

Review to “Hygroscopicity of Size-Selective Aerosol Particles at Heavily Polluted Urban Atmosphere of Delhi: Impacts of Chloride Aerosol”

The authors present field measurements of size-resolved aerosol hygroscopic growth at 90% RH and bulk aerosol composition of non-refractory PM₁ during wintertime in Delhi, India, and investigate the impacts of chloride on aerosol hygroscopicity and its potential to enhance aerosol-bound liquid water. The paper provides observational evidence of Ammonium Chloride as the major contributor to aerosol hygroscopic growth and liquid water content in Delhi, which highlights the role of Ammonium Chloride in aerosol-water interaction and related haze development. I would recommend publication once the following concerns are addressed.

Major comments:

- 1) The manuscript is a bit long and wordy to me. The authors put too much effort on the overview of the 1.5-month field measurement, and enumerate the ranges of many aerosol properties, e.g., PM₁ mass concentration, chemical composition mass of different species. For example, “BBOA mass concentration varied between 0.0 to 134.7 $\mu\text{g}/\text{m}^3$ ”, I feel sentences like this are not as informative, and should be reduced as much as possible.
- 2) I strongly suggest the authors add a representative case study including major gaseous pollutants, aerosol size distribution, chemical composition, and GF-PDF of 1~2 sizes, to showcase the driving effect of NH₄Cl on aerosol hygroscopicity and see if NH₄Cl exists in all size ranges (i.e., 20~200, from GF-PDF).
- 3) As shown in Fig. 3a, the κ of 20 nm particles look quite scattered to me. For the GF-PDF of 20 nm, I am curious about how many counts of 20 nm particles were sampled for each cycle. As the counting statistics may affect the inversion of GF-PDF. According to a recent study, a total of at least 100 particle counts might be a requirement for reliable GF-PDF inversions (<https://doi.org/10.5194/amt-15-2579-2022>).
- 4) As shown in Fig. 4, the diurnal variation of κ is overshadowed by the hydrophobic mode (e.g., HGF<1.2 for 100 nm). I would suggest the authors to isolate the hygroscopic mode and calculate the corresponding κ . To do so, you could either set a fixed threshold of HGF or fit the bi-modal GF-PDFs and calculate κ using the more hygroscopic mode. By doing this, the authors could probably compare the κ of the hygroscopic mode to pure NH₄Cl.
- 5) Line 351-352: The authors attribute the two-peaked pattern in the GF-PDF to daytime photochemical reactions. If that is the case, why does the HGF decrease at noontime when photochemical activities are supposed to be even stronger.
- 6) Regarding the minor difference in $\kappa_{200\text{nm},90\%}$ for H-BB and H-HOA events, I doubt if that because the two events are not well separated from each other. Do the authors have a general criterion for

separating the three different events, or at least show how much overlapping there is between the two events.

Minor comments:

- 1) Line 101: Use subscript in “(NH₄)₂SO₄”.
- 2) Line 159: Wrong expression in equation (1).
- 3) Line 236: Full spell “MPSS” where it is mentioned for the first time.
- 4) Line 239-241: The value of OA mass concentrations does not seem to be consistent with that reported in Gani et al., (2019).
- 5) Line 300: HGF_{90%_200nm} of “1.12-1.179”, but average to 1.41 ± 0.09 ?
- 6) Line 405: Full spell “ALWC” where it is mentioned for the first time.

Comments for figures:

- 1) Figure 1a and 1b: The lines are overlapped with the shaded boxes, looks like in-continuous data. Describe the shaded boxes of different colors in the caption.
- 2) Figure 2o: The y-axis label is blocked.
- 3) Figure 2d: The diurnal WD pattern looks quite different from the wind rose plot (i.e., Figure S6). The latter suggested a negligible fraction of southerly wind.
- 4) Figure 3: line 332, is Fig. 3d for 150 nm?
- 5) Figure S5: Use a consistent unit, ppb or $\mu\text{g m}^{-3}$.
- 6) Figure 8: Add a legend for the pie chart here.