This manuscript by Shelley et al. investigates aerosol trace element solubility and deposition fluxes over the polluted and dusty Mediterranean and Black Sea regions. The study provides valuable insights into the interplay between natural mineral dust and anthropogenic pollutants by analyzing aerosol samples to quantify the soluble and total concentrations of various lithogenic and anthropogenic elements. The authors explore how different air mass origins, such as North African dust and European pollution, influence these concentrations and examine the impact of atmospheric deposition on nutrient ratios and element budgets in the Mediterranean Sea.

While the observations are limited to a specific period, the paper nonetheless contributes significantly to understanding aerosol trace element solubility and its implications for marine nutrient cycling. Additionally, the manuscript is well-organized and clearly written. Below, I offer some minor suggestions for the authors consideration:

We thank the reviewer for their helpful comments. Our responses are given in blue text below, with changes to the manuscript in italics.

1. **Section 2.3.3**: Some ions mentioned in this section are not discussed in subsequent sections, such as Br^- and K^+. I suggest removing these ions unless they are addressed later. Furthermore, is there data on NH_4^+ recovery that could be included?

   It is true that we do not discuss some of the ions mentioned here, but we prefer to leave them in the list so that others are aware that the data exist.
   Certified NH_4^+ concentrations are not available for ION-915 and KEJIM-02. Recoveries of NH_4^+ in our internal standards were within 5% of their target value (n = 4).

2. **Section 2.5**: As the authors acknowledge, there are substantial uncertainties in the estimation of deposition velocities (Vd). If this section is to remain in the main body of the text, I recommend providing a more detailed uncertainty analysis. For instance, could the authors incorporate model results to assess the impact of these uncertainties?

   We are not sure what type of modelling the reviewer had in mind, but we note that numerical models of atmospheric deposition are subject to very similar levels of uncertainty to those encountered in the calculations we have performed.
   In order to highlight the potential impact of the uncertainty in deposition velocities, we have added ranges to the values given for the potential contributions of atmospheric deposition to soluble element budgets in the Mediterranean. The text below also takes account of a related comment by Reviewer 2.

   "Assuming that the aerosol samples collected during GA04 were representative of deposition to the region, mean dry deposition fluxes (Table 2) during summer (June - August) can account for 11 (3.6 – 32) % (Mn), 3.2 (1.1 – 9.6) % (Ni), 8.6 (2.9 – 26) % (Zn) and 1.0 (0.3 –

2.9) % (Cd) of the annual deficit in the surface budgets reported by Middag et al. (2022) in the western Mediterranean and 1.4 (0.5 – 4.3) % of the Ni deficit in the eastern basin (values in parentheses represent the range due to a 3-fold uncertainty in deposition velocity). The values for Ni in the eastern basin may be lower limits, as noted above."

3. **Section 2.6**: The manuscript states that "samples were assigned to one of five air mass types, indicative of likely aerosol source characteristics as described below." Was this classification based on a subjective assessment, or was an algorithm or objective method used? Were there any ambiguous cases that the authors had to resolve, and if so, how?

   While tools such as openair (Carslaw and Ropkins, 2012) allow trajectories obtained from static locations to be classified using cluster analysis, we are not aware of any equivalent tools that can be applied when the trajectory origin point is moving (as is the case during cruises). From that point of view, our classification can be regarded as subjective to some extent. However, the broad classifications that we have used are quite distinct and there were very few cases of ambiguity in the classification. Most of those were related to dust transport, which can be affected by trajectories at multiple heights, but in those cases we had the physical characteristics (colour) of the samples as an additional aid to classification.

   We have added an additional figure to the Supplement (new Fig S1), which shows more detailed trajectories for the samples highlighted in Fig. 2, as well as adding text to the description of how air mass types were assigned (see response to Reviewer 2).

4. **Section 3.1**: In Figure 2, rather than showing an example trajectory for each air mass type, would it be possible to display the mean trajectory for each class? Additionally, the colors used for the borders of the LE and LAN areas are too similar to the trajectory colors, which makes the figure somewhat confusing. I suggest modifying the color scheme. Lastly, the text mentions 'LAM,' but the figure is labeled as 'LA'—this inconsistency should be corrected.

   We have modified the figure in the following ways. PSA regions are now shown using black patterns to differentiate them and reduce confusion with the colours used for trajectories. The missing "M" has been restored to "LAM". Thank you for spotting that. We have not opted to show mean trajectories because the ship's movement "disconnects" the means from the ship's track, which we do not think aids understanding. Further changes to the figure were made in response to comments from Reviewer 2. The revised figure can be seen in our response to those comments.

5. **Section 3.2**: In Figure 3 (and in similar figures later on), it would be clearer to differentiate air mass types by using distinct colors for each type rather than using different colors for the various elements, which does not seem to add much clarity to the interpretation.

   Thank you for this suggestion. We have modified the figures (3 – 6 and S1) as suggested. Figure 3 is shown as an example below. Note that in this example, low concentrations make

some of the coloured bars difficult to see, but this is not the case in the other figures. Since the order of the types is the same in all the figures, the desired information is still present.

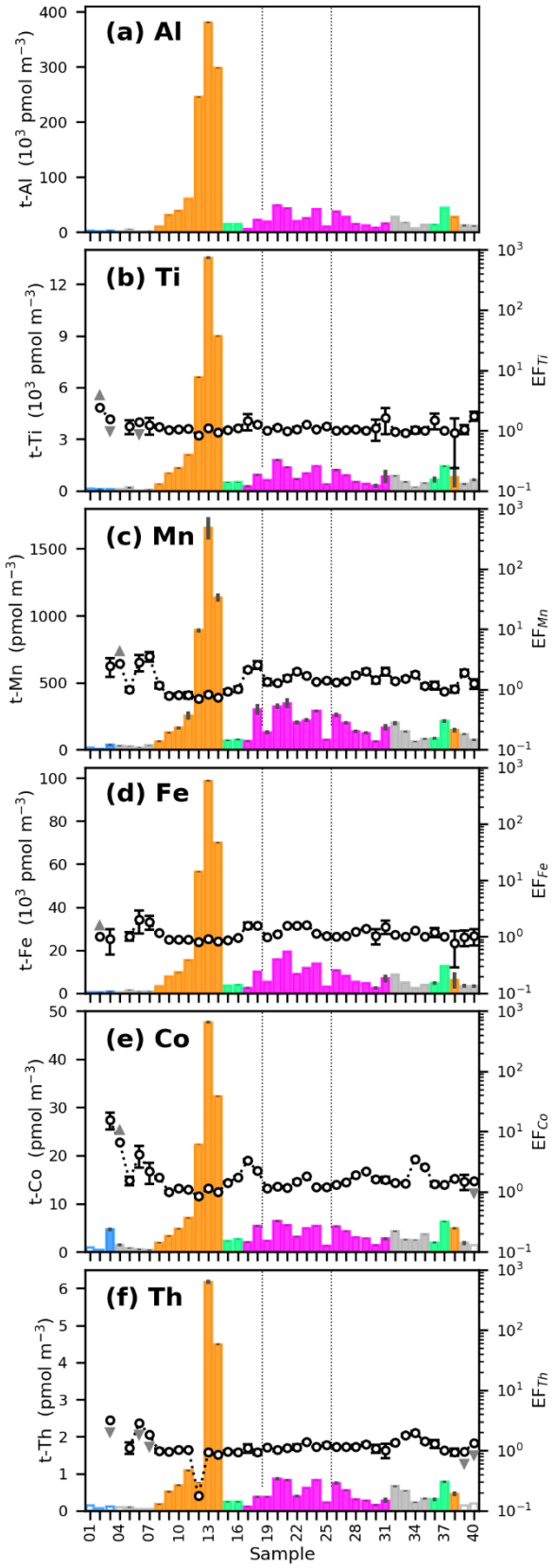

Figure 3. Total concentrations of lithogenic elements ± 1 SD (pmol m^{-3}), with enrichment factors relative to Al overlaid (circles). Unfilled bars indicate that analyte was below the limit of detection and bar represents 75% of the limit of detection. An EF was not determined if both Al and the element of interest were below the limit of detection. Up- / down-ward pointing grey arrows near EF markers indicate that values are minima / maxima because Al / the element were below the limit of detection. *Bars are coloured according to the air mass type of each sample*, blue = RNA, grey = WEU, orange = NAF, green = MED, pink = EEU. The dashed grey vertical lines indicate the legs of the cruise, with Leg 1-3 being left to right.

6. **Section 3.6**: The authors list several factors that influence element solubility. While they appropriately acknowledge that some mechanisms cannot be fully explored due to missing data, I believe that the discussion of acidity effects could be strengthened. Specifically, when discussing the role of acidic species concentrations, the authors should clarify to readers that while these concentrations provide useful insights, they do not directly represent aerosol pH. It would be helpful to explicitly state the limitations of using these parameters as proxies for aerosol acidity.

   We agree that this makes a valuable addition to the discussion here. We have added the following text:

   "*Ultimately, solubility enhancement through acid processing is dependent on the pH environment of the aerosol on an individual particle basis. This environment will vary strongly through the aerosol population, due to differences in internal mixing of acidic and alkaline species and trace elements (e.g. with particle size (Fang et al., 2017; Baker et al., 2020)). Furthermore, changes in the liquid water content of the particles (which is dependent on relative humidity and the hygroscopicity, and hence chemical composition, of the particles in question) can result in dramatic changes in pH, even when acid/alkaline ion balance varies little (Pye et al., 2020; Baker et al., 2021). Information about these factors is not available for the GA04 dataset and the insights provided by the above discussion of ion-solubility relationships are therefore limited.*"

Additional References

Baker, A. R., Kanakidou, M., Nenes, A., Myriokefalitakis, S., Croot, P. L., Duce, R. A., Gao, Y., Ito, A., Jickells, T. D., Mahowald, N. M., Middag, R., Perron, M. M. G., Sarin, M. M., Shelley, R. U., and Turner, D. R.: Changing atmospheric acidity as a modulator of nutrient deposition and ocean biogeochemistry, Science Advances, 7, eabd8800, 10.1126/sciadv.abd8800, 2021.

Carslaw, D. C., and Ropkins, K.: openair — an R package for air quality data analysis, Environmental Modelling & Software, 27-28, 52–61, 2012.

Pye, H. O. T., Nenes, A., Alexander, B., Ault, A. P., Barth, M. C., Clegg, S. L., Collett Jr, J. L., Fahey, K. M., Hennigan, C. J., Herrmann, H., Kanakidou, M., Kelly, J. T., Ku, I. T., McNeill, V. F., Riemer, N., Schaefer, T., Shi, G., Tilgner, A., Walker, J. T., Wang, T., Weber, R., Xing, J., Zaveri, R. A., and Zuend, A.: The acidity of atmospheric particles and clouds, Atmospheric Chemistry and Physics, 20, 4809-4888, 10.5194/acp-2019-889, 2020.