Refere 1: Rachel Shelley

We thank Rachel Shelley very much for her valuable review of our manuscript. Below are our detailed responses to her 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. All English mistakes have been corrected in the revised manuscript from manuscript with tracks changes provided by the reviewer (these corrections are not detailed in this reply). We provide with this reply a revised version of the manuscript and a track version allowing to visualizing changes from the submitted manuscript.

General comments

The manuscript, 'Wet deposition in the remote western and central Mediterranean as a source of trace metals to surface seawater', by Desboeufs et al. presents data on trace metals (TM) in rain and surface water samples. The current title is generic and could be more descriptive such as 'A comparison of two contrasting wet deposition events in the remote western and central Mediterranean and their impact of trace metal behaviour in surface waters.'

The authors present rainwater TM (12 elements), Al, P and N species concentrations and EFs, and discuss these parameters in terms of their fluxes and impacts on concentrations of dissolved and particulate fractions in the upper 20 m of the water column, including the SML. The relatively large number of parameters under discussion results in rather a long paper. Nevertheless, this dataset is the first of its kind for the remote Mediterranean Sea and highlights differences between TM concentrations in rainwater from studies (mainly) in the 1990s conducted at coastal locations. Therefore, this study provides valuable new data from a relatively remote region that is influenced by anthropogenic emissions from Europe, episodically interspersed by pulses of Saharan dust, both of which are delivered to the surface ocean via atmospheric deposition (wet and dry). Wet deposition is thought to have a disproportionately high influence on TMs in surface waters as they are thought to be delivered already in a soluble (and potentially bioaccessable) form. This study illustrates that only some of the TMs studied were delivered predominantly in soluble form; an important point that the authors could emphasise more. Furthermore, rain samples are notoriously challenging to collect at sea. Having paired water column and rain samples is a valuable addition to the scientific literature. I recommend that this manuscript is accepted pending major revisions, given that a restructuring is suggested.

Specific comments

There is no mention of dry deposition – it is important to note that dry deposition is also an important source of TMs.

R: We agree with the reviewer. In general dry deposition could be a source at least equal and even higher to wet deposition. However, 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. This information was not mentioned, so we have added a sentence in this purpose in the introduction, p2 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)."

Moreover, we have added a specific comment in the conclusion: "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."

Finally, at the end of the abstract, we mentioned the need to obtain data on dry deposition: "Our results suggest that the wet deposition constitutes only a source of some of dissolved TMs for Med surface waters. The contribution of dry deposition on the atmospheric TM inputs need to be investigated."

Specific values from this study are needed in the Results section text. I have made suggestions in the text as to where these would be of value.

R: We have added values where requested, e.g. section 3.3., p16, L422: " The TMs were mainly in dissolved forms in the ML (Kd from 0.006 to 0.5), except for Fe, whose dissolved and particulate concentrations were in the same order of magnitude (Kd around 2)." Or L432: " However, dCo concentrations (from 10 to 20 pM) were among the lowest ones measured during stratification period in Western Med Sea (~120 pM, Dulaquais et al., 2017)."

I suggest restructuring this manuscript to combine the Results and Discussion sections. This will make this manuscript easier to follow and stop the unnecessary repetition. It would also help to reduce the length of the manuscript.

R: We haven't changed the structuring of the paper. We thought that the structuring in two parts seems to us to be the most adapted to distinguish the primary data that could be used as they are from the interpretation that we have made of them. The different changes have reduced the length of paper and removed major repetitions.

One of the sticking points for me is the mismatch between SML sampling, ML sampling and rain sampling. I fully appreciate the challenges associated with sampling the SML. Rain, wind speed and sea state all impact the integrity of the SML making it extremely challenging to collect a SML sample immediately prior to and after a wet deposition event. It has been demonstrated that the SML reforms rapidly when wind speed drops below a threshold and that TMs have a short residence time in the SML (minutes to hours, compared to a few days in a 20 m mixed layer). I am concerned that the delay in sampling caused by the meteorological conditions would have missed the TM dynamics in the SML in response to the wet deposition, likely skewing the results towards a higher ratio of dissolved to particulate TMs being observed. The authors note that dissolution of TMs continues after deposition. I think the data is of value but I would like to see some more discussion of the limitations and challenges of sampling the SML at the resolution that would be needed to demonstrate the 'true' impact on TM concentrations following wet deposition. The authors do go some way towards discussing the limitations of the SML sampling towards the end of the manuscript, but I feel it needs to be addressed sooner and in more detail.

R: As noted by the reviewer, the interpretation of the SML data was greatly complicated by the fact that the samples could not be taken simultaneously with the ML sampling and 24h after the Rain FAST, and also by the potential effect of waves, rain... on the results obtained for this layer. From these observations, a large part of conclusions were speculative. Moreover, the analysis of SML was not key in this purpose of this manuscript since the aim of this manuscript was to estimate the role of atmospheric inputs as source of TMs for surface waters. The analysis of SML data made the discussion more cumbersome and masked the main highlights. In consequence, we have decided to remove all the data (data being available in the database and publishing in partly in Tovar-Sanchez et al., 2020) and

discussion on the SML. However, we considered the role of SML in the interpretation and discussion of ML stocks, by using the conclusions of Tovar-Sanchez et al., 2020 (see all the revised section 4.2.1).

I also feel that the significance of these findings is a little overstated as only two rain events were sampled. However, combined with findings from previous studies, it is apparent that the data supports the assertion that wet deposition impacts the mixed layer inventory of TMs. As I mentioned in the General Comments, one of the most interesting findings for me was the relative proportions of dissolved to particulate fractions and how this changed in response to the wet deposition, even with the limitations associated with the SML data.

As mentioned after in specific replies, the particulate to dissolved ratio were added in Figures 5 and 8, and these values were used for supporting discussion both on the atmospheric fluxes and on the comparison with marine stocks.

Line 34. Define DOC: *done*

Line 44. ML not yet defined: *done*

Line 49. Give examples/define continental aerosol in brackets
R: We don't think that we need to define here "continental aerosol", this term is used in opposition to marine aerosol to evoke the influence of continental particles emission even in the remote sea (in agreement with the cited reference). The continental aerosol in the case of Med Sea is described after in the text.

Line 52. Biosphere is too general. I suggest replacing it with microbial as it's this community that are directly accessing the dissolved nutrients in seawater: *done*

Line 52. State which nutrients: *done*

Line 79. I suggest preceding the sentence that starts in this line with 'To the best of our knowledge, ...' as there are studies that have determined concentrations of TMs in rainwater and seawater simultaneously. There are several other studies that have combined rain and seawater concentration data in recent years (e.g. Buck et al., 2010; Shelley et al., 2017) and others that have incorporated wet and dry deposition, combined with seawater concentrations and SML concentrations, although they may not have published their findings yet. What differs with this study is that a suite of elements were determined from the same location, rather than when the ship was underway, in a region of highly variable atmospheric dust inputs where no such data has previously been published. Therefore, I also suggest changing 'reported' to 'published' (line 80) to avoid instances of reporting of unpublished data at conferences or in theses.
R: We agree so we have started the sentence as suggested and we have specified in the text what made our strategy original, p2 L89: " Yet, to the best of our knowledge, the direct impact of wet deposition on TM concentrations in surface seawater has not been studied in a same location by concurrently collecting both rainwater and seawater samples before this work, whether in the Med Sea or in other oceanic regions.

Line 95. Possibly more important in terms of why coastal atmospheric TM concentrations might not be representative of open ocean samples is gravitational settling during atmospheric transport. Add a statement to this effect.
R: As the majority of literature contains data reported from coastal rain sampling, this part of text is to show that these rain samples may not be representative, not only in term of concentration but also of

composition of offshore deposition events. We agree that intensity and variety of emission sources is not the only factor to explain this difference, however the gravitational settling is a process among others (dispersion, chemical processes, rain-out..) to explain why the TM composition issued from continental aerosol are not representative in open sea. So we have changes the sentence by adding the processes during transport, p3 L104: "However due to the continental and local sources of pollution and the variety of anthropogenic aerosol sources (Amato et al., 2016) and the physico-chemical processes during atmospheric aerosol transport (chemical ageing, dispersion, gravitionnal settling, in-cloud reactivity..)(Weinzierl et al., 2017), the TM rain composition of the coastal zone may not be representative of atmospheric deposition to the remote Mediterranean"

Line 109. Delete 'Research Vessel' and remove brackets from around R/V: *done*

Line 116. Delete (PC) as you don't use this abbreviation anywhere else in the text: *done*

Line 112. Change to 'The rain collector': *done*

Line 126. Reorder this list and restructure this section. The focus of this manuscript is TMs so they should come first. I suggest i) TMs, ii) DOC, iii) major seawater and atmospheric ions, iv) pH. After the list describe the methods for each parameter in the order they are listed. Currently, the list and the methods are mixed together.

R: done: "The dissolved fraction was separated into four aliquots dedicated to i) TM determination by inductively coupled plasma coupled methods (ICP), ii) major ion by ion chromatography (IC), iii) dissolved organic carbon (DOC) determination by high-temperature catalytic oxidation (HTCO) on a Shimadzu total organic carbon analyzer (as described in van Wambeke et al., 2021a), and iv) pH measurement."+ description of sample treatments in this order.

Line 144 and 147. Not all analytes are discussed in the manuscript. Please indicate which ones are discussed.
R: A sentence has been added in p5, L179: "Only TMs, major nutrients, i.e. N and P forms, and Al are discussed in this manuscript."

Line 154. More detail needed. I am not clear what the rain blanks were. What were the blanks for the dissolved fraction? Empty bottles swished out with MQ or acidified MQ - one unfiltered aliquot for total wet deposition and a filtered aliquot for the dissolved fraction?
R: The description of blanks has been completed in p5 L186: "Blank samples were prepared by rinsing the funnel with 150 mL of ultrapure water (18.2 mΩ cm) with the same protocol of rain collection."

Line161. Delete: 'For the sample concentration computations, we subtracted these blanks values to elemental concentrations obtained in rain samples.' Replace with 'Blank concentrations were subtracted from all sample concentrations.' State where the analytical figures of merit can be found.: *done*

Line 172. '..., detection limit for all analytes was 0.5 ppb, ...': *done*

Line 174. Inconsistent use of date format. Sometimes words - day month, sometimes - month day, sometimes dd/mm. Also include the year in the chosen date format.

R: Done, the date format has been homogenised.

Line 198. By rinsing the glass plate with 0.5 L of ultra-pure water – presumably this water was collected as the blank solution? If so, state this.

R: As mentioned before, this part was for SML sampling so it has been removed in the revised manuscript.

Line 196. Delete MQ water and replace with 18 mΩ cm-1.: *Done (unit for resistivity is mΩ.cm)*

Line 206. Word order – change to: TM samples were also collected in the water column using a titanium trace metal clean (TMC) rosette (mounted with 24 teflon-coated Go-Flo bottles) before and after the rain events (Bressac et al., 2021). Although rosette deployments were performed over the whole water column, we focus here on the 0-20 m mixed-layer (ML): *Done*

Line 217. Replace digested with UV-treated.: *Done*

Line 222. Which metals? Mo and Pb or not all samples were preconcentrated for all metals listed? Reword to make this clearer.

R: Now metals are listed (p7, L224)

Line 236. Not exactly true but the vast majority of Al is crustal. Therefore, it is currently accepted as one of the best proxy elements. It is predominantly crustal, not only.:
R: "Only" has been changed by "predominantly"

Line 239. Reword to: considered significantly enriched, which points to a non-crustal source (Rahn, 1976). :
R: Done and completed with "For most metals, enrichment shows important input from anthropogenic sources, due to their low content in other non-crustal sources such seaspray or biogenic aerosols (Jickells et al., 2016)".

Line 244. Change to wet deposition fluxes.: *Done*

Line 254 and many other instances. Sometimes you use ship, sometimes R/V, perhaps best to stick with one for consistency.: *We have homogenised in using "R/V"*

Line 256. Include equation and explanation for using VWM rainfall.
R: The sentence was modified and completed with units (L295): "the wet deposition fluxes in our rain samples were calculated by multiplying the TM concentrations (µg L-1 or µmol L-1) by the total precipitation (mm).". We though that we don't need to add the equation (F= CxP), the sentence being precise enough.

Line 261. best tool to estimate rainfall in the surroundings of the R/V because....
R: the sentence has been completed by "since this method is a direct measurement of precipitation with both a best time and spatial resolution in comparison of model estimation."

Line 264. Replace more or less with approximately:
R: Approximately is not adapted here. "More or less" was used to give the extent of area where rain rates were integrated, so "more or less" has been changed by "±" (L306)

Line 267. Integrated rain rates, not integral: *done*

Line 271. Replace layer with SML: *done*

Line 276. ML, ML depth or MLD – multiple examples throughout the text. Check for consistent use.: *done*

Line 286. Is there a reference for this approach?
R: We could not provide a reference. Partition coefficient is a usual concept in chemistry to describe the partition of a compound between two phases (or two solvents). Here, this concept is adapted for TM distribution between particulate and dissolved phases.

Line 291 on – Section 3.1. No need to use the word hereafter.: *OK*

Line 302. Replace surrounding with vicinity: *done*

Line 307. Replace rate with volume. Rate requires a time dimension. Word order improvement suggested in edited version of manuscript.:
R: "rate" has been replaced by "precipitation" and suggestions of word order has been done (L348)

Fig. 1. Format the date consistently: *done (now Fig. 2)*

I would like to see Fig. S3 here for comparison rather than it be buried in the Supplementary Material.: *The Figure S3 has been added in Figure 2 to compare rainfall of the two rain events.*

Line 337. Reword for clarity. Suggested: The dust plume was concentrated between 3 and 4 km at the beginning of the station occupation, then expanded down to the marine boundary layer (about 500 m amsl) by the end of the day on 3 June 2017. The mass integrated concentration of dust aerosols derived...: *done*

Fig. 2. Can you devise another labelling scheme? You have panels a-c and labelling within panel a of arrows a-c, and in panel c of box a-c. All these a-cs get a bit confusing.
R: Now Fig. 3: We have completd letters (a, b, c) for panel labelling with panel position (top, middle and bottom panels) and changed letters (A,B,C) for selected periods to numbers (1,2 and 3).

Line 355. Change to time then date to make it easier to follow.: *done*

Line 367. Repetition. Delete estimates.: *done*

Line 368. Delete around: *done*

Line 388. What exactly do you mean by stocks? Sometimes it seems to mean concentrations, others partitioning. I think it would be better to replace stocks with partioning, if that is what is meant.
R: The stocks calculation is described in the section 2.7, stocks are not concentrations or partitioning but the integrations of marine concentrations in the ML. Indeed, in the section 3.3, only concentrations were discussed, so the title has been changed, it is only "Marine Concentrations" (Line 431).

Line 389. Are you really talking about deep water masses or just water below the ML?
R: We have clarified: " water below the ML and deeper (e.g. for Fe: Bressac et al., 2021)" (L432)

Section 3.3. Values are needed. Please add where indicated in edited version of manuscript.
R: Values have been added where indicated in commented manuscript between Lines 435 and 451.

Line 397. Add concentration of dMo in seawater (~107 nM from the reference you cite).
+
Line 400. Did any of these studies determine pMo and/or dMo? If not, say that your ones are the first
R: done L440: "..the highest TM concentrations in the surface seawater were found for Mo in the dissolved fraction (~120 nM), these values are the first measurements published in Med Sea and are in agreement with the high abundance of dissolved Mo in seawater in other oceanic regions (~107 nM, Smedley and Kinniburgh, 2017)."

Line 403. Add values of dZn and pZn.: *done*

Line 404. How high? Is contamination suspected?
R: As explaining in Line 448, the observed high concentrations remain in the range of reported Zn concentrations in the literature. However, we cannot be sure that the outlier values didn't reflect a contamination, so we have added a sentence to evoke this possibility: "even if we cannot exclude a possible contamination for these outlier concentrations."

Line 405. The concentrations in the SML were lower than in the ML and Pb dominated both in dissolved and particulate phases. I don't understand, what did Pb dominate?
R: As mentioned before all data of SML has been removed in the revised manuscript

Fig. 4. Box plots are two words not one. Could the panels be plotted on the same scale to make comparison easier?
R: Now Fig. 5: Scales have been homogenised and we have added the Kd values in this figure to easily observe if particulate or dissolved forms predominate in the ML for each TM. This adding enable to support all the discussion on the evolution of TM fractions after rain in section 4.2.2

Line 418. Med to Mediterranean: *done*

Line 424. Reword. The FAST rain concentrations were within the published range, whereas the ION rain was in the low range, confirming a background signature at this station. : *done*

Line 426. Define DIN/DIP:
R: DIN and DIP are now defined in section 2.1. " The speciation of dissolved P was estimated by determining dissolved inorganic phosphorus (DIP) from phosphate concentrations expressed as P and the dissolved organic phosphorus (DOP) from the difference between total dissolved phosphorus (TDP), obtained by ICP-MS, and DIP, obtained by IC. The dissolved inorganic nitrogen (DIN) were defined as sum of NO2, NO3- and NH4+, expressed as N."

Line 428. Bulk deposition – do you mean wet + dry? Clarify. : *done*

Line 430. (in the case of DIN/TDP), but averaged about 100 in bulk deposition – state if this was in unfiltered rain. You have switched from talking about DIN/DIP to presenting data for DIN/TRP. Note this.
R: TDP (total dissolved phosphorus) is the dissolved fraction of phosphorus which could be measured technically only after filtration. The switch (DIP in TDP) was just for the extreme value of ratio, i.e. 1200 not for all the sentence, but we are aware it was not clear. To clarify this, we have changed the sentence

with: " with average ratio about 100, the highest reaching 170 for DIN/DIP and 1200 for DIN/TDP" (L469)

Line 433. Include range : "*no high concentration" means "under limit of detection" so we have clarified*

Line 434. Replace emphasised with observed: *done*

Line 436. Add 'the' before Mediterranean. Wrong tense – change have been to were and include value : *done*
+
Delete Islands: *done*

Fig. 5 caption. Suggested reword: Comparison of dissolved (D) and total (T) TMs concentrations along with data from 14 former studies carried out in to previous studies in the eastern and western Mediterranean Sea.: *done in agreement with reword in the RC2.pdf (now Fig. 6)*

Line 452. Replace notably with especially: *done*

Line 456. Are you comparing like with like here? You state that concentrations likely decrease offshore and, although emissions of TMs may have decreased, if the literature data is from coastal sites you don't have direct evidence for this decline over the open ocean. However, you could infer it from reductions in leaded fuel and coal combustion. Is the decreasing atmospheric input evident in the full depth water column samples? Perhaps this is what the Fu et al data shows but the reader can't check this as the reference is 'in prep'.
+
Later you mention the Dulac thesis as evidence. This paragraph needs rewriting and making more concise in order to strengthen your final, important statement. As it stands, this section is too speculative.
+
Line 456. Switch order to Pb and Cd as the impact of a reduction in atmospheric Pb inputs is well documented. You could cite the GEOTRACES IDP as evidence here.
+
Line 464. Reword: '...related to the decrease in anthropogenic emissions. Thus, our results show the data from before [year] should not be used as a current reference for coastal rain composition due to recent environmental mitigation of TM emissions. '
+
Line 465. Your rain was open ocean not coastal so doesn't really show that historical coastal TM concentrations are higher. The comparison is with the thesis. You could say that coastal values are not representative of open ocean values in general.

R: All the comments from Line 456-465 were about the paragraph on the comparison with TM concentrations in previous studies. We agree that the paragraph was confusing by discussing conjointly coastal/offshore concentrations and previous/recent dataset. We have rewritten the paragraph in ordering arguments:
"Most of the referenced works on coastal rainwaters date from the late 1990s. There is a continuous decline of TM emissions since 90's due to regulatory efforts (Pacyna et al., 2007). The subsequent decrease of the anthropogenic Cd and Pb imprint on atmospheric inputs from European coasts to open sea is well documented (OSPAR, 2008; Travnikov et al., 2012; Geotraces IDP2021). Since the phasing-out of leaded automobile gasoline, the decrease of atmospheric Pb concentrations is also observed in Mediterranean atmosphere (Migon et al., 2008). The low TM concentrations of ION and FAST rain

samples, in particular Cd and Pb, suggest a pronounced decrease of TM inputs in open Mediterranean due to environmental mitigation on TM emissions. Moreover, the coastal deposition is generally not representative of open sea inputs, e.g. due to proximity of anthropogenic sources in coastal areas. Thus, the 90's data should not be used as a current reference for open Mediterranean rain composition."

Added references:
Travnikov,O., Ilyin I., O.Rozovskaya, M.Varygina, W.Aas, H.T.Uggerud, K. Mareckova, and R. Wankmueller, Long-term Changes of Heavy Metal Transboundary Pollution of the Environment (1990-2010), EMEP Status Report 2/2012
OSPAR Commission, 2008. Atmospheric deposition of selected heavy metals and persistent organic pollutants to the OSPAR maritime area (1990 – 2005). Publication 375/2008.
GEOTRACES Intermediate Data Product Group, The GEOTRACES Intermediate Data Product 2021 (IDP2021). NERC EDS British Oceanographic Data Centre NOC. doi:10.5285/cf2d9ba9-d51d-3b7c-e053-8486abc0f5fd, 2021.

Section 4.1.2. This section jumps around a bit, Rain ION then Rain FAST, the Rain ION again, then both, etc. It would be easier to read if one station was discussed and the other one contrasted to it
+
Line 474. Reword and clarification needed - For both rain samples, the EF of Zn was on average five times higher than the EF found in rain samples from coastal sites (?) in the Mediterranean region (Özsoy and Örnektekin, 2009; Al-Momani et al., 1998; Losno, 1989). However, extremely high enrichments of Zn in rainwater have been reported from island sites in the Mediterranean Sea, for example, Frau et al. (1996) reported geometric mean EFs of ~ 6500 in both crust-rich and crust-poor rains from two sites in southern Sardinia, and Fu et al. (2017) reported EF > 1000 for Zn in atmospheric bulk deposition on Lampedusa Island. The Zn EF at station ION is the same order of magnitude as at these island sites which suggests... – something about the evidence for an anthropogenic background signal in the open Med.
+
Line 486. Potential local influences - what do you mean here? To resolve the contribution of various local industries and shipping or local inputs from long range transport?
+
Line 487. As the atmospheric gas and particle measurements do not indicate an anthropogenic influence, you cannot say that the concentration and EF values confirm an anthropogenic influence – your data tells a conflicting story.

R: Regarding the different comments on the section 4.2.1, this section has been rewritten considering Rain ION results and concluding on the anthropogenic background on scavenged particles by rain:
"The anthropogenic origin of particulate TM and P have been reported by several studies on atmospheric deposition monitoring in the western Mediterranean (e.g., Guieu et al., 2010; Sandroni and Migon, 2002; Desboeufs et al., 2018). For example, Desboeufs et al. (2018) showed that there is a large contribution of anthropogenic combustion sources to the P, Cr, V and Zn background deposition fluxes. Aerosol composition monitoring over the Mediterranean coastal area showed the role of land-based sources and ship traffic sources on TM contents (Bove et al., 2016; Becagli et al., 2017). As all the deposition measurements sites were located in coastal areas, it was difficult to exclude the influence of these local sources for explaining the observed anthropogenic contribution. Here, EF values showed a clear anthropogenic signature for all TMs except Ti, Fe and Mn in the offshore Rain ION sample. In particular, the EF of Zn in Rain ION was on average five times higher than the EF found in the rain samples previously studied from coastal sites in the Mediterranean region (Özsoy and Örnektekin, 2009; Al-Momani et al., 1998; Losno, 1989). Nevertheless, extremely high enrichments of Zn in

rainwater have been reported from island sites in the Med Sea, for example Frau et al. (1996) reported geometric mean EF of ~6500 in both dust-rich and dust-poor rains from two sites in southern Sardinia, and Fu et al. (2017) reported EF >1000 for Zn in atmospheric insoluble bulk (wet+dry) deposition on Lampedusa Island. As previously discussed (section 3.1.1.), Rain ION was representative of a Mediterranean background marine rain event. The Zn EF at station ION was the same order of magnitude as at these island sites which suggests a high anthropogenic background signal of Zn even in open Med. More generally, the high EFs in Rain ION mean that even over the remote Med Sea, the chemical composition of background aerosol particles is likely continuously impacted by anthropogenic sources."

Line 468. Define EF here: *it is already defined in section 2.*

Line 469. Replace Earth with continental crust : *done*

Line 471. Delete slightly. You have stated that TMs with EFs >10 are considered significantly enriched. : *done*

Line 474. Was the atmospheric bulk deposition on Lampedusa bulk wet deposition or bulk wet + dry? If so, for clarity best to just say rain. If bulk dry you need to state that too. : *done*

Line 485. New paragraph. '...particulate TM and P concentrations in seawater have...': *We did not change since the cited references are for TM and P concentrations in deposition, not in seawater.*

Line 497. How does this study compare to the factor of three increase in concentrations between background and dust influenced rains – use your values – and state how this would impact EFs – more Al drives down EFs, Al predominantly from mineral dust, etc.
+
Line 505. Vague, just say that concentrations were higher and EFs suggest the reason for this was the impact of the dust plume. The EFs don't really tell us this. This section needs tightening up.
R: the paragraph on dust contribution in EF values have been completed as suggested by reviewer (values and explanation on the effect of Al concentrations on EF values) p21, L541:
"The EF values of TMs for Rain FAST were significantly lower than for Rain ION (Fig. 7) but similar to Saharan rains (Guerzoni et al., 1999b; Özsoy and Örnektekin, 2009), confirming the dust signature for this rain. The comparison between dust-rich and background rains generally reveals a net difference of concentrations (at least higher by a factor 3 in dust-rich), notably for Al, Fe, Mn and Cr (Guerzoni et al., 1999b; Özsoy and Örnektekin, 2009). Here, an increase in concentrations between rains ION and FAST was observed for the majority of TMs: Al (x28), Ti (x50), Mo (x23), Fe (x13), Mn (x9), V (x5), Pb (x3.5), Co(x3) and Cu (x2) but also for P(x4) (Table 2). The combination of higher concentrations and EF values <10 found in Rain FAST show that the dust contribution was important on deposition fluxes of many TMs and P during this event. However, the high Al concentrations in rain FAST drives mathematically down EF values, masking potentially other sources signature."

Line 516. Can you suggest a reason for the difference?
R: We have added two sentences to suggest an explanation (p22, L568):
"It is known that anthropogenic Fe is more soluble than dust-bearing Fe (Desboeufs et al., 2005, Jickells et al., 2016). Regarding the evolution of TM emissions (see section 4.1.1), we suspect that this difference could be due to a higher contribution of anthropogenic signal for Fe in dust-rich rains in 90's in Sardinia that in the recent rain samples."

Fig. 6. Could you make the two panels the same size and the bars the same width?: *done (now Fig. 7)*

Line 525. Could draw parallels with aerosol samples here, e.g. Jickells et al. (2016)

Line 535. Dust loading is only one proposed control and may not be a direct control – provide explanation e.g. perhaps the CaCO3 concentration is or the reduced impact of acidic gases, relative humidity, etc. – ref Baker et al., 2021.

R: We agree that the dust loading is not a direct control. Here, we noted that the dust load reflects the contribution of dust vs anthropogenic particles in rain samples. Even if the dust loading have an indirect control on solubility, e.g. through modification of pH due to CaCO3 dissolution, or through the saturation effect of high dissolved Fe concentrations, the Mediterranean rains monitoring showed that the TM solubility decrease with the dust load increase because high dust load implies a more crustal origin of metals (Theodesi et al., 2010).

Line 538. Therefore, your data does not support this argument. Disagreement within statement. The EFs close to crustal TMs in Rain FAST combined with the higher concentrations point to the role of mineral dust in reducing the fractional solubility of TMs in rainwater – or the presence of mineral dust overwhelms the background signal resulting in a net decrease in fractional solubility – although the net effect is the same, I favour the latter explanation.

R: We agree that the arguments on this section were not very clear and that the discussion was not supported by the results. We have rephrased all this section and described more in details the different steps of the discussion from reviewer's proposition:

"From our two rain samples, it is difficult to propose a control explaining the difference in solubility values. However, the pH values were very close in the two samples (Table 2), excluding a pH effect on solubility values. A much lower solubility of TMs in Rain FAST is consistent with the EFs indicating a crustal origin of TMs in Rain FAST (Fig. 7). As discussed before, background atmosphere in Med Sea seems to be continuously influenced by anthropogenic particles. Even in dusty-rain FAST, it is highly probable that a part of metals presented an anthropogenic imprint, not visible on EF values but with solubility similar than in Rain ION. Thus, the decrease of solubility between the two rain samples could be either due to the lowest solubility of TMs in mineral dust (as suggested by from aerosol leaching experiments), either results from the presence of mineral dust which by increasing the TM total concentrations overwhelms the anthropogenic background signal, or both."

Line 540. Mo has a high concentration in SW. If it has a predominantly marine source this could explain your uniform solubility. It would not explain your increase in EF necessarily but if there was an increase in seasalt aerosols it could do. You have the data to test this hypothesis. How reliable was the Mo solubility data? How close to LoD and blanks?

R: We had indeed completely neglected the marine origin of Mo in our discussion to explain the non-crustal part. We are very grateful to the reviewer for directing us to this explanation. The uncertainties of solubility were displayed on Figure 7, and confirm the solubility stability is significant in the two samples. So we have used this explanation in EF discussion (4.1.2) and completed the arguments by calculating EF ratio in rain relative to seawater (p20, L514):

"Mo is the most abundant TM in seawater (Smedley and Kinniburgh, 2017) and in particular in Med Sea (see section 3.3). Thus, at the difference of other TMs, the non-crustal part of Mo could be associated to seasalt aerosols rather than anthropogenic signal. The EF ratio relative to seawater ((Mo/Na)seawater = 8.9 10^{-7} in mass ratio, Millero, 2013) were 7.4 for Rain ION and 4.6 for Rain FAST confirming the marine origin of this element in both rain samples."

And mention that in solubility discussion (p23, L597): "As discussed from EF values Mo was associated to seasalt aerosols in both rain samples, explaining the similarity of solubility."

Moreover, due to the marine origin of Mo, we have excluded it in the comparison between atmospheric fluxes and marine stocks (section 4.2.2)

Line 551. How large? *done: "here ~25% RSD for total precipitation and 1 to 74% RSD for concentrations"*

Line 566. This is different than the Al percentage used in your EF calculations (~ 8% Rudnick and Gao, 2003). Your EFs would be lower if you used concentrations based on 7.1% Al (Guieu et al., 2002) or your fluxes would be higher if you used the Rudnick and Gao %. It would be worth noting this variability. There is certainly an argument for using Saharan elemental ratios in your EF calcs (as discussed in Shelley et al., 2105).

R: We used 7.1% Al to estimate the dust flux in order to compare this value with the flux value found by Bressac et al. (2021) on the same event since they used this value. Doing the flux calculation with 8% Al gives a value of 58 mg. m^{-2} which falls within the uncertainty of the dust flux given here. The variability related to Al content is lower than the one related to the total precipitation. We do not believe that a discussion of the variability of dust flux is necessary here, since the absolute value is not relevant to the discussion, which focuses on the magnitude of the event (only the order of magnitude is key and it is not modify by the using of different Al content).

Line 563. Fluxes aren't measured directly so reported, calculated or estimated are better words.: *done. We changed by "calculated"*

Line 565. Reword: The aerosol columnar concentration during the dust event was estimated to be between...: *done*

Line 566. This is x3-4 higher than your flux estimates. A sentence to clarify why you support this view.:
R: the explanation is already provided in the next sentences suggesting that all the aerosol column was not totally washed-out by rain as observed on depolarization values from lidar measurements.

Fig. 7 caption. Add 'Note different scales on the y axes.': *done (Now fig. 8)*

Fig. 7. It would be useful to see the Al wet deposition flux as this is what you are using as the basis to estimate the bulk wet deposition fluxes. It would also be nice to have a third panel showing the ratio of dissolved to particulate TMs – or Kds.: *done (Now Fig. 8)*

During the reprocessing of Figure 8, we identified that we had a problem of unit conversion in the used script to produce the figure from the dissolved TM concentrations in pM in the rain ION. In consequence, all the estimated fluxes were higher than a factor 10. So we have corrected all these fluxes in the revised figure.

Line 579. Explain how these two parameters differ. Are they not the same thing in the context of this work?:
R: It is probably an English problem since here the two parameters are "chemical composition" and "total precipitation" which are not the same thing. We have reworded to remove this ambiguity: "derived from the chemical composition and total precipitation of rain samples"

Line 579. Explain why. They (Co, Cd, Mo) also have the greatest uncertainties – low concentrations and high blank contributions:
R: The uncertainties on Co, Cd and Mo are now described in several parts of the manuscript, notably in section 2.1. for LoD and 3.2 for uncertainties on their concentrations. We do not think that we need to re-mention here this fact (moreover, the uncertainties appear clearly on the figure 8).

Line 583. Because these elements (Cr and Ti) were primarily found in the particulate

phase? *We have added a comment to explain this point: "due to their low solubility in rain FAST"(p25, L652)*

Line 584. Emphasised is stretching it for data from just two samples
R: The sentence starts by "Our results suggest", we believe we have exercised due restraint in extrapolating our results.

Line 586. With the exception of Mn, these are predominantly considered pollution derived elements. It would be worth mentioning this.
R: We have added a sentence to this effect: "Yet, these former elements (except Mn) are usually considered issued from anthropogenic sources."

Line 587. Poorly soluble, lithogenic elements. Al should also be reported here. I'm assuming it showed the same behaviour as Fe and Ti?
R: We didn't mention Al in this part which is focused on trace metals. Al being used for calculating dust flux, it is also poorly soluble and indeed its behaviour is very close to Fe and Ti.

Line 596. Do you mean the orders of magnitude range? Say how many orders of magnitude.
R: No, we mean "orders of magnitude", not range, e.g. pNi fluxes around 10^{-2} µmol m^{-2}, whereas pCd fluxes are around 10^{-4} in case of wet dust deposition.

Line 590. What does 'dust-related elements' mean? All elements discussed or only lithogenic elements?
R: Dust-related elements are also the lithogenic elements. It is a question of vocabulary, in the atmosphere community working on desert dust emissions and transport, the term lithogenic is not used unlike the oceanographic community. To precise, we have added desert dust-related elements: Fe, Mn and Ti (p26, L662)

Section 4.2.2. I'm wondering if concentrations or partitioning are better words than stocks (this applies throughout the paper).
R: Here, they are neither concentrations, nor partitioning. As described in the method, we have integrated TMs concentrations on the depth of mixed layer. The result is so the total content of TMs by m-2 in the mixed layer. Another term could be inventories of TMs. We used "stocks" since that implies also the notion of availability of these TMs.

Line 594-597. Move to Methods.: *done*

Fig. 8. Move to methods + Line 671. It would be useful to see this – put in Supplementary Material.
R: We have added a section 2.4 "Concurrent sampling strategy" detailing the information about sampling time-resolution and calculation of ML enrichment after rain (Now fig. 1)

Fig. 8 caption. Given how quickly the SML TM concentrations respond to atmospheric inputs and their relatively short residence times in the SML (mins - hours in Ebling and Landing, 2015), how can you be sure that your SML sampling resolution was capturing the impact of the wet deposition events on the SML, especially given that wind speed (and sea state) is a critical factor in determining the integrity of the SML?
+
Line 614. As it reforms so quickly and rain disrupts it, how can you be sure that the SML sampled after the rain at FAST was in contact with the atmosphere and accumulating TMs from wet deposition?
+

Line 616. Suggesting that there was a surface advective current?
+
Line 619. Dissolved or particulate or both?
+
Line 623. I wonder if this is because of the mismatch between SML sampling and rosette casts.
+
Line 632. Again, the SML samples may not be representative of the wet depo inputs. In contrast, the ML likely was
+
Line 655. Some ML residence time calculations could be of use here
+
Line 648. If we assume that the SML samples were representative of this layer before and after rain (and even if we don't), the similarity in Mo concentrations suggests to me that this element is not primarily delivered by atmospheric inputs
+
Line 661. Many people have shown a delayed response of the dissolved pool to atmospheric inputs and hypothesised why there is a lag.
+
Line 671. I'm not completely convinced it does because of the resolution problem. It certainly suggests it might given Tovar-Sanchez's findings of increases 24 h after rain but the signal is likely to be (significantly) diminished after this time.

R: Figure 8 is now Figure 1. All the discussion excludes now the case of SML and the case of Mo due to its marine origin. Moreover, as mentioned in the initial manuscript "For ML, the large variability in total and dissolved stocks between the two casts ML25 and ML27 before the rain makes the establishment of a background concentration levels before rain difficult.". Indeed at the difference of FAST station (Guieu et al., 2020), we have not the proof that the surface lateral advection was limited during ION station and hence that the ML sampling "before" rain (ML27 in figure 1) which occurred 2 days before the rain, was really representative of initial conditions. In consequence, in order to clarify our discussion on the role of atmospheric inputs in the marine TM stocks, we have decided to exclude also the results concerning ION station and to keep only the case of FAST Station which the most constrained both by time-and depth- resolution samplings and by analysis of dynamic conditions of water masses (Guieu et al., 2020). On this basis and the reviewer's comment, all the section 4.2.2. was completely rewritten by integrating Kd values and residence time calculations (from Bressac et al., 2021 and Tovar-Sanchez et al., 2020) and Figure 9 was simplified. The interpretation of results was also discussed in term of post-deposition processes.

Line 629. Additional to what? *R: we have changed by "external"*

Line 686. Delete 'at the scale' and replace with 'in the': *done*

Line 704. Which ones? Theodosi reports this for the TMs he studied but not your full suite. There are some that do, some that don't. See Jickells et al., 2016 and Baker et al., 2020. I appreciate these papers report data for the Atlantic rather than the Med but as they report data from Saharan and European air masses they are relevant.
R: Jickells et al. (2016) show that the dust loading effect is current for all dust-related metals, as Fe or Al. By considering recent results of Baker et al.(2020), we have completed the sentence: " Fe, Mn and Pb solubility decreases with increasing dust load in Mediterranean rain samples (Theodosi et al., 2010), suggesting that this estimation is probably a maximum flux for such deposition event. However, recent studies from aerosol collected in Atlantic Ocean showed that Co and Mn solubility was little affected by dust load at the difference of Fe (Baker et al., 2020)."(p29, L776)

Line 734. Only dissolved or dissolved and particulate?
R: This conclusion is only for "dissolved" since the comparison in Fig. 10 was limited to dissolved fluxes and stocks

Line 752. How about an extra statement suggesting additional dry deposition sampling to directly compare the inputs of wet and dry deposition in future? The contribution of dry deposition seems to have been overlooked in this study, in the sense that there is no comment about the contribution of dry deposition. What is thought to be the relative contribution of wet-dry dust deposition events in the Med. Is wet deposition thought to have a disproportionally large impact on TM seawater concentrations during stratification?
R: We have added a specific comment on dry deposition in the conclusion (p31, L824): "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 of dry deposition in open Med Sea are needed in order to estimate its contribution in TMs atmospheric inputs."

Technical corrections

There are many examples of incorrect agreements and syntax. I have attempted to catch them but may have missed a few. For the most part, these small errors do not impact too much on the readability of this manuscript, but it would be greatly improved by correcting them. I have included an edited version of the manuscript to help identify where these occur as they are too numerous to include in this review.

All corrections have made in the revised manuscript

It seems unnecessary to use the Med Sea abbreviation as it is used inconsistently throughout the manuscript.
R: We homogenised the using of Med Sea

Trace metal or trace element? You switch part way through.
R: We homogenised the using of trace metals (TMs)

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 intense rains favours the role of wet deposition. It is the reason why the paper is here focused on wet deposition. We have added sentence in this purpose in the introduction (see specific comments) and 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, L704: "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". We believe we have exercised due restraint in extrapolating our results. For example p29, L748: " 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 marine TM 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 can be impacted by local sources.

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 p28, L670 to 682 (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, L805: " 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 period 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 (L420): "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 emissions 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 emitted aerosols.

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 L586: "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 events 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 624 to 632, 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