16 October 2025

Dear Professor Guangjie Zheng,

Thank you very much for handling our manuscript submitted to ACP for the consideration of publication (manuscript number: egusphere-2025-2235; title: Atmospheric chemical processing dictates aerosol aluminum solubility: insights from field measurement at two locations in northern China).

The second version of our manuscript has been reviewed by two referees again. Ref #1 only has a few very minor comments, while ref #2 still has some major concerns. We have carefully addressed these comments and revised our manuscript accordingly. We believe that the revised manuscript can be accepted for publication, and highly appreciate these comments which have helped us further improve our work.

We would like to take this opportunity to thank you and the two referees for all the inputs. Please feel free to contact us if you need further information.

Dr. Mingjin Tang, Professor
Guangzhou Institute of Geochemistry
Chinese Academy of Sciences
Guangzhou 510640, China

Comments by referees are in blue.
Our replies are in black.
Changes to the manuscript are highlighted in red both here and in the revised manuscript.

Reply to referee #1

This manuscript presents a well-designed and thorough investigation of the spatial and seasonal variations in aerosol aluminum (Al) solubility, focusing on atmospheric aging by comparing solubilities at Xi'an and Qingdao. The authors integrated extensive field measurements, performed statistical analyses, and illustrated the effects of aging process in modulating Al solubility.

Reply: We would like to thank referee #1 for reviewing our manuscript again and recommending it for publication after minor revision. We have addressed his/her comments and revised the manuscript accordingly, as detailed below.

The manuscript would benefit from explaining the relatively large overlapping Al solubility data. The viewpoints are scientifically solid, however, the gap between the statistical analyses and conclusions requires more explanation. For example, in Figures 6 and S1 excluded outliers for Xi'an, but a very high solubility datapoint is kept for Qingdao instead and potentially makes the regression model significant.

Reply: Indeed Al solubility reported in our work show relatively large overlapping. This is unfortunately unavoidable for most (if not all) of field measurements, since environmental conditions are very complicated in the real atmosphere.

For Figures 6 and S1 in the previous versions of our manuscript, we excluded outliers at Xi'an but did not exclude outliers at Qingdao. We also carried out statistical analysis after excluding the outliers at Qingdao, and this almost led to no change. In the revised manuscript (page 21) we have made the following change to the caption of Figure 6: "…(d) supermicron particles at Qingdao (the *r* value changed from 0.81 to 0.74 if the data point with the highest Al solubility was excluded)." Similar change was also made to the caption of Figure S1 in the revised supplement (page 10): "…(d) supermicron particles at Qingdao (the *r* value changed from 0.81 to 0.77 if the data point with the highest Al solubility was excluded)."

Below are technical corrections to be noticed:

1. Line 36, in the abstract, there is a new line that's not supposed to be there.

Reply: We would like to thank referee #1 for pointing out this error, which we should have avoided. In the second round of review, referee #2 insisted that the abstract was overly general. As a result, we have substantially modified the abstract in the revised manuscript (page 2); in addition, we have carefully checked the entire manuscript and supplement to avoid errors which we should avoid.

2. Legend in Figure 7 should have a box like other figures in the manuscript.

Reply: As suggested, we have updated Figures 7 (page 22) and 8 (page 23) in the revised manuscript.

Reply to referee #2

Thank you for the revisions and your response to my comments. However, I note that the authors have not made substantial modifications to the manuscript, nor have they adequately addressed my concerns regarding the scientific robustness of the paper's conclusions. Measuring the depth of analysis by page number is inappropriate; a paper should delve into scientific exploration deeply, even if it is concise. The core conclusion proposed by the authors—that atmospheric chemical processing alters the solubility of aluminum in aerosols—while not necessarily incorrect, lacks convincing support from the current explanations. The authors fail to rigorously demonstrate that this is a dominant factor influencing aerosol aluminum solubility.

Reply: We would like to thank referee #2 for reviewing our manuscript again. We tried to address the comments he/she raised in the first round of review. Since referee #2 still has some major concerns, we have carefully addressed these remaining concerns and revised the manuscript again, as detailed below. We highly appreciate these comments which have helped us significantly improve our work.

1. Regarding the abstract, I maintain that the current version is overly general. I provided specific suggestions for improvement in my previous comments, yet the authors have made almost no changes to the abstract. I believe that carefully crafting the language to distill the core scientific information would not significantly increase the word count and could even make it more concise. The current abstract still lacks essential scientific evidence and in-depth quantitative analysis, making it unsuitable for a qualified research paper. Furthermore, the authors have not proofread this critical section carefully, as evident from the misaligned lines (between lines 36 and 37) and the presence of extra spaces or characters. I urge the authors to treat the revision process with greater seriousness.

Reply: We would like to thank referee #2 for pointing out the error we made in the abstract, which we should have avoided. For the third version (the latest version) of our manuscript, we have carefully checked the entire manuscript and supplement to avoid errors which we should avoid.

As referee #2 insisted that the abstract is overly general, we have decided to take his/her suggestion and revised the abstract: we have included core scientific information in the abstract, and deleted some non-critical words (in order not to exceed 250 words). Below is the updated abstract which can also be found in the revised manuscript (page 2): "Deposition of mineral dust aerosol into open oceans impacts marine biogeochemistry, and the deposition flux can be constrained using dissolved aluminum (Al) in surface seawater as a tracer. However, aerosol Al solubility, a critical parameter used in this method, remains highly uncertain. We investigated seasonal variations of aerosol Al solubility for supermicron and submicron particles at two locations (Xi'an and Qingdao) in northern China. Aerosol Al solubility was very low at Xi'an, showed no apparent variation with seasons or relative humidity, and was not correlated with sulfate or nitrate; in contrast, Al solubility was much higher at Qingdao, exhibited distinct seasonal variability, and increased with relative humidity and the abundance of sulfate and nitrate. All these features observed for Al solubility at the two locations can be explained by the effects of atmospheric chemical processing. Mineral dust transported to Xi'an (an inland city in Northwest China) was still not obviously aged and thus chemical processing had little effect on aerosol Al solubility; after arriving at Qingdao (a coastal city in the Northwest Pacific), mineral dust was substantially aged by chemical processing, leading to significant enhancement in aerosol Al solubility. Our work further reveals that aerosol liquid

water and acidity play vital roles in the dissolution of aerosol Al by atmospheric chemical processing. We suggest that chemical aging can lead to spatiotemporal variation of aerosol Al solubility, and this should be considered when using dissolved Al in surface seawater to constrain oceanic dust deposition."

2. I understand that the solubility of aluminum in dust deposited into the ocean can vary across different maritime regions and times, potentially significantly. However, this study only observes aluminum solubility at two terrestrial sites. The connection to the inference about oceanic dust deposition is not direct. Even if we clarify the spatiotemporal characteristics of aerosol aluminum solubility, how does that allow us to better constrain oceanic dust deposition? If the solubility of aluminum in dust varies greatly, how can we effectively use dissolved aluminum concentrations in seawater to constrain oceanic dust deposition? The logic behind this is unclear to me.

Reply: A good knowledge of spatiotemporal characteristics of aerosol Al solubility can inform us how to develop parameterizations of aerosol Al solubility, which can be used to better constrain dust deposition. To make it more explicit, we have made the following two changes in the revised manuscript.

1. Page 2: "We suggest that chemical aging can lead to spatiotemporal variation of aerosol Al solubility, and this should be considered when using dissolved Al in surface seawater to constrain oceanic dust deposition."

2. Page 4: "In order to better constrain the oceanic dust deposition using dissolved Al in seawater as a tracer, we need to develop parameterizations for aerosol Al solubility, and this requires spatiotemporal variability of aerosol Al solubility to be understood and processes and mechanisms which drive such variations to be elucidated."

Although our field observations were conducted only at two terrestrial sites, the results reveal that atmospheric chemical processes play an important role in controlling the variation of aerosol Al solubility. In the revised manuscript (page 27-28) we have added one sentence to discuss the implications and caveats of our work: "Although our measurements were only conducted at two sites, our work provides important insights into processes driving spatiotemporal variability of aerosol Al solubility, and such understanding can aid us to develop aerosol Al solubility parameterizations."

3. Concerning the issue of local resuspended dust, as the authors mentioned, its aluminum solubility is typically lower than that of desert dust, which is a consensus in many studies. However, the higher solubility observed in Qingdao compared to Xi'an does not automatically imply that local resuspended dust has a minimal influence in Qingdao. A more plausible explanation could be that emissions of local resuspended dust are much greater in Xi'an, thereby lowering the overall solubility there. In contrast, Qingdao might have less local resuspended dust, resulting in a relatively higher observed solubility. This is not even the most critical point. The more crucial issue is that if the interference from local resuspended dust is substantial, the paper's conclusions regarding the properties and transport of desert dust cannot be explained clearly and rationally.

Reply: We would like to point out that referee #2 may misunderstand what we stated. We did not state (previous studies did now show either) that Al solubility was lower for local resuspended dust than desert dust. In fact, Al solubility was always very low for soil and mineral dust samples examined in previous studies. In the revised manuscript (page 4) we have made the following change to make this clearer: "The initial Al solubility is generally low (typically <1.5%) for soil or mineral dust samples (Mulder et al., 1989; Duvall et al., 2008; Shi

et al., 2011; Aghnatios et al., 2014; Li et al., 2022)". Indeed we cannot exclude the contribution of local resuspended dust. This is why in the second version of our manuscript we changed "desert dust" to "mineral dust", in order not to exclude the contribution of local resuspended dust.

It is very likely that the contribution of local resuspended dust was lower in Qingdao than Xi'an. However, as Al solubility of local resuspended dust is not higher than desert dust, lower contribution of local resuspended dust can NOT explain either the much higher Al solubility (up to) observed at Qingdao or the dependence of Al solubility at Qingdao on RH and relative abundance of secondary species. In other words, we need to look for sources/processes which can enhance Al solubility. In Sections 3.2 and 4, we discussed several possibilities and came to the conclusion that atmospheric chemical processing dictates aerosol aluminum solubility.

4. In my previous comment, I pointed out that desert dust rarely occurs in Xi'an during winter because the major dust sources in northern China are typically snow-covered, with frozen or moist soil that prevents dust emission even under strong winds. Therefore, the dust observed in Xi'an during winter is likely predominantly local resuspended dust. In their response, the authors shifted the focus by stating that many studies show dust is a significant component of aerosols in Xi'an. However, this refers to the conditions in spring, not winter.

Reply: Besides spring, Asian dust events also occur in winter. On the other hand, we agree with referee #2 that local resuspended dust can also play a significant role; this is why in the second version we used "mineral dust" instead of "desert dust", in order not to exclude local resuspended dust. In the revised manuscript (Page 10) we have made the following change to provide further clarification: "Furthermore, besides spring, Asian dust also occurs in winter (Cai et al., 2020; Wang et al., 2020), and a previous study (Huang et al., 2014) suggested that the dust-related source, including local resuspended dust, contributed 56% to $PM_{2.5}$ during a severe haze event at Xi'an."

5. If the authors hypothesize that the dust samples originate from the Loess Plateau, which is close to Xi'an, they must provide substantial evidence to support this claim. It is important to distinguish concepts clearly: the Loess Plateau is generally not considered a dust *source* region but rather a depositional area for aeolian dust. The primary dust sources affecting China are located in southern Mongolia and China's own deserts (e.g., Taklamakan, Badain Jaran, Tengger, and Kubuqi deserts). These source regions are almost all over a thousand kilometers away from Xi'an, not "quite close" as suggested.

Reply: The Loess Plateau is a depositional region for Asian dust, but it is also an active source of Asian dust. In the revised manuscript (page 6) we have made the following change to clarify this: "Xi'an is an inland city in northwestern China, located at the southern edge of the Loess Plateau which is also an active source of mineral dust (Cao et al., 2008; Jeong, 2020; Haugvaldstad et al., 2024), and the aging extent of mineral dust at Xi'an was found to be quite limited (Wang et al., 2014; Wu et al., 2017)."

We fully agree that some dust sources are quite far from Xi'an, and the aging of dust particles transported to Xi'an is rather limited mainly because anthropogenic emission in Northwest China is much smaller. As a result, compared to the first version, we have made the following change in the second version (line 251-255, page 13, the third/current version): "There are several important dust sources in Northwest China, being far from (up to a few thousand km) or close to Xi'an. More importantly, anthropogenic emission in Northwest China is much smaller than the North China Plain, and thus the aging extent of mineral dust transported to Xi'an was rather limited (Wang et al., 2014; Wu et al., 2017)." Moreover, the original sentence in the second version "Xi'an is an inland city in northwestern China, and the

aging extent of dust was found to be quite limited at Xi'an ~~due to its proximity of desert regions~~ (Wang et al., 2014; Wu et al., 2017)" has been changed in the revised manuscript (page 6) to "Xi'an is an inland city in northwestern China, located at the southern edge of the Loess Plateau which is also an active source of mineral dust (Cao et al., 2008; Jeong, 2020; Haugvaldstad et al., 2024), and the aging extent of mineral dust at Xi'an was found to be quite limited (Wang et al., 2014; Wu et al., 2017)."

6. Finally, regarding the authors' explanation for the smaller difference in aluminum solubility between the two cities in spring—attributing it to faster transport due to higher wind speeds, thus less aging—it is important to note that major dust events are typically associated with strong winds during transport from west to east. Does this imply that the solubility of aluminum is less affected during these significant dust events, which are precisely the events of greatest interest for transport and deposition into the oceans? This point requires further clarification.

Reply: Indeed our work implies that the enhancement of aerosol Al solubility at Qingdao is limited during large dust events when large amounts of dust was emitted and despoited into the ocean. However, this does not necessarily imply that aerosol Al solubility remains low when dust particles are further transported to the open oceans, as Qingdao is a coastal site. To discuss this issue, we have added the following sentence in the revised manuscript (page 27-28): "Our work implies that during large dust events increase in aerosol Al solubility may be rather limited when dust is transported to Qingdao; nevertheless, when dust is transported further eastward to the open ocean, atmospheric chemical processing may substantially increase aerosol Al solubility."