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 #2

The paper systematically investigates the seasonal variations in aerosol aluminum solubility in two northern Chinese cities (Xi'an and Qingdao), revealing the critical influence of atmospheric chemical processes on aluminum solubility. The research topic holds certain scientific significance, as the study of aerosol aluminum solubility is expected to provide key parameters for accurately estimating mineral dust deposition fluxes in the context of marine biogeochemistry and global climate change. However, the paper devotes a significant portion of its content to presenting test data without in-depth analysis or detailed interpretation, resulting in insufficient depth and scientific value. Many conclusions are drawn at a speculative level, lacking solid evidence to substantiate them, which undermines the credibility of the article. The core conclusions of the paper currently carry considerable uncertainty and require further refinement; they should not be hastily drawn. Extensive revisions are recommended, and my suggestions and comments are as follows.

Reply: We would like to thank ref #2 for reviewing our manuscript and recommending it for publication after major revision.

We do not quite agree with the comment that we present data without in-depth analysis or interpretation. Our original manuscript uses ~7 pages (page 8-15) to present our result (Section 3), and ~10 pages (page 15-24) to analyze, discuss and interpret our results (Section 4). Due precisely to our in-depth data analysis and discussion, we could explain the difference in aerosol Al solubility at two locations and come to the conclusion that atmospheric chemical processing dictates aerosol aluminum solubility. There may be other unknown mechanisms which can explain our data, but the concolusion we have reached (i.e. atmospheric chemical processing dictates aerosol aluminum solubility) are consistant with our results and scientifically robust.

Nevertheless, we understand that there is always plenty of room for improvement. We highly respect and appreciate all the comments ref #2 raised, and have revised our manuscript accordingly; when we do not quite agree with ref#2, we have provided proper explanation. Please find more details below.

1. The abstract is overly vague and generalized. For instance, it should explicitly summarize the seasonal variation patterns of aluminum solubility in the two regions, the correlation with the relative abundance of sulfate and nitrate, and how their dependence on relative humidity (RH) differs between the two locations, rather than merely stating that they are "different."

Reply: Indeed we would like to provide further details about aerosol Al slolubility in the abstract. However, the abstract cannot exceed 250 words, as required by the journal. As a result, we cannot provide these details in the abstract; instead, we present them in the conclusion.

2. It is unclear why the authors claim that spatial differences in aerosol aluminum solubility must be fully considered when constraining oceanic dust deposition using dissolved aluminum concentrations in surface seawater. Although the solubility of aluminum in dust differs between Xi'an and Qingdao, and the authors attribute this to varying degrees of aging, dust transported from the same source region to the same oceanic area should undergo the same aging process, resulting in consistent aluminum solubility. It is unclear how the authors arrived at this conclusion.

Reply: We respectlt disagree with ref #2. First of all, over the oceans aerosol Al solubility can show spatial variation; furthermore, for a given region over the coean, aerosol Al solubility can vary great temporal variation, as dust particles arriving at a same point but at different times may have undergone different aging processes.

In fact, as pointed out in our original manuscript (page 3, line 62-64), previous work found that aerosol Al solubility could vary over the oceans by more than one order of magnitude.

3. The Introduction fails to clearly focus on the current major controversies and sources of uncertainty regarding the range of aluminum solubility variations and their influencing mechanisms. Some studies suggest a significant correlation between aluminum solubility and acidic components, while others hold opposing views. The Introduction should more clearly summarize these conflicting findings and unresolved mechanisms, identifying the specific "gaps" or "contradictions" the current study aims to address, thereby strengthening the research motivation.

Reply: Ref #2 rasied a very good point. As suggested, in the revised manuscript (page 5) we have added a few senetences to summarize the key gaps in our understanding of aerosol Al solubility: "It can be concluded that although aerosol Al solubility in the atmosphere was explored by several previous studies, our understanding is still very limited. For example, it remains unclear why aerosol Al solubility shows large spatial and temporal variation. Some work suggested that atmospheric chemical aging could enhance aerosol Al solubility, but the mechanisms and key environmental factors have not been elucidated. Furthermore, the effects of particle size on aerosol Al solubility have not been well understood."

4. How were the interferences from locally resuspended dust aluminum in Xi'an and Qingdao excluded? How is it proven that the dust observed in Xi'an and Qingdao originates from the same source and differs only in aging?

Reply: Indeed local resuspended dust, in addition to desert dust, could contribute to aerosol Al at both locations. In response to this comment, we have made the following changes:

1) In the revised manuscript we have changed "desert dust" to "mineral dust" so that we do not exclude the possible contribution of local resuspended dust.

2) Mineral dust from different desert regions and local resuspended dust cannot explain higher Al aerosol solubility in Qingdao, as previous work showed that Al solubility was low for soil samples from different regions. In the revised manuscripy (page 13) we have added one sentence to provide further explanation: "Mineral dust from different desert regions and local suspended dust cannot explain higher Al aerosol solubility observed at Qingdao, as previous work showed that Al solubility was low for soil samples from different regions (Shi et al., 2011; Wuttig et al., 2013; Aghnatios et al., 2014; Li et al., 2022; Hsieh et al., 2023)."

5. The study claims that its findings can be generalized to the "North Pacific dust pathway" or even the "global dust-ocean interface." However, the current design is based on only two sampling sites (one inland and one coastal), lacking gradient observations (e.g., multi-point trajectory analysis) or broad representativeness. The observed differences may be dominated by the unique characteristics of the sampling sites themselves (e.g., urban pollution, local humidity). How can such significant uncertainty be explained?

Reply: We think that this comment is related to the last paragraph in our manuscript where we discuss the implication of our work.

We found that compared to Xi'an (an inland site), aerosol Al solubility at Qingdao (a coastal site) was much higher, and we attributed this to chemical processing; as a result, it is justified to expect that aerosol Al solubility will further increase when mineral dust aerosol is transported to the Pacific. We also found that aerosol Al solubility at Qingdao showed

temperail variations, as the extent of aging also varied with time; as a result, at a given location over the oceans, aerosol Al solubility may also vary with time. Therefore, it is very justified to state that "when leveraging dissolved Al concentrations in surface seawater as a tracer to estimate deposition flux of mineral dust aerosol into open oceans, considering the spatial distribution of aerosol Al solubility, instead of using a uniform value on the global scale, can help us better constrain the oceanic deposition flux of mineral dust."

6. Why is aluminum concentration highest in winter? Mineral dust is not commonly observed in Xi'an during winter. Does this indicate that the source of aluminum in Xi'an is not mineral dust?

Reply: In fact many studies showed that mineral dust is a major component in aerosol particles at Xi'an. The major reason why aerosol Al concentrations was higher in winter than summer and autumn is that meteorological conditions in winter favored accumulation of aerosol particles. In the revised manuscript (page 10) we have added one sentence for further explanation: "Total aerosol Al concentrations were higher in winter than summer and autumn at Xi'an, and one major reason is that meteorological conditions favored the accumulation of aerosol particles (including aerosol Al) during winter (Cao and Cui, 2021)."

7. Lines 170-173: All the sites the authors compare are island observations, which are not strongly comparable to Xi'an. There are numerous observational results from inland China—why are these not mentioned for comparison?

Reply: Ref #2 rasied a good point. Total aerosol Al showed similar seasonal variations at other inland sites in North China (such as Zhengzhou and Beijing). As suggested, in the revised manuscript (page 10) we have included two studies carried out at Zhengzhou and Beijing, and deleted the two studies carried out at Hong Kong and Taiwan: "This was consistent with previous studies carried out in other locations in East Asia, such as Zhengzhou (Wang et al., 2019), Beijing (Zhang et al., 2013), Huaniao Island in the East China Sea (Guo et al., 2014), and Japan (Sakata et al., 2023)."

8. Lines 184-193: The comparative data presented here merely show that aluminum concentrations are higher at sites closer to dust source regions—a conclusion that is obvious and lacks significant scientific value. Could the authors supplement the discussion with differences in aluminum content (μg/g) in dust aerosols at sites at varying distances from dust sources? Analyzing changes in aluminum content during transport and their underlying mechanisms would be more scientifically valuable. The same applies to the analysis of soluble aluminum. Readers would prefer to see variations in aluminum content rather than just absolute concentration changes related to distance from dust sources.

Reply: It can be a good alternative to discuss change in aluminmun content at different sites; however, such data is not available as most of previous and our studies only report mass concentrations of aerosol Al. It can be expected that the increase in the transport distance will lead to decrease in Al content, since Al concentrations will decrease gradually while other aerosol components may increase.

9. The study presents aluminum concentrations in supermicron and submicron particles but merely displays the data without explaining its scientific significance. Why do aluminum concentrations differ between particle sizes? What mechanisms underlie these differences? How do the seasonal variation characteristics of aluminum content differ between particle sizes, and what causes these differences? The authors' data analysis needs strengthening; it should not be limited to simple data presentation.

Reply: In the revised manuscript (page 9) we have added the following sentences to describe and explain the general feature of size dependence of aerosol Al: "For each season the median concentration of total aerosol Al was usually higher in submicron particles than supermicron particles at both locations (and there were some exceptions, as shown in Figures 1a and 1b). This is related to size dependence of mineralogy and elemental compositions of mineral dust aerosol, which is not well studied and deserves further investigation." We also would like to point out that the focus of our manuscript is to understand aerosol Al solubility (as discussed in Section 4) while Section 3 is used to present relevant results .

10. Lines 220-224: What drives the seasonal variation in aluminum solubility? Why do differences in aluminum solubility exist between particle sizes? How do seasonal variations in aluminum solubility differ between particle sizes, and why? If there are no differences between particle sizes, then studying size-dependent features is unnecessary.

Reply: In fact, in our original manuscript we have discussed what drives the seasonal patterns of aerosol Al solubility at Xi'an and Qingdao (Section 4.2), and also discussed size dependence of aerosol Al solubility (Section 4.3). Ref #2 is kindly referred to relevant sections for further details.

11. Line 233: Here, the difference in aluminum solubility between Xi'an and Qingdao is attributed to transport distance. What evidence supports this claim, or is it merely speculation? How large is the uncertainty of this speculation, and how can it be validated?

12. The authors attribute the differences in aluminum solubility between the two cities to aging during transport. However, it should be noted that the distance from Xi'an to the Taklamakan Desert exceeds 3,000 km, while the distance from Xi'an to Qingdao is about 1,000 km. In other words, transport from Xi'an to Qingdao increases aging time by only about 30%, which is not a substantial difference. Without solid evidence proving the significance of this 1,000 km aging process, the core conclusion is difficult to accept. At present, this conclusion appears to be speculative.

16. Line 258: If the aluminum solubility in Xi'an's dust is very close to that at the source region, how can the impact of aging over nearly 3,000 km of transport on aluminum solubility be explained? Conversely, why does the 1,000 km transport from Xi'an to Qingdao have such a pronounced effect on aluminum solubility?

Reply: These three comments (No. 11, 12 and 16) are closely related, and therefore are addressed together. Aerosol Al solubility at Xi'an was not different from for dust samples collected over deserts, while was much higher at Qingdao. As we further discuss in Section 4, we suggest that all the features related to aerosol Al solubility can be only explained by the extent of chemical aging.

In addition to the Taklamakan Desert which is ~3000 km from Xi'an, there are several important dust sources in Northwest China which are quite close to Xi'an, such as China Loess Planteau. More importantly, anthropogenic emission over dust source regions in Northwest China is much smaller than North China Plain, and therefore mineral dust particles arriving at Xi'an are not much aged, as supported by previous work. In the revised manuscript (page 13) we have made the following changes to provide further explanation: "In three seasons (summer, autumn and winter), aerosol Al solubility at Qingdao was higher than that at Xi'an (Figure 3, Table S5). 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). On the contrary,

Qingdao is much farther from deserts; consequently, after long-distance transport over the North China Plain where anthropogenic emission is very large, mineral dust aerosol which arrived at Qingdao was substantially aged (Trochkine et al., 2003; Takahashi et al., 2011; Jeong, 2020), thereby leading to enhanced dissolution of aerosol Al and thus the increase in Al solubility."

13. Line 239: Why is the difference in aluminum solubility between the two cities minimal in spring? Spring is precisely the season when dust events are most significant in inland China. If aluminum solubility does not exhibit differences during this critical dust period, does this suggest that the tested aluminum solubility differences are not attributable to dust-derived aluminum?

Reply: As we further discuss in the original manuscript (line 268-278), aerosol Al solubility in spring was not different from dust particles collected over desert regions at either Xi'an or Qingdao. This is because higher wind speeds during dust events hindered the accumulation of atmospheric pollutants and shortened the transport time, limiting the aging of mineraldust aerosol.

Initial Al solubility was very low (and essentially identical) for mineral dust particles emitted from different regions. The difference in aerosol Al solubility at Xi'an and Qingdao was caused by atmospheric chemical aging, and this is the central message our work wants to deliver.

14. On the other hand, the impact of aluminum deposition on marine ecosystems should be evaluated from the perspective of dust load weighting. If, as the study shows, aluminum solubility is lowest during spring—when dust loads are highest—then the vast majority of annual dust-derived aluminum has poor solubility, calling into question its contribution to marine ecosystems. Moreover, does this imply that aluminum solubility in Xi'an and Qingdao is largely similar for most of the year, with differences only appearing in the less dusty seasons (summer, autumn, and winter), thus contributing minimally to annual totals? The authors should scientifically assess these differences in light of total dust transport.

Reply: I agree with ref #2 that one may conclude from our work that aerosol Al solubility is low in spring when dust loads are highest. However, it is beyond the scope of our manuscript to discuss the impact of dust deposition on marin ecosystems. The key meassage we would like to deliver is that atmospheric chemical aging dictates aerosol aluminum solubility and can explain its spatial and temporal variations.

15. Line 252: What are the concentrations of dust aerosols under different weather conditions? On clean or haze days, there is likely little dust transported from deserts, with local resuspended dust dominating. If aluminum solubility is similar under these conditions, does this suggest that solubility differences between desert dust and local resuspended dust in Xi'an are negligible, making it unreliable as a source indicator?

17. Line 283: How is it proven that dust sources are consistent across haze days, fog days, and clean days? How is it confirmed that aluminum solubility at the origin is identical and that the observed differences arise primarily from varying atmospheric chemical processes?

Reply: The two comments above (No. 15 and 17) are closely related, and thus are addressed together. In our original supplement (Tables S6 and S6) we have provided total and dissoleved aerosol concentrations under different conditions at Xi'an and Qingdao. Total aerosol Al concentrations were much higher for dust days. Because our manuscript is focused on aerosol Al solubility, we choose not to discuss in specific total Al aerosol concentrations under different weather conditions.

Indeed we cannot exclude the contribution of local resuspended dust. As a result, in the revised manuscript we have changed "desert dust" to "mineral dust". It is true that mineral dust aerosol at a given location can orginate from different source regions; however, original mineral dust, even from different regions, all shows very low Al solubility, as documented by a number of previous work. In the revised manuscripy (page 13) we have added one sentence to provide additional justication: "Mineral dust from different desert regions and local suspended dust cannot explain higher Al aerosol solubility observed at Qingdao, as previous work showed that Al solubility was low for soil samples from different regions (Shi et al., 2011; Wuttig et al., 2013; Aghnatios et al., 2014; Li et al., 2022; Hsieh et al., 2023)."

18. Line 285: The authors vaguely speculate that liquid-phase reactions enhance aluminum solubility. First, dust is a hydrophobic aerosol, and even on haze days, aerosol water content does not increase significantly. Can the authors provide data on how much aerosol liquid water content actually increased during their observations? Furthermore, what specific liquid-phase reactions promote aluminum dissolution? What triggers these reactions? Why do such reactions not occur on haze days in Xi'an?

Reply: Unaged mineral dust particles are largely hydrophobic, but they can still take up some water at high RH. More important, aging will increase the hygroscopicity of mineral dust. This exactly explains why aerosol Al solubility increases with RH at Qingdao (where dust particles are aged) but does not vary with RH at Xi'an (where dust particles are still not aged).

Based on previous literature and what we know for aqueous chemistry of Al, we suggest that acid and ligand processing can both enhance aerosol solubility, but at present it is difficult to disentangle their individual contributions. In the revised manuscript (page 16) we have added one sentence for further discussion: "Acid and ligand processing can both enhance aerosol Al solubility, although at present it is difficult to disentangle their individual contributions."

19. Based on the authors' analysis, the observed increase in aluminum solubility in Qingdao is more likely due to local atmospheric chemical processes (if their speculation is correct) rather than aging during transport. On haze days in Qingdao, it is unlikely that the entire dust transport pathway experiences haze conditions; instead, these are more likely dust days, representing cases with minimal aging and the least increase in aluminum solubility. Dust days are the most critical cases for annual dust transport from source regions to Xi'an and Qingdao. This suggests that large amounts of dust transported from deserts to Xi'an and Qingdao do not undergo significant aging to increase aluminum solubility. Instead, local haze in Qingdao may enhance aluminum solubility, but this accounts for only a small fraction of annual dust, which is not the dominant component.

Reply: Indeed our work indicates that in spring when large amounts of mineral dust aerosols are transported to Qingdao, aerosol Al solubility does not increase a lot because the aging of mineral dust is very limited. However, since the focus of our manuscript is on the variation of aerosol Al solubility, we choose not to make this statement more explicitly. Modeling studies can give more quantitative results in this aspect.

20. Figure 5 should include the p-values of the fits. Has the fit passed significance testing? The correlation appears weak, making it difficult to support the authors' claim of an inverse relationship.

Reply: Compared to previous work which reported such inverse dependence, the inverse dependence we reported is quite obvious. We have further tried to fit this inverse dependence using the exponential function, because such quantative relationship can be very useful for

modeling studies. Although the *r* values are not very large, the *p* values are all <0.01. In the revised manuscript (page 17), we have updated Figure 5 to include *p* values.

21. Line 344: On rapidly transported dust days, dust in Xi'an may exhibit external mixing with acidic components, whereas on haze days, over 95% of dust is internally mixed with acidic components. The authors could separately analyze aluminum solubility's relationship with acidic components on haze days and dust days to validate their explanation.

25. Line 410: I recommend categorizing samples into dust days and haze days to examine the impact of mixing state.

Reply: These two comments (No. 21 and 25) are closely related, and are therefore addressed together. We checked the dependence of aerosol Al solubility on acid species on haze and dust days separately, and there was still on correlation; one reason is that the numbers of haze and dust days were very small.

As suggested by the referee, we further examined the samples with low Al solubility, high RH and high acidity. These samples were mostly found in clear days, perhaps due to the influence of local resuspended dust for which chemical aging was very limited. In the revised manuscript (page 24), we have added one sentence to discuss these special samples: "We also note that samples with low Al solubility but high RH and high acidity were mostly found in clean days, perhaps due to the influence of local resuspended dust for which chemical aging was very limited."; moreover, I have added anotehr sentence to discuss future work which can provide further insights: "Single particle analysis which provides mixing state information can give further insights."

22. In Figure 6, "r" is used, while Figure 5 uses "r^2," and Figure 6 includes p-values. It is recommended to standardize the plotting conventions.

Reply: As suggested, in the revised manuscript (page 17) we have updated Figure 5, and the new version uses "*r*" and includes *p* values.

23. Line 377: The analysis here focuses only on the relationship between local RH variations and aluminum solubility in Xi'an. If high RH promotes liquid-phase reactions that increase aluminum solubility, then aluminum solubility should also rise during high-RH conditions in Xi'an. The role of local chemistry should not depend on the distance from the desert.

24. Line 379: It is difficult to understand why high RH in Qingdao increases aluminum solubility but not in Xi'an. Atmospheric chemical processes should be similar—are there other controlling factors influencing this aluminum solubility mechanism?

Reply: These two comments (No. 23 and 24) are closely related, and are therefore addressed together. The different RH dependence of Al solubility at two two locations was due to the difference in aging extents. In the revised manuscript we have made the following two changes to make our explanation more explicit:

1) "This again may imply that chemical processing had very limited impact on aerosol Al solubility at Xi'an, as mineral dust particles mostly remained externally mixed with secondary species and their aging extent was very limited (Wang et al., 2014; Wu et al., 2017)." (page 21).

2) "In contrast, RH played an important role in regulating aerosol Al solubility at Qingdao, because mineral dust particles observed at Qingdao had been transported through the North China Plain and were substantially aged." (page 22)

26. Line 416: I disagree with labeling dust in Xi'an as "fresh dust," given that it originates over 3,000 km away.

Reply: In fact in our original manunscript (Line 414-416), we state that "aging extent of dust particles as rather limited at Xi'an", and "fresh" was used to describe dust samples used by Shi et al. (2011) who investigated topsoil samples collected over deserts. In order to avoid misundstanding, in the revised manuscript (page 24) we have changed "fresh" to "unaged".