

Reviewer #1 Comments:

This manuscript by Ueda et al., investigated the morphological features, mixing states and water solubility of Fe-containing particles in aged fine aerosol particles over the Indian Ocean. The topic of this study is attractive and interesting, which is very useful for understanding the ageing of particles and the dissolution of Fe-containing particles during the transport of aerosols and the morphology variations of Fe-containing particles. Overall, the manuscript is logical, and the main issues are very well discussed in this paper. I would therefore recommend this manuscript for publication after the authors have addressed the following comments.

Response:

We appreciate the many constructive comments offered by Prof. Weijun Li, which have improved our manuscript considerably. Revisions have been highlighted as red in the text of the revised manuscript. This manuscript was checked according to the journal guidelines by a native-English speaking professional with experience in the review of technical documents in this field.

Comments:

Major concerns:

- 1. The units of horizontal variation of mass concentrations of (a) nss-SO₄²⁻, (b) NH₄⁺, and (c) Fe in PM_{2.5} in Figure 2 are $\mu\text{g}/\text{kg}$ and ng/kg , however, there are $\mu\text{g}/\text{m}^3$ and ng/m^3 in Table 2, which is confused for me, please explain the differences between the two units and make them uniform.*

Response:

Those mass concentrations were measured in units of $\mu\text{g}/\text{m}^3$ and ng/m^3 . For Figure 2, the Fe mass concentrations are shown as a mixing ratio ($\mu\text{g}/\text{kg}$ and ng/kg), which facilitates readers' comparison to model output data presented in Figure 8. Mixing ratios were calculated using the daily average of temperature and atmospheric pressure measured onboard. We added the explanation presented above to section 3.1. The units in the revised Tables 2 and 3 were changed to present units of $\mu\text{g}/\text{kg}$ and ng/kg uniformly.

Comments:

- 2. Figure 2c (mass concentrations of Fe) has two more data points compared to figures 2a and 2b. Why?*

Response:

As explained in section 2.2, we measured ions (such as (a) nss-SO_4^{2-} and (b) NH_4^+) using samples collected on a quartz fiber filter, and metals (such as Fe (c)) using samples collected on Teflon filter. Because stocks of quartz fiber filters were finished up, the sampling was stopped before that using a Teflon filter. We added some mention of each sampling period to section 2.2 of the revised manuscript.

Comments:

3. Line 232: *“For non-sea-salt components, the relations between the doubled nss-SO₄²⁻ molar concentration and the NH₄⁺ plus nss-K⁺ molar concentration were usually between 1:1 and 2:1 (Fig. S1b), suggesting that nss-SO₄²⁻ originated from ammonium sulfate, ammonium bisulfate, and ammonium potassium rather than from sulfuric acid.” Here, “ammonium potassium” should be “potassium sulfate” ?*

Response:

Thank you for noticing that mistake. That was corrected to potassium sulfate.

Comments:

4. Line 227-228 and line 232-233: *“The values nss-K⁺, which are regarded as originating mainly from biomass burning.....; For non-sea-salt components, the relations between the doubled nss-SO₄²⁻ molar concentration and the NH₄⁺ plus nss-K⁺ molar concentration were usually between 1:1 and 2:1”. The author argued that nss-K⁺ was mainly from biomass burning, please provide more evidence to support this view.*

Response:

As already described, nss-K^+ has often been used as a tracer of biomass burning (Andreae, 1983; Kawamura and Kaplan, 1987; Narukawa et al., 1999). In addition, highly frequent agricultural burning around the windward area of our observation site in autumn was reported from some studies (Bray et al., 2019; Shaik et al., 2019). Therefore, we thought that the air mass of the north site in this study might have been affected by biomass burning around the windward continental area. However, we thought that the other possibilities should not be denied completely. We carefully revised the description with references as shown in the first paragraph in section 3.1.

Comments:

5. In the section: *“3.2 Individual particle features and co-existing states with Fe of sulfate and soot”,*

the author chooses samples #01 and #07 as example to illustrate the individual particle features, however, I fail to get the idea why the author selects these two samples. Is there any special in these two samples, the reasons should be given.

Response:

Because morphological features of samples #01-06 were similar, we used sample #01 as a representative example, and sample #07 as the other examples. Figures for photographs and morphological types of all samples are presented in supplemental materials (Figs.S3 and S4 before revision). In the revised manuscript, to show similarity of samples #01-06, the graph for morphological types was moved to Figure 3, with some revision of the first sentences in section 3.2.

Comments:

6. *Compared with the filed observation results, the CAM5-ATRAS model underestimates Fe by nearly 1/3 (from Figure 9a), why? Are there other unknown sources of Fe or is there a large uncertainty in the Fe emission inventory used by your model?*

Response:

As you have commented, a great amount of uncertainty exists in Fe emission inventories from all sources including anthropogenic materials, dust, and biomass burning origin. While greater underestimation of Fe exists in earlier model estimation, our model has improved Fe simulations with detailed representations of anthropogenic Fe (Matsui et al., 2018b) and new emission data of anthropogenic Fe by Rathod et al. (2020). Although with underestimation of nearly 1/3, our simulations have shown higher Fe concentrations and better agreement with observations than earlier estimates (Liu et al., 2022). The comparison to TEM results suggests that our model underestimates the fraction of anthropogenic aluminosilicate Fe, such as illite and kaolinite. Therefore, their emissions might be underestimated, leading to underestimation of total Fe mass concentrations. Additionally, different spatial and temporal scales between observations and models might also explain the model-observation differences in this study. These explanations were added to section 3.4.

Comments:

Some other minor issues:

1. *Line 132: Cl⁻, NO₃⁻, SO₄²⁻, Na⁺, NH⁺....., here, “Cl⁻ andNH⁺” should be revised to “Cl⁻ andNH₄⁺”. Please check the similar issues in whole text.*
2. *The numbers and letters are so small that they can't be read clearly in Figure 4 and Figure 7,*

please adjust the font size.

Response:

Thank you for pointing out the matters listed above. We rechecked and revised the manuscript.

Reviewer #3 Comment

The authors investigate aerosol particles' iron (Fe) properties over the Indian Ocean aboard a research vessel. The article presents important information on these particles' morphology, concentration, and degree of solubility. In general terms, the article is well-written. However, some minor observations need to be addressed. The article can be accepted after answering the following questions.

Response:

We appreciate the many constructive comments offered by anonymous reviewer #3, which have improved our manuscript considerably. Revisions have been highlighted as red in the text of the revised manuscript. This manuscript was checked according to the journal guidelines by a native-English speaking professional with experience in the review of technical documents in this field.

Comments:

Abstract

line 15. the authors mention that they analyzed particles in the size range 0.3-0.8, using a cascade impactor. As written, it appears that they studied a continuous range of particle sizes. However, the methodology mentions that the sampler only has three nominal sizes (1.6, 0.8, and 0.3 μm). Please mention these three sizes in the abstract.

Response:

TEM samples were collected using a three-stage cascade impactor, but only those of the third stage were used for this study. Particles larger than the 50% cut-off diameter of the stage are collected to efficiency of higher than 50% on the stage. Therefore, in the third stage of the impactor, particles from about the third stage of cut-off diameter (aerodynamic diameter) to about the second stage cut-off diameter were collected. For this study, we specifically examined submicrometer particles that can include many anthropogenic aerosols, and analyzed only the stage samples that mainly submicrometer particles collected. Therefore, we described only the size range of the stage used for analysis as necessary information in the *Abstract*. Although use of only a third stage was also explained before revision, it might have been difficult to understand that point in the *Abstract*. Therefore, we revised the text in the *Abstract*.

Comments:

Methodology

The water dialysis process was important to study the water's mixing state and elucidate the Fe's solubility. Could you expand the description of this technique and mention the instrument used for it? Indicate the bibliographic source from which equations 1 and 2 were obtained.

Response:

Water dialysis method with image analysis had been used for quantification of the volume of water-soluble materials by Okada (1983). This method needs no other special instruments of already written EDS and TEM, although a petri dish and simple dropper are used. For the method described by Okada (1983), they estimated the particle volume before and after water dialysis using image analyses. For our present studies, we tried and applied water dialysis to quantify changes of Fe before and after water dialysis using EDS analysis. Therefore, the experimental method can be cited from earlier studies, but the quantitative method of Fe by the equations is our original. We also used the same method in Ueda et al. (2022), but did not mention the equations. For this study, demonstration of these equations was supported in supplemental materials before revision. In the revised version, we moved the demonstration in the main text (sections 2.3.1 and 2.3.2) and reinforced the description of this technique.

Comments:

Please revise the wording in the first line of the first paragraph of section 2.1. It is not easy to understand the name of the cruise ship where the study was conducted.

Response:

The sentence was revised as presented below.

Before revision>> Atmospheric observations were conducted over the Indian Ocean during the R/V Hakuho Maru during KH-18-6, which took place on 6–28 November 2018.

After revision>> Atmospheric observations were conducted over the Indian Ocean during the R/V *Hakuho Maru KH-18-6* cruise, which took place on 6–28 November 2018.

Line 200: Check the spelling in this sentence.

Response:

The spelling is no problem because the spelling represents the origin of the model's name. We have used the same form in reports of earlier studies (e.g. Matsui, 2018; Liu et al., 2022).

It is recommended to use the same concentration units in tables and figures. Tables 2 and 3 report the concentrations in $\mu\text{m}/\text{m}^3$ and ng/m^3 , respectively. While in Figure 2, the units are reported as $\mu\text{g}/\text{kg}$.

Response:

Mass concentrations of ions and metals were measured in units of $\mu\text{g}/\text{m}^3$ and ng/m^3 . For Figure 2, the Fe mass concentration is shown as a mixing ratio ($\mu\text{g}/\text{kg}$ and ng/kg) for readers to compare them to model output data of Figure 8. Mixing ratios were calculated using the daily average of temperature and atmospheric pressure measured onboard. We added the explanation given above to section 3.1. The units in the revised Tables 2 and 3 were presented uniformly as $\mu\text{g}/\text{kg}$ and ng/kg .

Comments:

Results

It is suggested that labels (a), (b), (c), and (d) have to be added to each panel in Figure 8. The figure caption should also be improved to make it easier to read.

Response:

We added the related labels and revised the captions.

Comments:

Figure 10 has many elements. It is suggested to separate them and present some results individually. In addition, the figure caption is very long and difficult to read.

Response:

We separated the figure to Figures 10 and 11. The caption was also revised.