Interactive comment on "Contrasted release of insoluble elements (Fe, Al, REE, Th, Pa) after dust deposition in seawater: a tank experiment approach" *by* Matthieu Roy-Barman et al.

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General comments:

The manuscript by Roy-Barman et al. will be an excellent contribution to Biogeosciences, and to chemical oceanography in general. The work constitutes a novel way to estimate the mobility of mostly-insoluble elements during dust dissolution, and the findings are highly relevant to a bourgeoning literature on using Th isotopes to quantify dust dissolution and trace element inputs. The methods, data quality, and interpretations are generally sound. Before publication, there are a several points that need to be addressed, and I encourage the authors to reconsider alternate possibilities for

their recommendations for the use of dissolved Th isotopes to quantify dust fluxes (see general comments below).

Specific comments:

1) I had a hard time following the REE section of the results (section 3.3.). I think this is because of the way Figure 2 is plotted. The sharp increase in REE at the start of the FAST experiment is difficult to see in Figure 2b because the t=0 and t=1 points look like they are at the same place on the x-axis. Maybe it would be good to label the t=1 point as being immediately after dust addition (if this is the case?). It should also be clarified that the rapid increase in REE is only seen at FAST, and not at ION (unless I am having a hard time seeing this at ION because of the same plotting issue).

This same point holds true for ^{232}Th and Figure 3. Also, page 7 line 2 suggests that ^{230}Th/^{232}Th ratios as a function of time are plotted in Figure 3, but I do not see them there – I would quite like to see these ratios plotted with sampling time!

2) I'm curious from an analytical perspective, though not necessarily for the paper, where they think their high ^{231}Pa blanks might come from, since there is so little ^{231}Pa in nature. That said, the precision the authors achieved on such small quantities of ^{231}Pa, in such small samples, is remarkable.

3) I am curious about the inferred dissolution of detrital calcium carbonate in the dust mentioned in 3.6. even though this isn't the main point of the paper. This process must be occurring above the calcite saturation horizon – what is driving this dissolution?

4) I struggle with the arguments in the first paragraph of section 4 (page 7, particularly lines 34-41). What time interval are these waters seeded over? The dust input units given of 10g dust/m2 are not flux units without a time component, so it is difficult to assess whether this is a feasible dust flux compared to the highest dust pulses or mean annual dust fluxes. Because the dust corresponding to the average areal input was added in such a short time period, I think the sentence in lines 37-38 "Hence, the

PEACETIME experiments..." must be taken out.

5) The difference in dissolution kinetics between Th/REE and Al is intriguing, and I would like to have seen the authors speculate on what drives it. Are different mineral phases releasing Th/REE than Al? A back of the envelope calculation, 3.6g dust added, 15% of it presumed to be CaCO3 that dissolves, 0.5ppm hydrogenous 232Th in lacustrine carbonates (Lin et al., 1996) and in pedogenic carbonates (Ludwig and Paces, 2002) gives 1160 pmol of 232Th that could be released into the tank from detrital CaCO3 dissolution. Dividing this by the 300L tank volume gives a Δ[232Th] of ∼3.9pM – very close to the 3-5 pM initial rapid increase in 232Th observed at both ION and FAST. This calculation should be done by the authors for REE as well. If this is the case, then Th dissolution in dust may be highly dependent on the detrital carbonate content – which would be a really interesting result. Regardless, it is important to explicitly note that Th and REE release from dust is clearly controlled by multiple phases – a fast dissolving one and a slow-dissolving one – and that this is different from Fe and Al release.

6) The relative fractional solubility of metals is used in calculations of dissolved metal flux from dust derived from thorium isotopes (Hayes et al., 2018; Pavia et al., 2020). It would be fantastic to calculate the release ratios of Fe, Al, and REE to Th (along with their uncertainties) and provide these in addition to Table 1. These would be an excellent addition to the aerosol-based solubility estimates currently used, though it would also be important to note that using these relative solubilities in Th-based Fe flux estimates could be biased if the differences in metal release kinetics between Fe and Th release are sufficiently large to decouple the release depths of Fe and Th as fine lithogenic particles aggregate and sink from surface waters.

7) There is a mismatch by a factor of 10 between the units on y-axis of Figure 6 and those cited in the text in section 4.3.

8) The points regarding differences in solubility estimates of Th between previous re-

sults and this study made in 4.4. are very important. Th solubility is one of the largest sources of uncertainty in Th-based dust flux estimates (Pavia et al., 2020), and different methods give different results. However, authors' points about lateral 232Th inputs are not justified by the data presented in this paper. The solubility of 232Th does not alter the removal residence time of Th in the upper water column. The 230Th-based removal residence times in the upper 250m of the South Pacific Gyre (depths over which Th-based dust flux estimates are integrated) are 1-2 years (Pavia et al., 2020). Note that the South Pacific Gyre is highly oligotrophic, similar to the Mediterranean – so 1-2 years is likely on the longer end of Th residence times for the upper 250m in the ocean. The diffusive lengthscale for lateral diffusion of Th in the upper water column, given by $\Delta x = \sqrt{(2K\_H \, \Delta t)}$ (Roy-Barman, 2009), for a residence time of 2 years and a lateral eddy diffusivity of 10ˆ3 m2/s, is 355km. So, regardless of the 232Th solubility, lateral lithogenic inputs from the continents should not affect 232Th distributions in the open ocean beyond this distance. Indeed, the lack of lateral inputs affecting the deep Mediterranean, with longer transport timescales from boundary lithogenic sources than in the upper water column in the open ocean, is noted in the conclusion. More work is needed to determine the role of vertical and lateral exchange on 230Th and 232Th budgets, but the conclusions of this study do not justify the conclusiveness of the statements regarding lateral Th fluxes (e.g. "cannot be done", page 12 line 33; "strongly biased by advective inputs" page 12 lines 31-32). These statements should be tempered accordingly. Similarly, the statement on page 13 lines 13-15 should be adjusted or removed, as the current study does not provide any insights on the importance of advection as a source of 232Th to the open ocean.

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