. But again, this was not the purpose of this paper.

Another important question is that based on the trace metals deposited to the Mediterranean Sea, would this result in negative effects on aquatic organisms.

Our measurements show that there has been a decrease in TM concentrations in rain since the 1990s. Only dissolved Co and Fe have their marine stock impacted by wet deposition event. As these metals are known for their positive physiological role on marine organisms, the probability that the atmospheric inputs induce a negative impact on marine biosphere is probably limited. However, this would require monitoring of rainfall fluxes in relation to phytoplankton growth. This is not the topic of this paper either.

Specific comments

Line 59: The sentences emphasized wet deposition, but the importance of dry deposition of metal-containing aerosols was not discussed.

The deposition from L62 to 69 is presented as total deposition, then L77 to 86. Even if fluxes of wet deposition could be in the same order of magnitude of dry deposition, see inferior, the rain inputs are delivered already in a soluble (and potentially bioaccessable) form. Moreover, due to Mediterranean Climate, the wet deposition were sporadic but often intense. In order to mention the contribution of dry deposition, we have added L86:" Due to Mediterranean sporadic and intense storms, the rain events by scavenging loaded air masses with anthropogenic aerosols or Saharan dust could lead to higher deposition TM fluxes than dry deposition (Desboeufs et al., 2021). 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)."

Line 110: Rainwater was collected during the period between 11 May and 10 June 2017. How many samples were collected?

R: One sample by rain, the detail of sampling is provided in section 2.

Line 290: The subheading can be more detailed. E.g. Atmospheric conditions prior to rain events.

R: We have changed the title by "Atmospheric conditions during wet deposition events"

Line 303: "in the night between June 28 and 29," Why are these dates different from those in Table 1?

R: The table 1 give the perio of sampling, whereas "in the night between 28 and 29 June" described the period of rain in the vicinity of the R/V position.

Section 3.1: The low sample size seems to be an issue. There was only one rain event representative of regional background conditions and one rain event representing wet deposition from a dust episode.

R: We are aware that this study was based on two rain samples. Rain sampling during cruise is very dependent on meteorological conditions and positions of R/V. Even if the strategy during the cruise was to chase wet deposition event, only two rain were sampled. This study was not dedicated to obtain a large database of rain composition, but was focused to estimate recent rain deposition composition and fluxes in open sea, since no data was published since 80's.

Lines 378-379: It is unclear if this is the dissolved or total concentrations of Fe and Zn in rain.

R: We have clarified (L413): "Regarding TMs in rain, Fe and Zn presented the highest concentrations both in the dissolved fraction and in total deposition with the same order of magnitude (10 to 25 µg L-1)."

Section 3.2: There should more discussion on how the chemical composition between the two rain events differ, e.g. are there different sources contributing to the scavenging of metals for the ION and FAST events? Are the sources of TMs in dust natural or anthropogenic? The dissolved concentrations for metals are not very different between the ION and FAST rain events. Are they statistically different? It appears that the total concentrations were much higher for the FAST event than the ION event. Any possible explanations as to why the dissolved concentrations are much more comparable between the two events than total concentrations?

R: This discussion is provided in the sections 4.1.2 and 4.1.3 as a function of EF and solubility values. Here, in the results section, only concentration values are given.

Lines 474-475: It was mentioned that the rainwater chemistry from this study cannot be compared to those from previous studies because emissions levels were much higher back then. Is it valid to compare the enrichment factors from this study with those from earlier studies? Why is the enrichment factor for Zn in this study higher than that of previous studies despite regulations on toxic trace metal emissions?

R: EF values enable to determine the origin of TMs between desert, anthropogenic or marine sources. Even if the emission have been reduced during the last decade, the EF values determine an enrichment relative to the upper crust, so this enrichment is dependent on aerosol composition. All the

anthropogenic sources present this enrichment whatever the intensity of their emission, it is only related to the composition of the emission.

Line 535: What causes the lower solubility of TMs in high dust events?

We agree that the reason of the lowest solubility for TMs associated with dust origin is not clearly explained. So we have added a sentence in L579: "However, it is known that metals that are mainly associated with crustal aluminosilicate mineral lattices such as Fe and Ti have very low solubility values, due to the difficulties to breaking bonds in the lattices (Jickell et al., 2016)."

Line 556: Is this the deposition flux over the course of the rain event? Can you quantify the time period associated with this deposition flux, e.g. mg/m2/day? Given the rain events last up to a few days, can the deposition fluxes be extrapolated to seasonal or annual fluxes?

Fluxes are calculated on the period of rain event. As mention before, we don't think that the extrapolation of our data to the seasonal or annual period is relevant since it is only two rain samples. Besides no discussion is carried in this sense.

Lines 591-592: How many intense deposition events occur in a typical year?

Dust event are sporadic, so it is not a regular phenomenon. It is the reason why the dust flux of our event is compared in the published data, L 617 to 624, in order to situate the sampled wet dust deposition event relative to the most intense event: "Although low compared to deposition fluxes reported in the western Mediterranean (Bergametti et al., 1989; Loÿe-Pilot and Martin, 1996; Ternon et al., 2010), our flux estimates are in the same order of magnitude of the most intense weekly dust deposition fluxes calculated more recently in Corsica between 2011 and 2013 (14% of fluxes >50 mg m-2) and is comparable to the mean weekly flux (93 mg m-2) reported for Majorca during the same period (Vincent et al., 2016)."

Section 4.2.2: Have you compared the trace metals profile in wet deposition and in seawater and in marine stocks? Are they comparable? Based on the trace metals deposited to the Mediterranean Sea, would this result in negative effects on aquatic ecosystems? Although the authors qualitatively discussed the role of post deposition processes on seawater concentrations, some data are needed to elucidate the importance of atmospheric deposition relative to the post deposition processes.

R: The atmospheric fluxes and marine stocks were calculated since these parameters are comparable at the difference of concentrations which is dependent on dilution effect. As mentioned before, it is impossible to estimate the negative effect of atmospheric deposition in this study and it is not the aim. No measurements of post-deposition processes have been quantify during the cruise, for example biological uptake, making impossible to known the importance of atmospheric deposition compared to these processes.

Lines 745-747: “we suggest to use the chemical composition of PEACETIME rains as a new reference for the studies TMs on wet deposition in Med Sea”. I suggest rewording this statement. This is only one wet deposition monitoring study in the Mediterranean region, and the paper presented the findings from two rain events. Other studies have been conducted to capture background and dust episodes in this region. The results on the trace metals composition and their solubility in wet deposition are not particularly new compared with previous studies.

R: As mentioned in the text to our knowledge, no data on rain composition in open Med Sea was published since the 80's.In consequence, our results are the only recent measurements available for this region.

Lines 750-753: HNLC has not been defined.

R: done