

Interactive comment on “Characterisation of aerosol provenance from the fractional solubility of Fe (Al, Ti, Mn, Co, Ni, Cu, Zn, Cd and Pb) in North Atlantic aerosols (GEOTRACES cruises GA01 and GA03) using a two stage leach” by Rachel U. Shelley et al.

Anonymous Referee #1

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This manuscript describes the results obtained from different leaching methods used to liberate soluble trace elements from aerosol samples collected over the North Atlantic Ocean. The majority of the work focuses on leaches with ultra-high purity water (UHP) and an acetic acid solution, although some results obtained from leaching with seawater are also reported. The dataset presented is of high quality and it has been subjected to a very thorough analysis. The manuscript is highly suitable for publication in Biogeosciences, although I feel that some clarifications of relatively minor points are

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necessary.

Although the manuscript focuses on the UHP and acetic acid leach results, Section 3.3.3 presents results obtained by leaching a subset of the aerosol samples using seawater. Currently the manuscript contains no information about how these experiments were performed. Please add this.

Non-seasalt (nss) sulfate concentrations in the aerosol samples were estimated using aerosol chloride concentrations as an indicator of seaspray content. I understand that this was the only indicator available, since major cations (e.g. sodium or magnesium ion concentrations) were not measured. However, chloride is not an inert species in marine aerosol, since it can be converted to hydrogen chloride and lost to the gas phase (Andreae and Crutzen, 1997). The extent of chloride loss is likely to be greater in polluted air types, such as those originating over Europe and North America, but all of the samples will be affected to some extent. Thus the nss concentrations presented here will be over-estimated and it would be helpful to note this in the text.

I found it difficult to compare the results presented in the main body of the manuscript with those in the Supplementary Information (and in the two earlier manuscripts in which previous work on these samples was presented) because different labels have been used for the samples in different places. If it is not possible to only use a single set of labels, please could all the different labels be added to the tables in the supplement?

There is also a specific issue with the naming convention for sample M3-GA03. The "M" here (and the map in Shelley et al., 2015) imply that this was a Marine sample. The discussion on lines 328 -332 specifically state that it had a European air mass back trajectory. Please clarify this.

I assume that positive matrix factorisation analysis (lines 350 onward) was done using total trace element concentrations. Please could this (or the correct information) be specifically stated.

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All of Figures 2 – 7 would be improved by the addition of error bars. This would greatly assist the reader in putting the relatively high variability in calculated parameters (TE ratios to Al, or percentage soluble fractions) for samples with low total concentrations into context with the low variability, high total concentration samples with North African origin.

Minor points:

Line 95: “seawater” spelling.

Lines 103-115. Much of this paragraph is repetition of material from previous paragraphs. It could easily be shortened.

Line 319: The eruption of Eyjafjallajökull took place in 2010, not 2011.

Line 353: I think Fig S2b should be referenced here, not S1b.

Line 369: The panels of Fig. 4 are not labelled on the figure.

Line 374: “the ranges of fraction solubility” - I think there is a misspelling here.

Lines 382-387: This is a very long and cumbersome sentence. Please consider splitting it.

Lines 444-445: “The differences source dependence of” Please correct.

Line 481: Panel h of Fig 6 shows data for cadmium. Lead data is on panel i.

Lines 509-511: This statement is very speculative. Please add further explanation or consider removing.

Line 534: I am a little confused by the opening statement of this sentence. This manuscript has been devoted to the direct measurement of TE solubility!

Lines 536-537: “in regions of high mineral dust deposition and/or productivity fractional solubility”. An odd construction. What is productivity fractional solubility?

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Lines 538-839: How is it possible to have an inverse relationship between TE fractional solubility and aerosol provenance?

BGD

References Andreae, M. O., and Crutzen, P. J.: Atmospheric aerosols: Biogeochemical sources and role in atmospheric chemistry, *Science*, 276, 1052-1058, 1997.

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