

The authors are grateful to anonymous reviewer 1 for providing feedback to the manuscript. Our detailed point-by-point response to the comments is reported below.

Reviewer 1

This study investigates the differences in metal concentrations between TSP (total suspended particulate) and fog samples collected concurrently at a coastal site in 2017. Using ICP-MS analysis and thermodynamic modeling, the authors attribute these variations to distinct complexation behaviors with inorganic/organic ligands. Key findings include enhanced Al/Fe solubility in fog water under neutral pH conditions, hypothesized to result from aqueous-phase processing during droplet activation. XANES analysis confirmed Fe-organic complexes despite thermodynamic predictions favoring hydroxide species. TEM/DLS measurements further supported colloidal Fe nanoparticles as an additional phase. I have the following comments.

Major comments:

1. The manuscript does not explicitly state whether total or water-soluble metals were measured via ICP-MS. Metal solubility should be quantified as the soluble fraction relative to total metal content. Equating bulk concentration to "solubility" is problematic. A revised discussion addressing this distinction is needed.

With ICP-MS we measured the dissolved fraction of the metals, that is from samples extracted in ultrapure water and filtered with a 0.2 μm filter. The methodology for the chemical analysis is reported in the companion paper by Formenti et al. which is now published in EGU sphere (<https://doi.org/10.5194/egusphere-2025-446>). The discussion reported in this manuscript always refers to the dissolved fraction while information on the solubility of the metals (in % over the total) is reported in the companion paper.

2. The method section lacks critical information for ICP-MS analysis. The manuscript only has one sentence for this. E.g. how are the samples prepared? What are the instrumental parameters. This affects how the metal data should be interpreted.

For TSP samples, all information can be found in the companion paper (Formenti et al., this issue).

For fog samples, the following text has now been added at line 147: "For dissolved trace metal determination, fog samples were aliquoted in the field into precleaned (with nitric acid) HDPE bottles and stored at 4°C until analysis. Prior to analysis, samples were filtered through a 0.22 μm filter (PES, Millipore) and acidified using high purity nitric acid (EMD Chemicals, 67–70% Omni Trace Ultra) to a pH less than 2. The samples were analysed for 40 trace metals using a Thermo Electron X-Series 2 quadrupole inductively coupled plasma mass spectrometer (ICP-MS). All elements were analysed using Kinetic Energy Discrimination (KED) mode using as collision cell gas a hydrogen (7%) and helium mixture. Scandium, germanium, yttrium, indium and bismuth were used as internal standards. As for TSP samples, the dissolved fraction is defined operationally as passing through a 0.2 μm filter which may include nano-sized particles as reported in a previous study (Giorio et al., 2025)."

Reference: Giorio, C., Borca, C. N., Zherebker, A., D'Aronco, S., Saidikova, M., Sheikh, H. A., Harrison, R. J., Badocco, D., Soldà, L., Pastore, P., Ammann, M., and Huthwelker, T.: Iron Speciation in Urban Atmospheric Aerosols: Comparison between Thermodynamic Modeling and Direct Measurements. *ACS Earth and Space Chemistry*, 9 (3), 649–661, <https://doi.org/10.1021/acsearthspacechem.4c00359>, 2025.

- Current spectral plots in Figure 5 are challenging to interpret due to overlapping lines/symbols and low contrast colors/lineshapes. Consider stacked panels or splitting into subfigures for key comparisons. For example, it is hard to tell from the figure that Fe was predominantly present in the (III) oxidation state. Additionally, descriptions on how the simulations were done (i.e. parameters) should be added to the method section.

We have now improved Figure 5 following the suggestion for the reviewer.

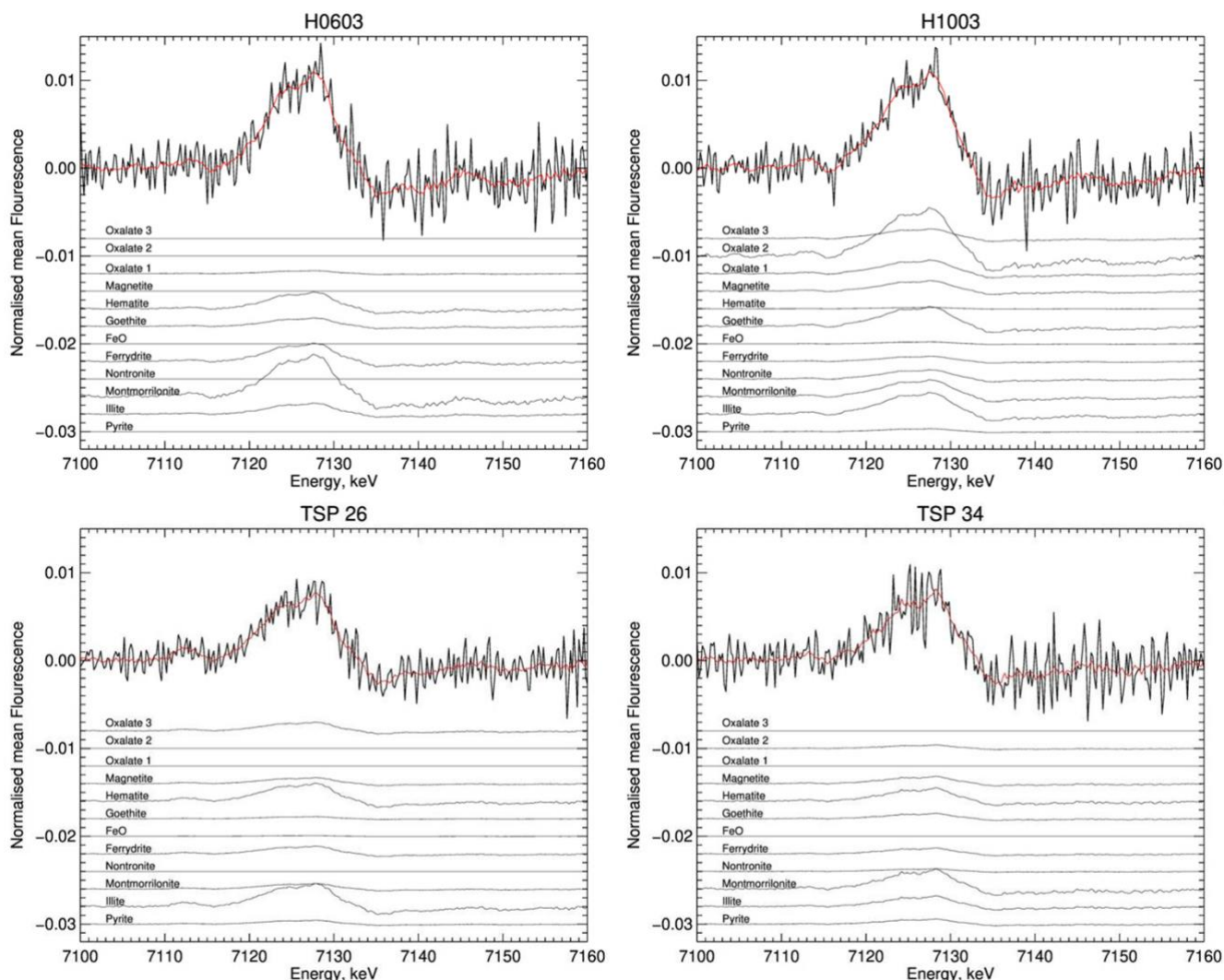


Figure 1. XANES spectra at the Fe K-edge for two fog samples (top, H0603 and H1003) and two TSP samples (bottom, TSP 26 and TSP 34) showing the results of the spectral deconvolution using a linear combination fitting with Fe measured reference standards of oxides, clays, organic complexes and sulphide. The individual contributions of the references to the total fit (red curve) are represented as stacked black lines. For sake of clarity, the contribution of organic complexes (named “oxalate 1” for $\text{Fe}(\text{C}_2\text{O}_4)_2\text{H}_2\text{O}$, “oxalate 2” for $(\text{NH}_4)_3[\text{Fe}(\text{C}_2\text{O}_4)_3]$ and “oxalate 3” for $\text{Fe}_2(\text{C}_2\text{O}_4)_3\cdot 6\text{H}_2\text{O}$ in the Figure) are multiplied by 10.

The description of the fitting procedure is reported in the companion paper from Formenti et al., this issue. “for Fe(II)-bearing minerals, the position of the centroid of the pre-edge is found at 7112.1 eV, whereas it is at 7113.5 eV for Fe(III)-bearing minerals.” and “The speciation of Fe was obtained by the least-square fit of the measured XANES spectra based on the linear combination of mineralogical references. Fits were conducted on the first derivative of the normalized spectral absorbance in the energy region between 7100 to 7180 eV, corresponding to -30 and +50 eV of the K-edge. Only the fits with a χ^2 closest to 1 were retained for further

analysis.” Information on the standard reference compounds is reported in the supplements of Formenti et al., in Table S2. The companion paper is now available as pre-print in EGU sphere (<https://egusphere.copernicus.org/preprints/2025/egusphere-2025-446/>).

4. The discussion on pH and metal solubility is largely lacking. It is known that acidification of metal species in the presence of sulfate is an important mechanism to make insoluble metal become soluble. The author should consider adding relevant discussions. It would be useful to plot the pH against/with metal solubility.

Metal solubility in TSP is discussed in the companion paper by Formenti, et al. We have now explained this at the beginning of section 4.3: “Solubility of metals in TSP is discussed for the whole campaign in the companion paper by Formenti et al., 2025, this issue. Here we focus the discussion on the comparison between dissolved metals in pairs of TSP and fog samples.”

Concerning the extraction protocols, while fog pH was measured and was always close to neutral, for TSP dissolved metal determination autogenic pH was used which may be more acidic (not measured). Despite this difference, that would cause a higher dissolved fraction for TSP compared to fog samples, we actually observed the opposite for most metals/samples and therefore it should not affect our conclusions. This difference has now been clarified in the methodology section at line 154: “One difference between the dissolved fraction in fog and in TSP may arise from the pH of the extraction solution, which was in both cases autogenic but may be more slightly more acidic for TSP (not measured) compared to fog samples (measured to be close to neutral).”

Minor comments:

1. Provide the year for this reference: “Formenti et al, this issue”. This appears many times in the manuscript.

The reference has been updated and added to the reference list.

2. “...is discussed in section 0.” Where is section 0? This also appears many times in the manuscript.

It looks like this is due to an issue in converting the word file to PDF. We have fixed this in the revised manuscript.

3. Revise single-sentence paragraphs at e.g. Lines 91/125/141 by integrating them logically into adjacent sections or expanding context where appropriate.

The sentences have been revised as follows:

Line 91: “The list of aerosol samples and concomitant fog samples relevant to this paper is presented in **Error! Reference source not found.**, while in the following paragraphs, we describe the strategy of fog and seawater collection and analysis.”

Line 125, describing the liquid water content measurements with the CASC has been moved to the previous section, where CASC collection is described.

Line 141 (147 in the revised version) has now been modified as reported in the answer to comment 2.

4. In section 4.1, “temperature with a narrow range (<5°C) and humidity (RH 95%)”. Provide a range for humidity.

We have now added the requested information: “RH 95 ± 4 %, min 78.5 %, max 100 %”.

5. Section 4.3, line 282, “higher concentrations of Al, Fen and Ni..” Missing Cu here?

We have now added Cu.

6. Define LWC upon first use.

Definition added.