

*Editor Comments:*

*I thank the authors for addressing most of the comments listed by both reviewers; however, the manuscript requires additional work before it can be accepted for publication in ACP.*

Response:

We appreciate your many valuable comments and criticisms. We have undertaken efforts to improve our manuscript as we can.

*Major/minor comments:*

*1. The authors argue that “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”; however, I found many grammatical errors along the document. I invite the authors to significantly improve the English of the entire manuscript.*

Response:

Thank you for the comments. We checked the entire manuscript and will continue our efforts. However, it is unfortunately impossible for a non-native speaker of the language to find all errors. Because we understand that well, we have sought the help of reliable native-English speaking professionals after doing our best. The proofreading has been done by multiple native English speakers who have experience at proofreading many papers that have been accepted to international academic journals, including *ACP*, before and since *ACP* was founded in 2001. The present manuscript was also rechecked after sharing this editor comment. Specifically, all of the language requirements for this journal were reviewed and checked. American English spelling and grammar were used, which might be objectionable to speakers from other countries, but which is specifically allowed in the journal guidelines. It would help if you could point to exact grammatical points that you regard as errors. Perhaps specific points can be discussed and resolved. We are committed to improving the manuscript to your high standard. More than anything, we believe that this journal will be fair to the science.

*2. Along the document there are too many qualitative words such as “Presumably”, “it might be”, and “might have”, among others. I invite the authors to avoid such statements and to be more quantitative.*

Response:

We agree that such statements might detract from the quantitative strengths of our presentation. In fact, we have used such words to distinguish supposition and speculation from mathematically probable and certain findings. Therefore, we checked the use of such words (in L334, L339, L343, L467, L529, L534, L593 before revision) critically throughout our manuscript. Although some of the expressions remain because of their context, we tried in most cases to replace or clarify those statements by revision.

*3. I invite the authors to reduce the large number of self citations.*

Response:

We deleted some self-citations and related sentences from the text with this revision (L82, L156–159, L189 and L338–341 before revision).

*4. L247-248: I think this is rather speculative and it is not supported by the present results.*

Response:

We had regarded potassium as an important tracer over the remote ocean based on earlier studies to which we referred before revision. However, we have reconsidered after reading reviewer and editor comments, although they mentioned no clear reason for denying that point. This interpretation seems weak in current studies. Therefore, sentences related to the implication of a relation between potassium and biomass burning were deleted with this revision. This revision does not have important effects on our main conclusions related to Fe-containing particles.

*5. Make units consistent between Fig 2 and Table 2 and 3.*

Response:

The units  $\mu\text{g/g}$  for Figures 1a and b, and Table 2 were changed to  $\text{ng/ng}$ .

*6. Figure 9. Add log scale in panel (a).*

Response:

Because Figure 9a specifically emphasizes the difference between the Northern and Southern

hemispheres, we do not think it is necessary to use a logarithmic axis. The logarithmic axis obscures the difference between the model and the observations for the Northern Hemisphere. Furthermore, we do not think that strict comparison of the difference has strong meaning for the quite low concentrations prevailing in the Southern Hemisphere. Therefore, we did not change Figure 9a. Instead, the average values of Fe concentrations were added to the text (L686).

*Technical comments:*

*L24-25: “aluminosilicate Fe contained in matter such”. Please re-write it.*

Response:

The sentence was changed to “anthropogenic aluminosilicate (illite and kaolinite) Fe contained in matter such.” in the revised manuscript.

*L26: Do the authors mean “Fe-containing particles”?*

Response:

In this case, we mean the Fe-containing part rather than Fe-containing particles including sulfate coating. Sentences of L26 were revised as presented below.

Before revision>> “Our observations suggest that Fe in particles over remote ocean areas has multiple shapes and minerals, and further suggest that its solubility after aging processes differs depending on their morphological and mineral type.”

After revision>> “Our observations revealed multiple shapes and compositions of Fe minerals in particles over remote ocean areas, and further suggested that their solubilities after aging processes differ depending on their morphological and mineral types.”

*L37: Add a reference after “areas”*

*L40: Add a reference after “areas”*

Response:

References were added.

*L41: What do the authors mean with “evaporation and condensation processes”*

Response:

The sentence was revised as presented below.

Before revision>> “By contrast, combustion Fe is emitted as both fine and coarse particles, through evaporation and condensation processes”

After revision>> “By contrast, combustion Fe is emitted as both fine and coarse particles through evaporation of metals at higher temperatures in thermal sources and through condensation processes that occur with diffusion and cooling”

*L58: Add a reference after “sources”*

Response:

References were added.

*L59: Add a reference after “Fe”*

*L60: This was mentioned 2 lines above*

Response:

We deleted the sentence of L59 because we thought that the sentence had no strong meaning for this study. Regarding L60, the sentence two lines above mentions Fe at the emission sources. However, the sentence of L60 mentions Fe change with atmospheric aging. Therefore, we did not change the sentence. However, we think that deletion of the sentence of L59 between the other two sentences made it easier to understand the difference (L63–L66 after revision).

*L63: Add a reference after “simulations”*

Response:

This sentence is related to the sentence immediately prior and references. Instead of presenting an added cited reference, the sentence was revised as shown below.

Before revision>> “...; Sakata et al., 2022). Earlier knowledge about relations between solubility, Fe mineral species, and aging processes has usually been based on bulk sample measurements, laboratory experiments, and simulations.”

After revision>> “...; Sakata et al., 2022). This earlier knowledge about relations between solubility, Fe mineral species, and aging processes has usually been based on bulk sample measurements,

laboratory experiment findings, and simulation results.”

*L65: Add a reference after “insufficient”*

Response:

The sentence was deleted because the meaning overlapped with the last sentences in the section.

*L73: Add a reference after “areas”*

Response:

Cited references were added. The sentence was also changed in minor ways according to the cited reference.

*L113-114: Why is it mentioned “6–16 November 2018” twice. This is unclear.*

Response:

We are sorry. The second period is 17–19 November 2018. That was corrected in the revised manuscript.

*L128: (a.s.l. 14 m).?*

Response:

We corrected the expression to "located at about 14 m altitude from sea level" in the revised manuscript.

*L132: “the home laboratory”?*

Response:

The words were deleted.

*L143: "Aerosol particles were collected for morphological particle analysis using TEM" What does it mean?*

Response:

We corrected the sentence and the subsequent sentence.

Before revision>> "Aerosol particles were collected for morphological particle analysis using TEM. Aerosols after diffusion drying were collected on carbon-coated nitrocellulose (collodion) films using cascade impactors."

After revision>> "After diffusion drying, aerosols were collected on carbon-coated nitrocellulose (collodion) films using cascade impactors. Then the morphologies and compositions of particles were analyzed using TEM."

*L144: "cascade impactors". Please add model and manufacturer.*

Response:

The cascade impactor used for this study was designed by the authors. Design information such as the materials and nozzle diameters was added to the revised manuscript. The 50% cut-off diameters were calculated using equation 5.28 in Hinds and Zhu (2022, Third edition Aerosol Technology, Wiley, pp109). The relevant reference was also added to the revised text. With this revision, we found correction of the Stokes number from the first edition of Hinds (1982), from 0.22 to 0.24. Therefore, the calculated result of the 50% cut-off diameter of second stage corrected from 0.8  $\mu\text{m}$  to 0.9  $\mu\text{m}$ .

*L146: "flow rate of 1.0 L min<sup>-1</sup>." It this correct? This looks pretty low to me.*

Response:

It is correct. Such a flow rate is well used in the TEM sampler, which has a single nozzle (e.g. Kojima et al., 2006; Neimi et al., 2006; Li et al., 2013; Adachi et al., 2020). The cutoff diameter of the impactor depends mainly on the flow velocity through a jet nozzle to the impaction plate. As an impactor of the higher flow rate, there is an impactor having multiple orifices, such as the MOUDI series. Such a multiple orifice impactor is designed to collect bulk particles to constitute a detectable amount for chemical analysis rather than for TEM analysis. Such a sampler also seems to be capable of use as a TEM sampler with some arrange holder. However, it does not require the accumulation of numerous particles for individual particle observation because dispersed particles on film are better

for observation. In addition, the TEM grid is small (5 mm diameter). Therefore, we think that a single-nozzle sampler with low flow has more generally been used for aerosol sampling for TEM analysis.

#### References

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*L224: Is “water” an aerosol species?*

Response:

Liquid water in the atmosphere is fundamentally present as water in hygroscopic aerosols or clouds and rain droplets. For example, for chemical analyses of bulk aerosol samples, we also agree that water and the other species should have separate explanations. However, in this case, the sentence explains the aerosol composition of atmospheric suspended particles in our model. Therefore, we regard water as an aerosol species.

*L243: “2018, when rain events occurred”. Where is this shown?*

Response:

Information about rain observation was added to Figure 1a and section 2.1. In addition, the sentence

was revised as presented below.

Before revision>> “North of the equator, nss-SO<sub>4</sub><sup>2-</sup> and NH<sub>4</sub><sup>+</sup> concentrations were especially high. These mass fractions were, respectively, 70–76% and 18–22%, except for data of 11 November 2018, when rain events occurred.”

After revision>> “These mass fractions were, respectively, 70–76% and 18–22%, except for data of 11 November 2018, when nss-SO<sub>4</sub><sup>2-</sup> and NH<sub>4</sub><sup>+</sup> were less around rain event occurrences.”

L243-244: “”*The values of nss-K<sup>+</sup>, also tended to be north of the equator. What does it mean?*”

Response:

The sentence has been corrected to “The values of nss-K<sup>+</sup> also tended to be higher north of the equator”.

L314-315: “*that they have hygroscopicity*”. *What does it mean?*

Response:

The sentence has been corrected to “that they are water-soluble materials”.

L316-317: “*are probably formed with secondary formation*”. *What does it mean?*

Response:

The sentence and the preceding sentence were corrected as presented below.

Before revision>> “Such particles are probably formed with secondary formation or coagulation of sulfate with an Fe-containing particle.”

After revision>> “Such sulfate on Fe-containing particles can be formed by condensation from sulfuric gaseous materials or coagulation of sulfate particles.”

L319: *I think “under their deliquescence humidity” can be deleted.*

(Probably L329)

Response:

We had intended to mention state in a metastable humidity (i.e. from deliquescence humidity to



efflorescence humidity). Although the particle state can be determined as liquid in higher-than-deliqescence humidity and solid in efflorescence humidity, the state in a metastable humidity can be solid or liquid depending on the experience of humidity. However, the state of ammonium sulfate particles in the atmosphere can be inferred from the particle morphology on the sample and in situ humidity because the shape of solid ammonium sulfate changes under the metastable humidity, as explained in text. Therefore, the sentence is corrected as presented below.

Before revision>>“Particles composed mainly of ammonium sulfate can be present as solid or liquid under their deliquescence humidity, according to atmospheric humidity experience.”

After revision>>“Particles composed mainly of ammonium sulfate can be present as solid or liquid in a metastable humidity condition between efflorescence and deliquescence humidities, according to their atmospheric humidity experiences.”

*L362: “the Fe-containing particles mixed with both Ca and Mg were only one”. What does it mean?*

Response:

The sentence was corrected as presented below.

After revision>> “, in our study, an Fe-containing particle in which both Ca and Mg were detected was only one...”

*L 379: Replace “Sects” with “Sections”*

*L492: “Fe/Pd” should be in italics.*

Response:

Those points were corrected in the revised manuscript according to the comments.

*Notification to the authors from Editor:*

*Please ensure that the colour schemes used in your maps and charts allow readers with colour vision deficiencies to correctly interpret your findings. Please check your figures using the Coblis – Color Blindness Simulator (<https://www.color-blindness.com/coblis-color-blindness-simulator/>) and revise the colour schemes accordingly.*

Response:

We checked all figures using the simulator. Because some colors in Figures 1, 6, 9 and 10 were too light for some color vision deficiency categories, we changed them. Mapping photographs (Figures 4 and 7) and Maps (Figure 2 and Figure 8) were also checked; all patterns of the simulator were checked. The colors were looked different, but same colors were not assigned to different values. The color contrast seemed sufficient to recognize important differences of values for this study in all cases. Therefore, we judged that our map colors can accommodate color vision deficiencies and can enable readers to interpret our findings correctly.