

Review of “Aerosol trace element solubility and deposition fluxes over the polluted, dusty Mediterranean and Black Sea basins” by R. Shelley et al.

An interesting work reporting trace metals, their solubility and dry atmospheric fluxes during GA04 cruise in Mediterranean (Med) and Black seas. The manuscript is well-written and deserves publication after addressing the following points:

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

General comments:

- Since the campaign covers a period during summer, as title I would suggest: Aerosol trace element solubility and deposition fluxes over the Mediterranean and Black Sea basins during summer time (or warm period if they wish).

We do not think that this change is necessary. The season during which the cruise took place is already mentioned in the first sentence of the Abstract.

- There is a good amount of works based on either long-term or annual basis sampling and reported fluxes of nutrients and elements in the Mediterranean and Black Sea which are missing. Please compare with them. Below a non-exhaustive list:

Theodosi C., Markaki Z., Pantazoglou F., Tselepides A., Mihalopoulos N., Chemical composition of downward fluxes in the Cretan Sea (Eastern Mediterranean) and possible link to atmospheric deposition: A 7 year survey, Deep-Sea Research Part II, 164, 89-99, 2019.

Desboeufs, K., Bon Nguyen, E., Chevaillier, S., Triquet, S., and Dulac, F.: Fluxes and sources of nutrient and trace metal atmospheric deposition in the northwestern Mediterranean, Atmos. Chem. Phys., 18, 14477–14492, doi.org/10.5194/acp-18-14477-2018, 2018.

Im U., S. Christodoulaki, K. Violaki, P. Zarbas, M. Kocak, N. Daskalakis, N. Mihalopoulos and M. Kanakidou, Atmospheric deposition of nitrogen and sulfur over Europe with focus on the Mediterranean and the Black Sea, Atmospheric Environment, 81, 660-670, 2013.

Markaki Z., M.D. Loye-Pilot, K. Violaki, L. Benyahya, N. Mihalopoulos, Variability of atmospheric deposition of dissolved nitrogen and phosphorus in the Mediterranean and possible link to the anomalous seawater N/P ratio, Marine Chemistry, Volume 120, Issues 1-4, Pages 187-194, 2010.

Theodosi C., Z. Markaki, A. Tselepidis, N. Mihalopoulos, The significance of atmospheric inputs of soluble and particulate major and trace metals to the eastern Mediterranean seawater, *Marine Chemistry*, Volume 120, Issues 1-4, 20, 154-163, 2010.

Guerzoni, S., Molinaroli, E., Rossini, P., Rampazzo, G., Quarantotto, G., and Cristini, S.: Role of desert aerosol in metal fluxes in the Mediterranean area, *Chemosphere*, 39, 229–246, [https://doi.org/10.1016/S0045-6535\(99\)00105-8](https://doi.org/10.1016/S0045-6535(99)00105-8), 1999.

Thank you. These papers were also mentioned by reviewer 2. We have included relevant comparisons where possible. See our response to Reviewer 2.

For Black Sea, Kocak et al., 2014 and references therein: Atmospheric deposition of macronutrients (dissolved inorganic nitrogen and phosphorous) onto the Black Sea and implications on marine productivity, *Journal of the Atmospheric Sciences* 73 (4), 1727-1739, 2014. Their total deposition measurements performed in Black Sea (Varna, Bulgaria) during similar period (2013-2014) were reported

Since we were not able to collect samples for dissolved inorganic nitrogen and phosphorus analysis during the Black Sea leg of GA04, this paper does not provide any information relevant to our manuscript. Examination of the papers cited by Kocak et al. did not provide many flux determinations comparable to those we report, but we did find total Al flux values in Theodosi et al. (2013), which we were able to compare to the values in Table 2. Our detailed response to this comment is included in our response to Reviewer 2.

Other comments:

- Few words on meteorological conditions are missing. For instance, any rain event occurred during the cruise? Wind speed variability? This information is valuable for the reader.

Unfortunately, we are not able to provide this information.

- Figure 4: How the authors explain the increased soluble levels and the extremely high solubility for terrigenous elements such as Al, Fe and especially Mn (almost up to 100% for the last) under the influence of EEU air masses (pink color). A short comment would be very useful.

We do not have anything to add to the comments that we already made on this (lines 320-324 of the original manuscript).

- Figure 7: Solubility as a function of aerosol load. Similar figure and results were reported by Theodosi et al., 2010 at Finokalia Crete for both wet and dry deposition. Please compare your findings with this work.

We have added a specific reference to this dataset. However, it is not directly comparable to the GA04 data, since it is derived from flux measurements and used a different method to determine the soluble fraction of Fe.

- For solubility: Did the authors calculate ionic balance as a better index of acidity or even better (SO₄+NO₃/NH₄) ratio (in equivalent)? Any relation of (SO₄+NO₃/NH₄) ratio with solubility?

We do not think that ion balance calculations are helpful in the interpretation of this dataset. See response to Reviewer 1.

- N/P ratio in the Mediterranean: Your finding about N/P variability across Mediterranean is in very good agreement with the study of Markaki et al 2010 and I think it is worth mentioning. "Markaki Z., M.D. Loyer-Pilot, K. Violaki, L. Benyahya, N. Mihalopoulos, Variability of atmospheric deposition of dissolved nitrogen and phosphorus in the Mediterranean and possible link to the anomalous seawater N/P ratio, Marine Chemistry, Volume 120, Issues 1-4, Pages 187-194, 2010"

We have added the citation, as suggested.

- For Black sea fluxes compare with the work of Kocak et al., 2014 and references therein.

See above.