

Atmos. Chem. Phys. Discuss., referee comment RC2
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Comment on acp-2021-712

Anonymous Referee #1

Referee comment on "Sources and processes of iron aerosols in a megacity in Eastern China" by Yanhong Zhu et al., Atmos. Chem. Phys. Discuss.,
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General Comments:

This paper quantifies the total Fe and dissolved Fe in the fine particles collected in Hangzhou megacity of Eastern China. The pollution levels, Fe content and solubility under different weather conditions are compared. The source contributions of dissolved and total Fe are apportioned by PMF analysis. Finally, the impact of atmospheric acidification processes on Fe solubility are discussed. This paper appears to be a data paper with litter data analysis of the beyond reporting the results. Although the authors try to explain the sources or atmospheric phenomena that lead to the changes of iron aerosols under different weather conditions, the discussion are too general. Additionally, the English language requires substantial improvement (both style and grammar) throughout the manuscript. Many sentences are not clearly written. The topic is certainly appropriate for Atmospheric Chemistry and Physics. However, there are quite a few major issues with the study that prevent me from recommending it for publication in the present format. It is possible that these issues could be addressed with a major revision. My specific concerns are addressed below.

- Line 43: Change "having" to "have".
- Line 49: Fe solubility in some anthropogenic sources such as coal fly is very low, so here the statement "an increase ... from anthropogenic source cloud lead to the increase in Fe solubility" is not accurate.
- Line 55: When the relative humidity is higher than 50%, some soluble inorganic components may begin to be hygroscopic, resulting in phase changes of particles, but some secondary organic components will not. For some aged aerosol particles after liquid-liquid phase separation, the organic coating also prevents inorganic components from contacting the atmosphere directly, which would affect the hygroscopic property of particles. Moreover, in the reference cited here, hygroscopic growth begins at 60%

and 55% for haze particles. Therefore, the "50%" or "secondary aerosol particles" here are not appropriate, please consider rewording them. Additionally, what do the surfaces of secondary aerosol particles mean? You mean the secondary materials coat the primary particles, or the particles are secondarily formed? Please clarify.

- Lines 62-64: It is not a complete sentence.
- Line 84: The identification of fog samples: as described in Table S2, the threshold value of 10 km is too high for fog visibility. If the relative humidity during fog is not considered in the definition of weather conditions, in my opinion, it will lead to the misclassification of the fog samples. The authors do not mention the RH in the definition of fog. Please clarify.
- Line 89: The daytime and nighttime samples are collected respectively. It is not quite clear how the sampling strategy was selected, and why the authors do not discuss the differences between daytime and nighttime samples.
- Lines 88-103: The PM₅ samples are used for the mass concentration analysis of total and dissolved Fe, but the particle size of the samples used for single particle analysis is not clearly indicated in this paper. Is there any difference in the cut point inlet between single-stage cascade impactor and TH-16A Intelligent sampler? The authors state that the collection efficiency is 50% for particles with an aerodynamic diameter of 0.1 μm and a density of 2 g cm⁻³, so the aerodynamic particle size of particles collected from single-stage cascade impactor is less than 100 nm? If so, I suspect that the collected particles have not yet grown.
- Line 155: A reference would be helpful.
- Line 226: Change "3.3.2" to "3.3.1".
- Lines 227-258: The authors applied Pearson correlation analysis between dissolved Fe and other elements to explore the primary of dissolved Fe. The elements do not consider the impact of atmospheric process, but the dissolved Fe is affected by the atmospheric process, so I am wondering whether the correlation analysis between the dissolved Fe and other elements can be used for source identification.
- Lines 241-243, 246-248, 252-253 and 256-258: The authors state that EF values of Ca and Ti are less than 10, suggesting a potential contribution of coal combustion,...; Pb, Zn and K had EF > 10, indicating a potential contribution of coal combustion, ... These statements make no sense.
- Lines 260-265: I believe that the figures represented in supplement appear to be more important. While reading that authors have referred to supplementary figures too many times, I would suggest merging or re-plotting some of the figures to bring supplementary figures in main text.
- Lines 267-273: The Figure 3 has provided the contribution of each source in detail, so the authors do not need to describe them again. The similar situations appear many times in the text.
- Lines 2677-283: Oakes et al. (2012) found that Fe solubility was 0.06% in coal fly, 46% in biomass burning, 51% in diesel exhaust and 75% in gasoline exhaust as the authors state in Lines 47-48. Here the industrial emission is the largest contributor to dissolved Fe in haze, fog, dust and clear days. Traffic emission is the secondary contributor. It warrants further discussion.
- Lines 302-306: As shown in Figure 5, the size of S-Fe particles in haze samples is larger than that of fog samples. The relative humidity during fog is higher than haze. Thus particles during fog are more wet, and softer, so the projected area after impacting copper is generally larger than those during haze. How are the data interpreted?
- As can be seen in Figure 6, the relative humidity of several fog samples is ~75%. So, the average relative humidity during the sampling period is less than 80% and visibility is less than 10 km. According to the definition in Table S2, these samples should be haze samples.
- 3.2: The authors use the data of TEM/EDS to interpret the secondary source, and regard S-Fe as an indicator of acid dissolution. The number contribution of S-Fe particles to Fe-containing particles are 76.3% and 87.1% in haze and fog days. It is not

surprising. How about the clear and rain days? I speculate that the proportion of S-Fe particles have a similar contribution.

- Line 312: Under fog condition, RH ranges from 71% to 99%. Again, what is the difference in RH between fog and haze samples?
- Line 320: The authors use SO_4^{2-} and NO_3^- to indicate aerosol acidification. Why does NH_4^+ not be considered in aerosol acidification?
- Line 327: Here the unit of $(2\text{SO}_4^{2-} + \text{NO}_3^-)$ is mmol? What is the factor of 2 for?
- Lines 327-330: The water content of the particles seem to be important. Is it possible to calculate the water content of the particles under different weather conditions, then do the correlation analyses between the dissolved Fe and water content?