We thank the reviewers for their comments and respond to their suggestions below in blue font.

Reviewer 1:
This work details results of aerosol measurements during a pollution event in South Korea. Detailed size and composition measurements are presented from Incheon and Seoul showing increased levels of pollution and the possibility of secondary aerosol production due to high relative humidity. This event had higher than expected levels of aerosol nitrate in comparison to previously studied pollution events. This is believed to be the case because of low temperatures (along with the high RH). Previous pollution events studied have been in the summer when higher temperatures did not favor nitrate formation.

The analysis is sound, and this paper provides another example of secondary production catalyzed by high humidity during these pollution events. The results are not necessarily novel but they do lead credence to the role of secondary aerosol formation during these events – high relative humidity associated with these events exasperates air quality by increased oxidation of locally emitted NO2 and SO_2. This also shows that this occurs in different seasons but can result in a different aerosol mix (higher nitrate).

Overall, the approach is reasonable. However, some revisions are needed before publication. A key flaw in the paper is one of its findings states that based on a size-resolved analysis of composition that composition of PM1 can be used to understand PM2.5 composition. However, this is not necessarily true based on this analysis. This just shows for this one campaign where the aerosol composition seems to be atypical from other pollution events that it is a good proxy. In addition, as noted in the text, this is not the case for clean or "transition" periods. More analysis is needed to determine how prevalent these nitrate rich aerosol pollution events are, to see if this correlation between submicron and supermicron composition holds. Additional suggestions are provided below.

Response: Thank you for the positive view of the work. We respectfully disagree that our finding of similar composition between $PM_{2.5}$ and PM_1 is atypical. We think this point conflates two separate issues, please see our specific response below clarifying this point.

However, the emphasis the reviewer placed on this event being atypical due to high nitrate led us to a renewed check of the literature. When we began this work, we found far more references focused on elevated sulfate in East Asian haze, but more recently work is coming out on high nitrate in haze events. We have added two such references, removed the word "atypical" from the manuscript and amended Section 6.4 (now titled Nitrate-dominated Haze Event) as follows:

> "High NO_3^- events were not particularly common, as shown especially for Beijing (Yang et al., 2017), requiring favorable conditions such as cold temperatures, high humidity, a shallow boundary layer, and high precursor levels. However, with reductions in sulfate precursor emissions, high nitrate events are increasingly reported in the literature (e.g., Xu et al., 2019; Zhou et al., 2022)."

Minor Revisions:

- Line 106: Fig. S1 should be in the main text and a note on the distance between Seoul and Incheon. Figure S1 should include the location of the Incheon Met Site

Response: We added the location of the Incheon Meteorological Site to Figure 1 (formerly Figure S1). Other figure numbers were adjusted to account for the figure addition to the main article file. We added the distance between Seoul and Incheon in the following line of text:

> “The focus of this study is on three specific monitoring sites in Incheon and Seoul (~30 km apart), which were compared to a wide network of other stations in those cities to confirm agreement in temporal variability and concentrations.”

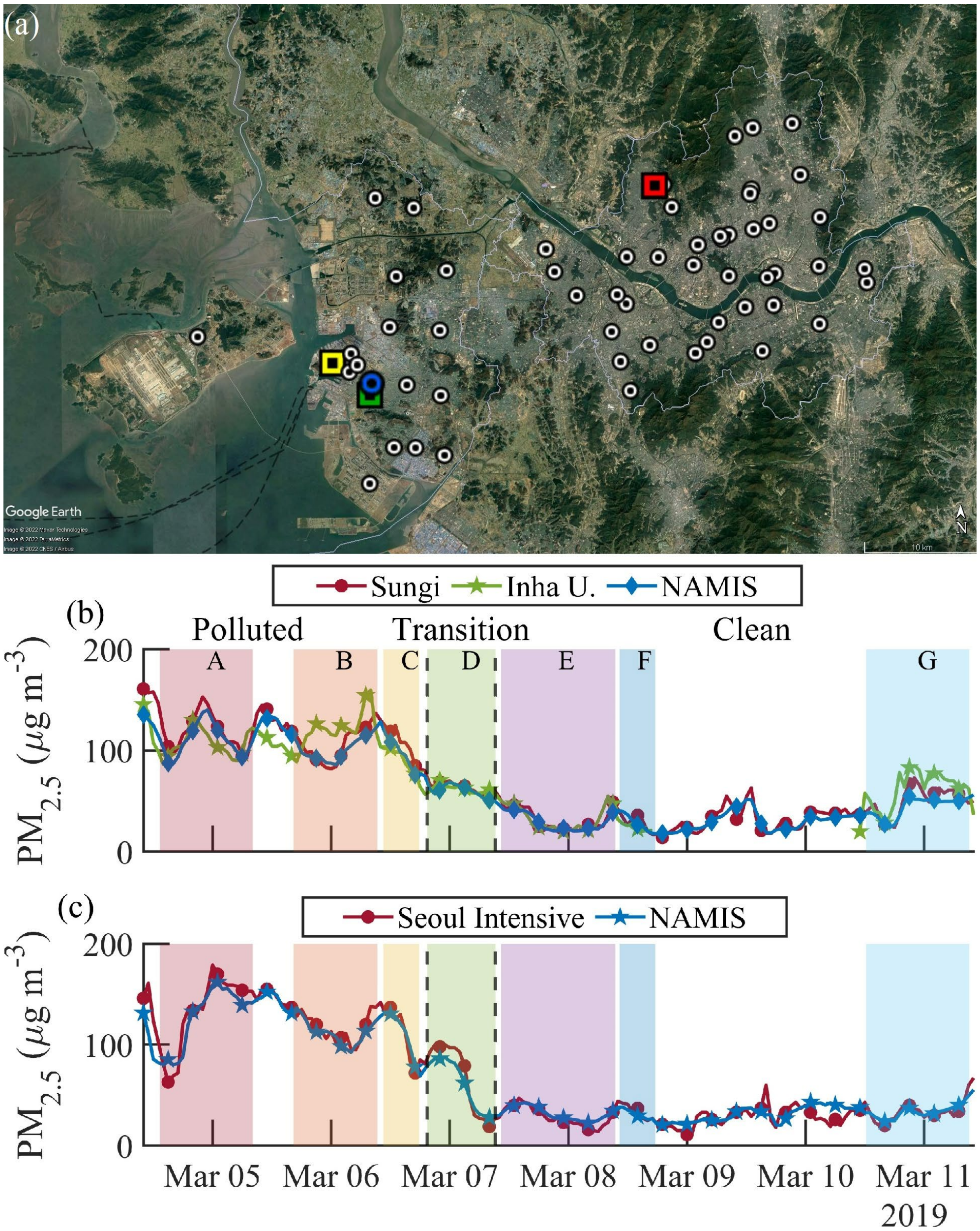

Figure 1. (a) Spatial map showing the 17 and 40 National Ambient air quality Monitoring Information System (NAMIS) stations in Incheon and Seoul, respectively, along with the three main surface sites relied on for this study (yellow = Incheon meteorological site, green = Inha University, blue = Sungi [also a NAMIS station], red = Seoul Intensive Monitoring Station). $PM_{2.5}$ comparison between (b) city-wide Incheon mean values and those for Sungi and Inha University, and between (c) city-wide Seoul mean values and those for Seoul Intensive Monitoring Station. Coefficients of determination (R^2) between the data points: (b) (Inha University) $R^2 = 0.82$ and (Sungi) $R^2 = 0.98$; (c) $R^2 = 0.\ 96$. Shaded regions of panels b-c are labeled with individual $DLPI^+$ sets overlapping in time. All times are reported in Korea standard time (KST), where KST is UTC + 9 hrs.

- Line 151: I am not that familiar with the OPC-Grimm 1.109, but I believe it does not actually measure aerosol mass. It measures size distribution and then a density and calibration is used to calculate mass (from an online manual this may be done with dolomite dust). This should be addressed in the text. In addition, it gives a reason to use the Sungi PM2.5 in subsequent analysis as it is the same method as used in Seoul (which you typically did).

Response: The OPC measures the number concentration of aerosol particles for 31 channels with size ranging from 0.253 to 31.15 µm. These number concentrations are converted into a mass concentration via mathematical extrapolation with a correction factor specific to the Grimm 1177 software. As is common in studies using OPC-Grimm data (e.g., doi: 10.4209/aaqr.2009.05.0037) we compared the $PM_{2.5}$ values from this method to another method; more specifically we compared $PM_{2.5}$ from the OPC-Grimm at Inha to the mean of data around Incheon from 17 NAMIS stations based on the BAM method and found excellent agreement ($R^2 = 0.82$). Therefore, there is confidence in the $PM_{2.5}$ data at Inha.

To address this comment we added the following line of text to the paper:

> “The OPC measures the number concentration of aerosol particles for 31 channels with size ranging from 0.253 to 31.15 µm, which then get converted into a mass concentration via mathematical extrapolation with a correction factor specific to the Grimm 1177 software.”

And as the reviewer noted, we already make use of Sungi $PM_{2.5}$ in comparisons to Seoul as they are based on the same measurement method for $PM_{2.5}$.

- Line 415-418: You state that the reductions in CO from the polluted to clean periods could be due to Chinese influence. However, if this were solely the case, wouldn’t CO be higher (or comparable) at Incheon than Seoul. Seoul always has higher CO than Incheon. Maybe some of the enhancement is due to Chinese transport but in addition, the shallow boundary layer may allow for an increase in CO due to local emissions. Looking at CO/CO2 would help in the future.

Response: Excellent point and we added this text:

> "Carbon monoxide concentrations in Seoul exceeded Incheon by 177 ppb on average during the polluted period, suggestive of added influence from local emissions in Seoul superimposed on top of the transported pollution."

We agree that the use of gas ratios is useful for future work.

- Line 434-436. It is unclear what you are calculating by relative fraction in supermicrometer to all sizes. Does the 43% mean 43% of the sulfate is in the supermicron and 57% is in the submicron? Please clarify.

Response: We start the paragraph with a new sentence to provide context for why we discussed the relative fraction of the supermicrometer sizes to all sizes:

> "One finding of this work is the significant amount of secondarily produced species in the supermicrometer range. More specifically, the relative fraction of SO_4^{2-}, NO_3^-, NH_4^+, and organic acids in the supermicrometer range (i.e., technically $D \geq 0.94$ µm) as compared to all sizes sampled at Inha ($D \geq 0.016$ µm) during the polluted period was 43%, 44%, 42%, and 36%, respectively, which is appreciable and potentially influenced by the humid conditions."

Furthermore, yes, the other 57% would be submicrometer but we think this is already quite clear and makes the text more distracting to have to clarify this in parenthesis or another entire sentence.

- Line 496-499: a statement is made that based on this campaign that PM1 composition measurements can be assumed to be the same as PM2.5 composition for modelling purposes. A cautionary note is included starting on line 501 stating that this is not the case for cleaner periods. While this is true for this case, it may not be the same for other haze events such as those mentioned when temperatures are higher and therefor the composition is different (less nitrate). It isn't clear how frequent these low temperature haze events are and in fact on line 524-527 you state this was a fairly uncommon event. The cautionary note here should be expanded to show the limitations of analysis from this one event for understanding the PM1-to-PM2.5 connection (here and also in the conclusion – line 557-558).

We do not understand why the reviewer believes that the apportionment of the composition between PM_1 and $PM_{2.5}$ would differ between haze events as a function of temperature. Temperature would only shift the balance between nitrate and sulfate, not their apportionment as a function of size. It is important to note the large difference in aerosol mass concentrations between haze and non-haze conditions. As shown in Figure 6, the mass concentration of $PM_{0.94}$ is nearly 5 times that of the clean period, while the $PM_{2.5-0.94}$ is 11 times that of the clean period. That is, during haze, the processes that drive aerosol production and accumulation at the surface overwhelm other contributors to the extant aerosol mass. We don't find the results shown here to be particularly unique. They confirm expectations from results of prior studies. To the extent that any caution is warranted, it is conveyed in the statement we have already made as follows in Section 6.3: "However, the differences evident in the transition and clean periods imply that under other atmospheric conditions PM_1 composition measurements will not fully reflect the apportionment of $PM_{2.5}$ aerosols". This is in fact one of the things that makes haze events interesting and different from other meteorological conditions: under other conditions differences between sub- and supermicrometer composition would be expected.

Further, we already had this text pointing to the importance of learning how common these low temperature haze events are:

> “…points to the importance of conducting measurements at different times of the year to more fully understand haze formation and its impacts on air quality.”

For these reasons we have not added any further text to address this point. However, as we explained above, we believe some of the confusion here arose from our use of the word “atypical”. Recent work has shown that this is becoming commonplace (after 2017) with recent emission reductions. So, we have changed the section title to: Nitrate-dominated Haze Event.

We revised the following text to

> “High NO_3^- events were not particularly common, as shown especially for Beijing (Yang et al., 2017), requiring favorable conditions such as cold temperatures, high humidity, a shallow boundary layer, and high precursor levels. However, with reductions in sulfate precursor emissions high NO_3^- events are increasingly reported in the literature (e.g., Xu et al., 2019; Zhou et al., 2022).”

Typos/Suggestions:

- Table 1: include elevation; I think all are near sea level but it should be included

Response: Elevation information has been added to Table 1 as shown below:

Table 1. Sample site name, elevation in meters above sea level (MASL), latitude and longitude coordinates of each site, and the parameters measured at each site.

Site	Elevation (MASL)	Latitude	Longitude	Measurements
Incheon: Inha University	23	37°27’2.08”N	126°39’20.87”E	Aerosol: $PM_{2.5}$, 467/528/652 nm wavelength absorption coefficient (α), size-resolved particulate mass samples
Incheon: Sungi	46	37°27’34.74”N	126°39’27.31”E	Aerosol: $PM_{2.5}$ Gas: Ozone (O_3), nitrogen dioxide (NO_2), carbon monoxide (CO), sulfur dioxide (SO_2)
Incheon Meteorological Site	70	37°28’39.85”N	126°37’28.40”E	Meteorological: Ambient temperature (T), wind speed and direction, ambient relative humidity (RH), ambient pressure (P), rain
Seoul Intensive Monitoring Station	30	37°36’38.40”N	126°56’1.36”E	Aerosol: PM_{10}, $PM_{2.5}$; speciated $PM_{2.5}$ concentrations of sulfate (SO_4^{-2}), nitrate (NO_3^-), chloride (Cl^-), sodium (Na^+), ammonium (NH_4^+), potassium (K^+), magnesium (Mg^{2+}), calcium (Ca^{2+}), organic carbon (OC), elemental carbon (EC), silicon (Si), titanium (Ti), vanadium (V), manganese (Mn), iron (Fe), nickel

				(Ni), copper (Cu), zinc (Zn), Arsenic (As), selenium (Se), lead (Pb) Gas: SO_2, NO_2, O_3, CO Meteorological: T, wind speed and direction, RH, P, rain

- Line 142: “Furthermore, the” should be “The”

Response: Fixed.

- Line 199/208: BAM is an abbreviation for Beta Attenuation Monitoring

Response: Fixed

- Line 251: how can secondarily-produced species include primary organic aerosols? Should it be “includes primary organic aerosols and secondarily-produced species (i.e., SO_4^{2-} and SOA)”

Response: Fixed:

> “Reanalysis output is provided for dust, sea salt, open biomass burning smoke, and “anthropogenic and biogenic fine (ABF)” that includes secondarily produced species (i.e., SO_4^{2-} and SOA) and primary organic aerosols mainly confined to the fine mode (< 1 µm).”

- Table 3: caption repeats the dates related to the polluted, transition and clean time periods. This has already been stated so do not repeat it (it just makes the caption more cumbersome).

Response: Fixed by removing that sentence from Table 3 caption.

- Table 3 and following: ozone, NO2 and SO2 should be reported in ppb. CO can be in either ppb or ppm

Response: Fixed and reflected in revised forms of Tables 3 and 4, in addition to Figure S9.

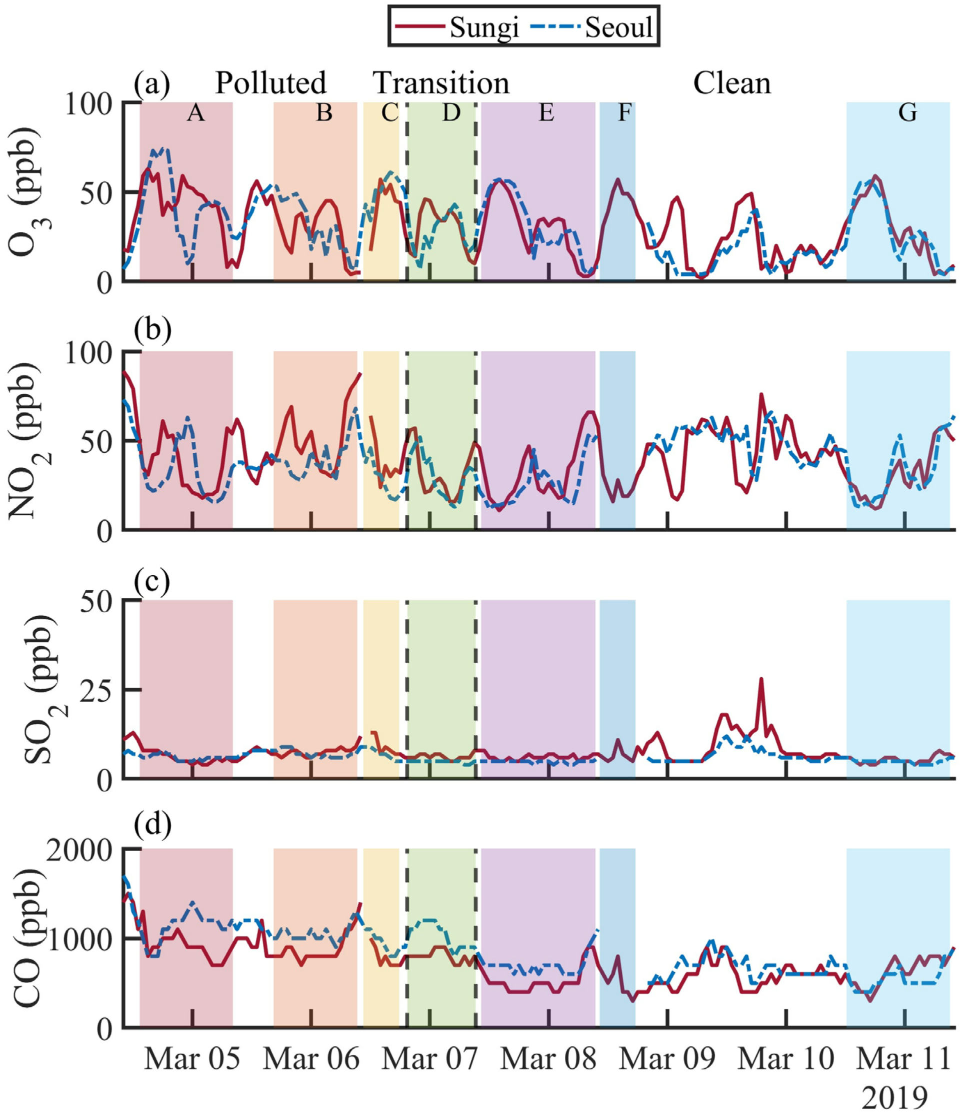

Figure S9. Time series of the following gas species measured at the sites denoted by the colors in the legend: (a) ozone (O_3); (b) nitrogen dioxide (NO_2); (c) sulfur dioxide (SO_2); and (d) carbon monoxide (CO). The dashed black vertical lines separate the (left) polluted, (middle) transition, and (right) clean periods. Shaded regions are labeled with individual DLPI$^+$ sets overlapping in time.

- Line 284-285: For comparison. What is the Seoul - Sungi value for the entire polluted period, entire transition period and entire clean period.

Response: We truncated the former sentence replacing the original comparison with the following added sentence that we feel is more helpful:

> "For example, the maximum/mean ± standard deviation in the $PM_{2.5}$ difference (µg m^{-3}) between Seoul and Sungi were as follows for the three periods: polluted = 59/10 ± 26; transition = 36/6 ± 28; clean = 42/-6 ± 14."

- Line 360: The sentence starting with "Oxalate is produced" should be moved before the sentence starting "The strong correlation"

Response: Fixed.

- Line 455-457: it is confusing to discuss both NOR and SOR value at the same time. Discuss nitrate in one sentence. And then sulfate.

Response: Revised:

> "The mean (± standard deviation) of NOR during the three time periods of the study was as follows: polluted = 0.39 ± 0.1; transition = 0.22 ± 0.10; clean = 0.09 ± 0.04. Similarly, the mean (± standard deviation) of SOR was as follows: polluted = 0.51 ± 0.06; transition = 0.44 ± 0.14; clean = 0.14 ± 0.07."

- Line 558: "for haze pollution." should be "for this haze pollution event."

Response: Fixed.

Reviewer 2:

The paper investigated size-resolved aerosol composition during a transboundary pollution event and concluded that a significant amount of secondary aerosols (mostly inorganic) in the accumulation mode (particle diameters ranging from 0.94 to 2.5 μm) can be formed during the severe haze episode. Their main conclusion is that the composition apportionment of submicron particles (PM0.94) is essentially the same as that of supermicron particles (PM2.5–0.94) if inorganic aerosols are the main constitution, which is most likely the case of a severe haze. This result may add value to the relevant research field as a new finding. However, this work focuses on mostly delivering their measurement results without in-depth analyses.

Response: By way of background, some of the authors on this work were deeply involved in the analyses that led to the aerosol synthesis paper from KORUS-AQ (Jordan et al., 2020) and the aerosol chapter in the Final Report from that campaign. During that work there were lengthy discussions of whether or not it was appropriate to assume that the $PM_{2.5-1}$ fraction had the same composition as the measured PM_1 composition. Hence, for those of us whose work primarily focuses on field measurements and data analysis, the result in this work that shows that the composition of the $PM_{2.5-0.94}$ fraction is in fact the same as the $PM_{0.94}$ fraction (during the polluted haze period) is an important one. Furthermore, we disagree about the lack of in-depth analyses as we go deep into the field data, use complementary datasets, and go beyond just plotting up raw data. We reach a number of important conclusions that rely on the in-depth analyses we did.

So, in my opinion, this work may be meaningful in terms of reporting another measurement datasets in the early spring season.

Response: If the reviewer is asking for more early spring data, there is none to our knowledge that is publicly available for the same study region of the nature we use (size-resolved composition). This makes the results of this work increasingly important and rare.

However, I do not think that this paper advances our fundamental understanding of secondary aerosol formation under favorable meteorological conditions (i.e., humid and cold environments). A large amount of secondary inorganic aerosols (sulfate, nitrate and ammonium) under stagnant conditions characterized by weak wind, shallow boundary layer, high humidity and/or low temperature, has been reported in numerous studies as the authors introduced. Large increases in sulfate and nitrate aerosols during the pollution event are therefore nothing special, even though a much large increase in nitrate aerosol is observed due to the lower air temperature as compared to the result found from the previous campaign (KORUS-AQ campaign). The latter is also an expected and already known result.

Response: We disagree. This reviewer assumes that the processes responsible for the production of the inorganic ions in East Asian haze are well understood. This is not the case. See Jordan et al. (2020) in the reference list and Travis et al. (2022) currently in review at ACPD (https://acp.copernicus.org/preprints/acp-2021-946/).

There are rare datasets examining such haze events in the study region with detailed size-resolved composition data. Our results are important and unique. They are needed to validate assumptions and speculations based on past bulk aerosol measurements. The findings about the enhanced nitrate due to colder temperatures may be common according to basic aerosol thermodynamics of ammonium nitrate formation, but the size-resolved nature of the data and the relative abundance of that species as compared to others as a function of size is uncommon. The actual absolute concentrations of the species we detected (especially relative to each other) is important information that in our view is special from the measurement perspective.

I personally wish they could focus more on oxalate (and/or organic acids) and aqueous secondary organic aerosol (SOA), which is much less known compared to secondary inorganic aerosol.

Response: We investigated oxalate and organic acids in this study. We direct your attention to Table S1. Adipate, Maleate, MSA, Oxalate, and Phthalate were all measured from the DLPI$^+$ size-resolved filters at Inha University. With the exception of oxalate, these compounds were all below detection in ≥ 85% of the samples. These are not major contributors to haze events. Nonetheless where they were above detection they were consistent with enhanced concentrations in the haze event (Fig. 4).

Formation of aqueous SOA occurs in haze events as has been reported in previous studies, but it provides a minor contribution compared to nitrate, sulfate, and ammonium. Looking at the difference in the mass concentrations (rather than enhancement factors) between the polluted and clean periods in Table 4 makes this clear. The sum of the Polluted-Clean differences for nitrate, sulfate, and ammonium is 72 $\mu g\ m^{-3}$ compared to the difference in OA of 7 $\mu g\ m^{-3}$, less than 10% of the 3 major inorganic components. During the pollution event, oxalate itself represented < 1% of the total mass concentration of the Inha samples. SOA is not the driver of haze events.

The fact that oxalate exhibits a greater enhancement ratio in Table 3 than that of OA in Table 4 is not surprising since not all OA is produced via aqueous processing and even components that are may be produced at different rates. Hence, oxalate is not a good proxy for OA overall in haze.

We point the reviewer to the following key pieces of text that we felt could help address this comment:

"OA was only enhanced by a factor of 1.7. In terms of mass concentrations, the difference between the polluted and clean periods for the sum of nitrate, sulfate, and ammonium was 72 $\mu g\ m^{-3}$ versus the difference in OA of 7 $\mu g\ m^{-3}$ (Table 4). The change in OA is < 10% of the change in the three major inorganic ions."

"The likely formation pathway for SO_4^{2-}, NO_3^-, NH_4^+, and organic acids in the polluted period was secondary production, which was assisted in part by high humidity as discussed in more detail in Sect. 6.2. Their common formation mechanism is supported by significant correlations ($r \geq 0.94$; see Fig. S5) during the polluted period between SO_4^{2-}, NO_3^-, NH_4^+, and oxalate, with the latter being the most abundant organic acid during the entire study period but especially in the polluted period (~70% of organic acid mass). Oxalate is produced efficiently via aqueous-phase chemistry (Sorooshian et al., 2007;Sorooshian et al., 2006;Wonaschuetz et al., 2012). The strong correlation between oxalate and SO_4^{2-} during the polluted period is important as a strong correlation between these species (in the absence of biomass burning) is considered a marker for secondary aqueous aerosol formation (Ervens et al., 2004;Yu et al., 2005;Zhou et al., 2015;Hilario et al., 2021a). The fact that oxalate exhibits a greater enhancement ratio in Table 3 than that of OA in Table 4 is not surprising since not all OA is produced via aqueous processing and even components that are may be produced at different rates. Thus, it is cautioned that oxalate is not a good proxy for OA overall in haze."

1. Throughout the paper, they stated multiple times the potentially important role of local emissions from Seoul in the higher levels of PM2.5 in Seoul than in Incheon or Sungi. Considering the larger population in Seoul, this is an obvious statement. It would be much better if they could provide some quantification of contributions of local emissions vs. transboundary transport to the higher PM levels in Seoul. With regard to local emissions, on the other hand, I wonder why PM2.5 concentration in Inha University (= 43.6 μg m^{-3}) or Sungi station (=36.2 μg m^{-3}) during the clean period is higher than that in Seoul (= 30.5 μg m^{-3}) (Table 3 and Table 4). Under clean conditions, it is expected that local emissions would contribute most to PM2.5 levels. I agree that emissions from Seoul would be higher than those in Incheon, but how can these results be interpreted?

Response: Quantifying the contribution of local versus transboundary is complex, depending not only on a more extensive dataset than we have for this event but also on model parameterizations needed for the calculation. Travis et al. (2022; https://acp.copernicus.org/preprints/acp-2021-946/) carefully examine the issues involved using the KORUS-AQ data set. Hence, such a calculation is beyond the scope of this work.

2. line 37, the high correlation between oxalate and sulfate is not directly provided with figures or tables in the manuscript. I presume that this is an important finding as they explicitly mentioned this in the abstract. So, I would suggest presenting a figure that directly shows timeseries of SO4, NO3, NH4, and oxalate (at least in supplement). In addition, further discussion seems to be still required. The authors noted that the high correlation between the two species indicates a secondary aqueous aerosol formation. Many studies reported elevated levels of SOA under humid or foggy conditions and speculated that these are associated with aqueous phase SOA formation. The higher oxalate concentration during the polluted period (0.7 μg m-3, Table 3) than that during the clean period (0.2 μg m-3) seems meaningful with a factor 3.5 enhancement, which is not negligible. However, OA in Seoul is only enhanced by a factor 1.7 (line 313-314). In fact, this enhancement actually reflects OC enhancement because they applied a constant factor (1.8) to OC to calculate OA. Although the authors said the mass closure is good, the remaining mass (PM2.5 minus the sum of all aerosol species except for OA) relative to OC is largest in the polluted period, followed by transition and clean periods. That is, for example, the sum of all aerosol species except for OA is 100.3 μg m^{-3} in the polluted period, and the ratio of the differential mass (= 127.2 – 100.3 = 26.9 μg m^{-3}) to OC (= 9.4 μg m^{-3}) is 2.86 while it is 1.75 in the clean period (Table 4). I understand that all of the remaining mass would not be OA, but part of it should be OA. I wonder if the authors can

provide some insight into (or evidence) the increased level of OA or SOA during the pollution event. Besides, I doubt if a constant factor of 1.8 can be applied to both polluted and clean episodes.

Response: We have added the requested time series figure (Figure S5) to illustrate the high correlation between SO_4^{2-}, NO_3^-, NH_4^+, and oxalate:

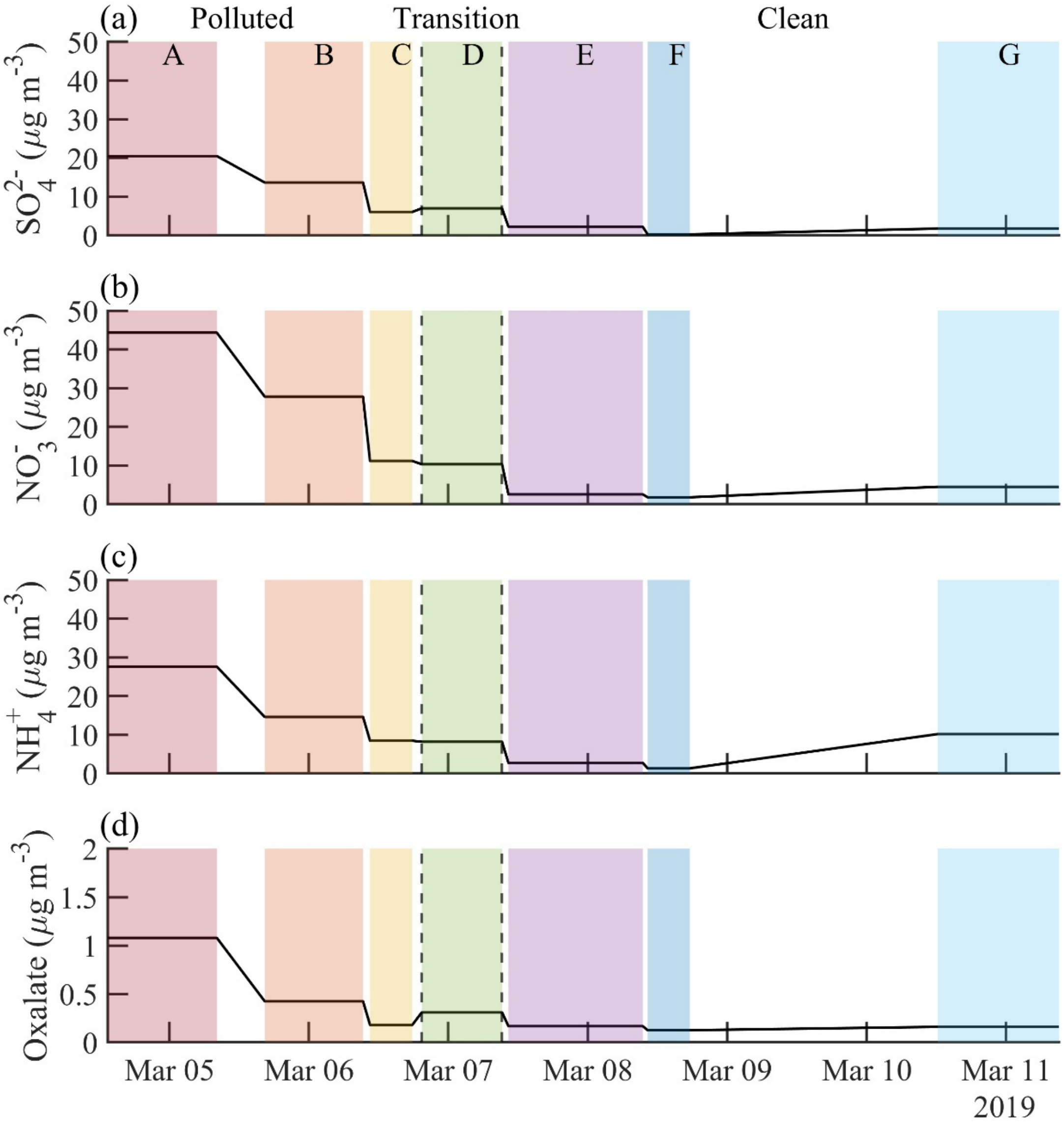

Figure S5. Time series of total sulfate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+), and oxalate measured at Inha University. The dashed black vertical lines separate the (left) polluted, (middle) transition, and (right) clean periods. Shaded regions are labeled with individual DLPI$^+$ sets overlapping in time.

The following sentence was updated to include this information:

> “Their common formation mechanism is supported by significant correlations ($r \geq 0.94$; see time series in Fig. S5) during the polluted period between SO_4^{2-}, NO_3^-, NH_4^+, and oxalate, with the latter being the most abundant organic acid during the entire study period but especially in the polluted period (~70% of organic acid mass).”

The point about the missing mass is interesting. The reviewer’s calculations are correct in that the ratio of $PM_{2.5}$ mass unaccounted for by speciated measurements in Seoul relative to OC in the polluted and clean periods was 2.86 (unaccounted $PM_{2.5}$ = 26.9 μg m^{-3}; OC = 9.4 μg m^{-3}) and 1.75 (unaccounted $PM_{2.5}$ = 9.8 μg m^{-3}; OC = 5.6 μg m^{-3}), respectively. However, we are not convinced it is necessary to go down this path of digging deeper into potentially more organic aerosol formation in the polluted period with uncertainties associated with other types of species we may not be fully resolving and lack of information about an accurate conversion factor of OC to OA as a function of time.

3. In Fig. 5, the sum of PM0.94 and PM2.5-0.94 (e.g., 66.9 μg m^{-3} in polluted case) and the sum of all species listed in Table 3 (= 73.3 μg m^{-3} in polluted case) do not match. Any explanations?

Response: The reviewer overlooked the Table 3 caption which said “Speciated concentrations from Inha represent mass for particles with diameters above 0.016 μm (i.e., full size range of $DLPI^+$ sampler)”; therefore, the sum of speciated masses in Table 3 includes sizes larger than 2.5 μm and thus cannot be compared directly with Figure 5. No changes are needed for this comment as it was just a misunderstanding by the reviewer.