#### Review bg-2021-87

### **Overview:**

This manuscript presents data on dissolved and particulates phases for iron and aluminium collected in the Mediterranean during the PEACETIME project. During the expedition a number of dust/precipitation events occurred and the authors attempt to derive dust fluxes from measurements before and after such events. Unfortunately the are missing some before data and have used data from other stations to try to bridge this gap, without taking into account possible differences in hydrology and past depositional histories between the stations concerned, this results in what appears to be a major overestimation of the estimated dust fluxes at some stations due to the use of inappropriate initial conditions. The lack of a Lagrangian hydrographic context for the stations also suggests that horizontal variations are being interpreted as temporal changes in the vertical.

### **General comments:**

## Influence of Hydrography, Eulerian or Lagrangian approaches:

A critical omission at present from this work is that there is no information on the hydrography and what is going on in the water. For a process study this is a major flaw as all too often changes in water masses may be interpreted falsely as the effect of rapid in water column processing. This is the reason why Lagrangian experiments are carried out in the ocean (Abbott et al., 1990; Boyd et al., 2005; Jickells et al., 2008; King et al., 2012; Krom et al., 2005; Law et al., 2005; Law et al., 2001) in order to remove the effects of water mass movement as much as possible. While the optimal way to carry out such a study is to use a tracer such as SF<sub>6</sub> (Law et al., 2001), it can also be as simple as putting in a drifting buoy as the sampling point (King et al., 2012). It appears from the description in the associated paper (Guieu et al., 2020) that the same eulerian point was occupied for each station and no attempt at a quasi Langranian study was made, though that work mentions that a Lagrangian adaptive sampling strategy was employed, but this apparently refers to using satellite data to avoid placing stations at fronts or in eddies.

### Absence of a hydrographical context:

I am really surprised that the authors did not think to use salinity as a tracer of recent precipitation events as this would also allow an estimate of the amount of rain that has fallen if there is minimal horizontal advection of water masses. It also appears that there is data for transmission and a LISST-Deep, both of which presumably would have picked up changes in the particles in the deep waters where the apparent excess particulate aluminium was located. Referencing data to density levels and examining the different water masses present (Schroeder et al., 2020) would help to resolve some of the issues around the lack of any hydrographical context in a future revision.

### Dissolved Al removal dominant over release from dust:

Using the median profile as the initial starting point for looking at changes in the particulate concentrations has a number of apparent flaws that are not fully explained or investigated in the current manuscript. (i) There may be significant differences in the hydrography between the station used as the median profile and the experimental station (ii) similarly the experimental station may include a transient nepheloid layer due to water mass differences and lastly this may lead then to (iii) the time scales of the inferred changes being different, that is a seasonal or slow moving

transient change is interpreted as an instantaneous response to a dust event. In the present work section 3.3.1 at present does not give the reader a good idea of how the fluxes were calculated and what assumptions were made in the calculations. It appears that the difference between the median profile and the most recent profile is assumed to represent the total flux of particulate aluminium to the region in a single event, this seems very optimistic.

*Elevated particulate aluminium at depth but what about iron?* One of the main mysteries in this paper is if there is also a vast excess of particulate iron at stations ST04 and ST05 as there is for the particulate aluminium as this data is not provided (or at least I could not find it). It would be useful to include the particulate Fe data in Table 2 for instance as a check on the dust deposition estimates using Aluminium.

*Time scales*: Earlier work (Caschetto and Wollast, 1979) showed strong seasonal changes in dissolved aluminium concentrations, which would suggest that the residence time for dissolved aluminium is on the order of months and certainly less than a year for the Mediterranean. At present the manuscript basically interprets every change on the same timescale as between repeat measurements but care has to be taken when the sampling is not truly Lagrangian as you may confuse variability at depth with fast removal or dissolution processes. What the data in this paper do inform about however is the effect of deposition in surface waters and reconciliation of the timescales involved that control the strong salinity relationship with dissolved aluminium (Rolison et al., 2015) as it fits well to a two end member situation with low Al Atlantic surface water and high Al Eastern Mediterranean waters, however this implies rapid mixing of surface waters in the Mediterranean to maintain the observed gradient. Thus it would be helpful to examine in the current work how horizontal mixing may dilute and transport dissolved aluminium after a dust event. I appreciate that in the current manuscript this aspect is downplayed so as to simplify flux calculations but it does appear that it is a critical aspect for maintaining dissolved aluminium concentrations in the Mediterranean.

Work on thorium isotopes in the Mediterranean (Roy-Barman et al., 2002; Roy-Barman et al., 2009) also suggests that the dissolution of continental particles stored in the margins needs to also be considered. This does not appear to have been examined in detail by subsequent researchers but could be explored here briefly. Similarly for the elevated particulate aluminium it would be useful to explore this with reference to earlier work on the pressure effects on aluminium speciation (Moore and Millward, 1984).

*Role of colloids missing from this work:* A further obvious omission in this paper is any mention of the role of colloids for Fe (Bergquist et al., 2007) and Al (Dammshäuser and Croot, 2012; Moran and Moore, 1989) in the surface ocean. Given that colloidal Al could also be part of the measured particulate aluminium pool observed in this study it would also be of benefit to the reader if there was some discussion on the potential role of colloids in controlling the fractional solubility of Al in seawater (Dammshäuser and Croot, 2012; Dammshäuser et al., 2011; Dammshäuser et al., 2013; Moran and Moore, 1989).

*Biogenic silica in Saharan Dust*: Saharan dust can contain a significant amount of biogenic silica (Folger et al., 1967), either as phytoliths or freshwater diatoms. So there is the possibility that the biogenic silica may not be formed in situ. Alternatively the biogenic silica may be overestimated during the analysis due to lithogenic dissolution (Ragueneau et al., 2005). This typically is estimated using dissolved Al as a tracer for the lithogenic fraction, which appears not to have been done in this case. This is an opportunity missed really as a sequential dissolution protocol looking at both the Si and Al would have provided strong evidence for Al weakly bound to the particles (i.e. that released

with an acetic acid step as was done in other studies (Berger et al., 2008; Landing and Bruland, 1987; Planquette et al., 2009). At present it looks like the biogenic Si is overestimated.

*Precipitation estimates:* There are satellite data available for precipitation <u>https://gpm.nasa.gov/data</u> - did the authors examine this dataset to examine the spatial and temporal scales of precipitation during the expedition? This would likely support the model runs used currently. There are also rain rate data included in the Guieu et al. (2020) work associated with this expedition.

*Error estimates*: In the dust flux estimates there is no inclusion of error estimates and it would be useful to include these to constrain the values accordingly.

# Specific comments:

- Line 57: In this context the work of Li et al. (2013) along with Moran and Moore (1988) are good examples of the apparent biological drawdown of Al.
- Line 68: It is not that unexpected as the deposition of dust is known to scavenge other elements from the water column also.
- Line 69: Do you mean fractional solubility with regard to aerosol solubility? As iron solubility in the water is a different measurement (Baker and Croot, 2010).
- Line 94: Please clarify that this is a Kevlar cable with a conducting insert in it and include information on the type of CTD that is on the rosette and the parameters measured on it.
- Line 95: The classical rosette is an aluminium oxide coated rosette? Please provide more information on this as the data in Supp Fig 2, do suggest that it is non-contaminating but the same data also suggest that the trace metal clean rosette could be contaminating (two obvious fliers). Alternatively it suggests there were major changes in the intermediate waters over a few hours (less likely).
- Line 95: Were nutrients taken from both sampling systems? There is no information here on if nutrients were taken and if so, how they were analysed.
- Line 112: There are no GEOTRACES consensus samples reported here for Al. Does this mean none were run? What data do you have that this method was giving good values?
- Line 113: Are the data corrected for the reagent blank?
- Line 120: The GSC consensus value is 1.54 ± 0.12 nM for Fe and the data are available here: <u>https://www4.obs-mip.fr/wp-content-</u>

omp/uploads/sites/31/2020/03/2019\_Consensus\_Values\_2009\_samples.pdf

- Line 125: The GEOTRACES Cookbook is citable (Cutter et al., 2010).
- Line 140: There are two or more fliers in Supp Fig 2, all of which come from the trace metal clean rosette so what is the reason for this? Some are in the surface so this is understandable but at 400 m at ION and 2000 m FAST, this seems more like a bad bottle as it would not be expected that the water masses would change the much in the few hours between casts. Are there any other elements that can help with determining if it is contamination from a bottle?
- Line 160: The usual description for this is that it is a sequential leach using the method of Paasche (1980) as modified by Nelson et al. (1989) using the spectrophotometric method of Mullin and Riley (1955). I would avoid using the thesis as a citation as it quotes the wrong paper for the silicate method and while it has the authors correct for silicate, the journal reference is actually a paper on phosphate (Murphy and Riley, 1962).

- Line 165: Table 1: The data for BSi, LSi, Al and Fe have high variability, with apparently one standard deviation being almost the same as the mean more so in many cases than the variability in the mass fluxes. This is therefore an inherently noisy data set, which likely represents the challenge of short term deployments, so it is important that this is also discussed in the manuscript in more detail as concentrating on the average fluxes presents a somewhat distorted picture.
- Line 172: Was this always in the mixed layer as observed by the hydrographical measurements. See the general comment above regarding the absence of any hydrographic context in this work.
- Line 185: How was the mixed layer defined here? A density or temperature criteria or something else? More details should be supplied.
- Line 219: As mentioned in the general comments above, there is satellite data for precipitation so there is no need to invoke model predictions. The authors could also use the change in surface salinity as an indicator.
- Line 239: Can the authors provide a little more information on how this dust flux was determined was it based on direct measurements of dust in the rain or via a proxy? This is important as without being able to see the Desbouefs et al. paper is may mean that this is actual a circular argument (e.g. that paper uses dissolved Al in the water to estimate the dust flux).
- Line 252: Using the median pAI profile without providing a hydrological context seems flawed in that it ignores differences in the hydrography. Additionally we are not provided with any statistical oversight of what the median profile represents. See the general comment on this above also.
- Line 256: The high particulate concentrations at depth may also represent a nepheloid layer. Is there any transmission data from the CTD to examine this possibility? The Guieu et al. (2020) work indicates that there was a transmissometer and a LISST sensor onboard that could answer these questions so it would be a good idea to include this data here. I am aware of earlier work which has shown that this area can form nepheloid layers at intermediate depths (Misic et al., 2008). Nepheloid layers have also been shown to be important for scavenging of Th and Pa in the Mediterranean (Gdaniec et al., 2018). Alternatively it could be from a mesoscale eddy so it would be good to rule that also using sea surface altimetry.
- Line 267: the units here are g m<sup>-2</sup> but no time frame is given, this is then compared to an annual flux in g m<sup>-2</sup> y<sup>-1</sup> so how is the reader meant to compare this when we are not told how the deposition flux is calculated? The key missing piece of information is the assumed residence time for particulate aluminium and it seems here that the assumption (unstated) is that the excess particulate aluminium comes from a single dust event. See the general comments on this above.
- Line 270: This statement is confusing, as the inference appears to be that the excess dissolved Al all arrived in a single event? It is also not clear then how this relates to the rain event where the dust flux is estimated at 40 mg m<sup>-2</sup>.
- Lines 277 & 281: These two sentences are opposing each other and I don't follow here how if the deposition is assumed to be homogeneous that then the spatial variability indicates precipitation patchiness and not sampling bias. What type of sampling bias is inferred here?
- Line 285: Table 2 deposition not 'depositon'
- Line 285: Item 4: Why is the loss correction made? Particulate Aluminium in the traps is still in the water column at the time it is collected by the traps so it seems to be some sort of double accounting.
- Line 285: Item 6: The authors should check these numbers as using 7.1% and a molecular weight for Al of 26.98 gives a dust flux of 9.18 g m<sup>-2</sup> for the ST04 station when using 24169  $\mu$ mol m<sup>-2</sup> as the pAl input. Similarly 6.98 g m<sup>-2</sup> for ST05.

- Line 289: The vertical profiles for the particulate Fe data are not shown in this work, only the integrated values and the depth/time when sampled.
- Line 298: This is perhaps a good example of why a Lagrangian framework is better to use than a eulerian one as clearly different water masses with different deposition histories are passing through the same site so the interpretation here based on a eulerian approach is flawed. See the general comment on this above.
- Line 300: So what is the excess Al seen at ST04 and ST05, slow sinking dust particles? Why then are they not seen at FAST?
- Line 305: As Kd is a ratio, using a normalization approach as shown in Figure 3 makes it harder to follow. Furthermore the use of the 4<sup>th</sup> of June for the 0-200 m inventory makes comparison with the 0-20 m data even more problematic Perhaps plotting the data as percentage of total Al might then be a better way to show how the relative amounts changed in the particulate and dissolved pools.
- Line 337: See the general comment about the lack of details on the hydrography.
- Line 340: Please include error bars in Figure 4 as it is not clear what the variability of the measurements is relative to the apparent changes seen here.
- Line 347: Another good reference for this which includes data at high dust loadings is Shelley et al. (2018) and in that work it is clear that the fractional solubility is very low under high dust loadings. Though it may be higher when deposition is as rain.
- Line 359: What is meant by '1D dynamic condition' here? That there is no lateral mixing? If the deposition is so patchy initially then of course lateral mixing will smooth this out quite quickly. Additionally horizontal mixing is typically orders of magnitude more than vertical mixing so you have to assume then that the deposition is homogeneous for the area you are sampling in (see the general comment on this above).
- Line 371: What do the grey filled circles correspond to? All the other stations samples in this study?
- Line 373: That an individual measurement is higher than the average, or that of a higher dust loading, is no surprise and does not require that there a rapid removal mechanism needs to be invoked as it depends on the time scale for the processes. In this case it the dust loading appears to be critical and as mentioned above there is an inverse relationship to the fractional solubility (Shelley et al., 2018).
- Line 391 and 405: Figure 6 (b). If this is a simple two component model the data would actually fit a curve if the value of Fe:Al was constant in the biogenic and lithogenic silicate fractions using a linear function as is computed here the intercept is not the actually the value of Fe:Al in the biogenic fraction and is in fact an underestimate of it. The authors can easily check this for themselves by plotting over the top values of the Fe/Al ratio at different LSi/BSi ratios assuming fixed Fe/Al for the end members. It also implies that the Fe:Al ratio for the LSi is less than the biogenic value which is what it should be for a bioactive element like Fe. Moral of the story is always be wary of linear fits to ratio plots!
- Line 405: Figure 6: It would be helpful for the reader if you also include the Fe/Al ratio for Saharan dust or continental crust.
- Line 405: Figure 6 what about the Fe/Al for stations ST04 and ST05 where the large excess of particulate aluminium is found, if this is indeed dust going down through the water column it should have a similar Fe/Al ratio as for the dust itself but the manuscript is completely silent on this! So please show the Fe/Al for these stations and the particulate iron profiles.
- Line 419: This is for dust inputs of 1.6 and 8.8 g m<sup>-2</sup> for a single dust event! With almost no change in the dissolved iron concentrations, this seems rather remarkable.

- Line 445: How about a nepheloid layer, resuspension of material transported to the interior. In regard to the timescales, it is also important to consider the role of colloids here (see the general comment on this above).
- Line 456: Though this process is also part of the low mesopelagic Fe regeneration efficiency referred to on line 445 so there is a contradiction here if bacterial remineralization is being suggested here but not earlier.

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