

Comment on acp-2021-1098

Anonymous Referee #2

Referee comment on "Evidence of haze-driven secondary production of supermicrometer aerosol nitrate and sulfate in size distribution data in South Korea " by Joseph S. Schlosser et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-1098-RC1>, 2022

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 ($\text{PM}_{0.94}$) is essentially the same as that of supermicron particles ($\text{PM}_{2.5-0.94}$) 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. So, in my opinion, this work may be meaningful in terms of reporting another measurement datasets in the early spring season. 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. 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.

- Throughout the paper, they stated multiple times the potentially important role of local emissions from Seoul in the higher levels of $\text{PM}_{2.5}$ 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 PM_{2.5} concentration in Inha University (= 43.6 $\mu\text{g m}^{-3}$) or Sungi station (=36.2 $\mu\text{g m}^{-3}$) during the clean period is higher than that in Seoul (= 30.5 $\mu\text{g m}^{-3}$) (Table 3 and Table 4). Under clean conditions, it is expected that local emissions would contribute most to PM_{2.5} levels. I agree that emissions from Seoul would be higher than those in Incheon, but how can these results be interpreted?

- 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 $\mu\text{g m}^{-3}$, Table 3) than that during the clean period (0.2 $\mu\text{g m}^{-3}$) 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 (PM_{2.5} 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 $\mu\text{g m}^{-3}$ in the polluted period, and the ratio of the differential mass (= 127.2 – 100.3 = 26.9 $\mu\text{g m}^{-3}$) to OC (= 9.4 $\mu\text{g m}^{-3}$) 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.
- In Fig. 5, the sum of PM_{0.94} and PM_{2.5-0.94} (e.g., 66.9 $\mu\text{g m}^{-3}$ in polluted case) and the sum of all species listed in Table 3 (= 73.3 $\mu\text{g m}^{-3}$ in polluted case) do not match. Any explanations?