Response to the Referee (RC) 1

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., PM_1 mass concentration, chemical composition mass of different species. For example, "BBOA mass concentration varied between 0.0 to 134.7 $\mu g/m^3$", I feel sentences like this are not as informative, and should be reduced as much as possible.

Response:

Authors thank the reviewer for highlighting this point. We have removed the sentences which were not much informative. Please see the changes in the revised manuscript (MS).

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 NH_4Cl on aerosol hygroscopicity and see if NH_4Cl exists in all size ranges (i.e., 20~200, from GF-PDF).

Response:

Authors thank the reviewer for the constructive comments. Your suggestion looks very legitimate.

However, we have specific limitations concerning aerosol chemical composition. ACSM gives only bulk chemical composition without size-resolved composition. Therefore, we chose only the most suitable 200 nm aerosol particle to look at the driving effect of NH_4Cl on aerosol hygroscopicity. We have already discussed all sizes of GF-PDF and hygroscopicity in case studies like H-BB, H-Cl, H-HOA, and Clean periods. However, we think making additional case studies to incorporate your suggestion seems too repetitive here. Therefore, instead, we described major gaseous pollutants and aerosol size distribution in addition to H-BB, H-Cl,

H-HOA, and Clean periods in the revised manuscript in such a way that you described in your comment.

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).

Response:

Authors thank the reviewer for mentioning the importance of counting statistics requirement for reliable GF-PDF inversions.

We have gone through the above-mentioned article and checked the counting statistics and GF-PDF of 20 nm. We found that the 20 nm particle counts are less than the recommended statistics from recent article for low aerosol loading times. Authors already followed various recommended filtering processes for good scans, as mentioned in the section, and discarded significant scans. However, authors think that for region like Delhi using HTDMA instruments it will be well justified to have a slightly lesser count.

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 NH_4Cl.

Response:

Thank you for your constructive comments. Your suggestion looks very legitimate.

We calculated the κ for HGF>1.2 per the reviewer's suggestion. However, κ (0.26 ± 0.03: HGF>1.2) was not comparable to the 0.93 hygroscopicity of pure NH_4Cl. In addition, assuming only inorganic salts contribute to HGF>1.2, the calculated hygroscopicity was found to be 0.52 ± 0.10, higher than the observed value of 0.26 ± 0.03. It could be due to the secondary organic aerosol significantly contributing to the HGF>1.2. As we mentioned earlier, we could not calculate hygroscopicity for HGF>1.2 due to the instrument limitation.

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.

Response:

Thanks for the comment.

We agree with the reviewer. Therefore, the primary peak (HGF < 1.2) shifted towards the higher due to the intense daytime photooxidation, as shown in Fig. 5. However, at the same time, lower inorganic contribution, especially NH_4Cl, and the secondary peak (HGF>1.2) shifted towards the lower HGF side, potentially responsible for the lower daytime HGF.

6) Regarding the minor difference in $\kappa_{200nm\_90\%}$ 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.

Response:

Thank you for your comment.

H-BB and H-HOA were separated according to significant BBOA and HOA concentration peaks, respectively. We feel that there is no overlapping of the events as these events do not subsequently happened, as mentioned in Fig. 2. Previous studies (Chakraborty et al., 2014 and Mandariya et al., 2019) in the IGP have shown that BBOA and HOA are difficult to distinguish as they both have significant mixed signatures (mz43, 55, 57, and 60) in their mass spectra. ACSM has limitations concerning OA mass spectra as it proved only bulk m/z without high-resolution m/z fragmentation mass spectra. It could be the reason for the possible mixing of H-HOA and H-BB events. However, as mentioned in the supplementary, we followed standard protocol to separate the HOA and BBOA sources. Therefore, there is a non-significant ($p>0.05$) difference in hygroscopicity, possibly due to the relative changes in primary, secondary OA, and inorganic species. In the H-HOA events, the negative effect of a significantly higher fractional (41%) contribution of HOA to OA possibly balances with a positive impact of a 7% increment in secondary OA relative to H-BB.

Chakraborty, A., Bhattu, D., Gupta, T., Tripathi, S. N. and Canagaratna, M. R.: Real-time measurements of ambient aerosols in a polluted Indian city: Sources, characteristics, and processing of organic aerosols during foggy and nonfoggy periods, J. Geophys. Res., 120(17), 9006–9019, doi:10.1002/2015JD023419, 2015.

Mandariya, A. K., Gupta, T. and Tripathi, S. N.: Effect of aqueous-phase processing on the formation and evolution of organic aerosol (OA) under different stages of fog life cycles, Atmos. Environ., 206(November 2018), 60–71, doi:10.1016/j.atmosenv.2019.02.047, 2019.

Minor comments:

1) Line 101: Use subscript in “(NH4)2SO4”.

Response:

Thanks for pointing it out. As suggested, this comment has been addressed in the revised manuscript.

(Line 99-101) "The humidity sensors positioned in the second DMA were calibrated automatically with 100 nm ammonium sulfate ($(NH_4)_2SO_4$) particles every 30 min at 90% RH to analyze the stability at high RH."

2) Line 159: Wrong expression in equation (1).

Response: Thanks for pointing it out. The equation has been corrected in the revised manuscript.

(Line 152)

$$\kappa_{H-TDMA\_90\%} = (HGF\_90\%^3 - 1)\left[\frac{1}{RH}\, exp\left(\frac{4\sigma M_w}{RT\rho_w D_o HGF_{90}\%}\right) - 1\right] \quad (1)$$

3) Line 236: Full spell "MPSS" where it is mentioned for the first time.

Response:

Thanks. As suggested, this comment has been addressed in the revised manuscript.

(Line 83-88 and 119-121) "Real-time atmospheric aerosol measurements were conducted simultaneously using Hygroscopic-Tandem Differential Mobility Analyzer (H-TDMA), Mobility Particle Size Spectrometer (MPSS), and Aerodyne Aerosol Chemical Speciation Monitor (ACSM, Aerodyne Research, Billerica MA) during winter (1st February 2020 to 16th March 2020) at the Indian Institute of Technology (IIT) Delhi in Block 5, at the height of nearly 15 m as shown in Fig. 1. The lab-2 is situated at the height of 15 m above the ground level and lab-1 is 50 m apart from lab-1."

4) Line 239-241: The value of OA mass concentrations does not seem to be consistent with that reported in Gani et al., (2019).

Response:

Thank you for your constructive comments. As suggested, we modified the statement for better explanation and this comment has incorporated in the revised manuscript.

(Line 261-265) "The OA ranged between 1 and 293 (46.5 ± 39.6) µg/m^3 with the predominant fraction of PM_1, consistent with the range of 53.3 to 166 (112) µg/m^3 observed during winter (December-February) at the present site (Gani et al., 2019). However, lower average OA

concentration could be explained by the measuring period of February-March, as aerosol loading starts decreasing in February after reaching its peak in December-January (Gupta and Mandariya, 2013).”

5) Line 300: HGF90%_200nm of “1.12-1.179”, but average to 1.41 ± 0.09?

Response:

Thanks for pointing it out. We realize that it is typo mistake. This sentence has now been corrected in the revised manuscript.

(Line 318-322) “The hygroscopic growth factors of 20 ($HGF_{90\%\_20nm}$), 50 ($HGF_{90\%\_50nm}$), 100 ($HGF_{90\%\_100nm}$), 150 ($HGF_{90\%\_150nm}$), and 200 nm ($HGF_{90\%\_200nm}$) size particles varied between 1.00-1.41, 1.05-1.39, 1.11-1.49, 1.12-1.63, and 1.12-1.79 with an average of 1.14 ± 0.09 (average ± standard deviation), 1.16 ± 0.06, 1.27 ± 0.07, 1.35 ± 0.09, and 1.41 ± 0.09, respectively.”

6) Line 405: Full spell “ALWC” where it is mentioned for the first time.

Response:

Thanks for pointing it out. This sentence has now been modified accordingly in the revised manuscript.

(Line 425-427) “Further, ammonium chloride has a more significant water uptake potential (Chen et al., 2022; Zhao et al., 2020), which can be justified by the solid correlation of aerosol liquid water content (ALWC) with a mass fraction of ACl in PM_1 as shown in Fig. 6(b).”

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.

Response:

Authors thank the reviewer for pointing it out. The Fig. 2 now has been corrected in the revised manuscript.

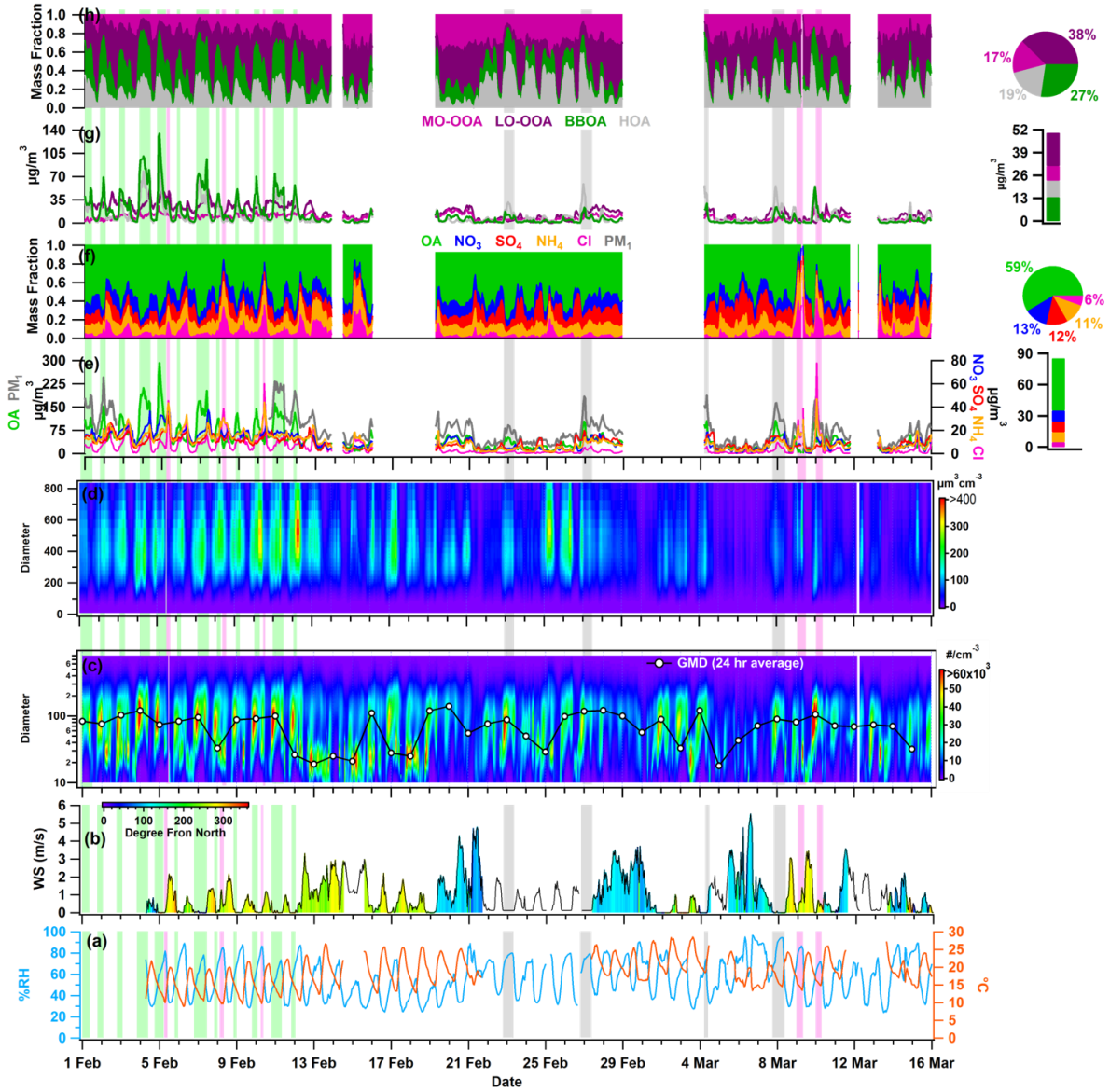

Figure 2: Temporal variability of ambient (a) relative humidity (RH), temperature (T), (b) wind speed (WS), wind direction (WD), (c) particle number-size distribution (PNSD), 24-average geometric mean diameter (GMD), (d) particle volume-size distribution (PVSD), (e) particulate matter (PM_1), organic aerosol (OA), nitrate (NO_3), sulfate (SO_4), ammonium (NH_4), chloride (Cl), (f) fractional contribution of OA, NO_3, SO_4, NH_4, and Cl in PM_1, (g) more oxidized-oxygenated OA (MO-OOA), less oxidized-oxygenated OA (LO-OOA), biomass burning OA (BBOA), hydrocarbon like-OA (HOA), and (h) fractional contribution of MO-OOA, LO-OOA, BBOA, and HOA in OA. The pie chart sub-plot represents the overall average contribution of species, and the bar sub-plot represents the overall campaign average value of different species. All other species are represented with specific color coding mentioned in legends. The light green, pink, and grey color shaded vertical line indicates the high-BBOA (H-BB), high-HOA (H-HOA), and high-Cl (H-Cl) events, respectively. The discontinuity in the data points marks the missing data or non-sampling time.

2) Figure 2o: The y-axis label is blocked.

We sincerely thank the reviewer for pointing it out. The plot has been corrected in the revised manuscript.

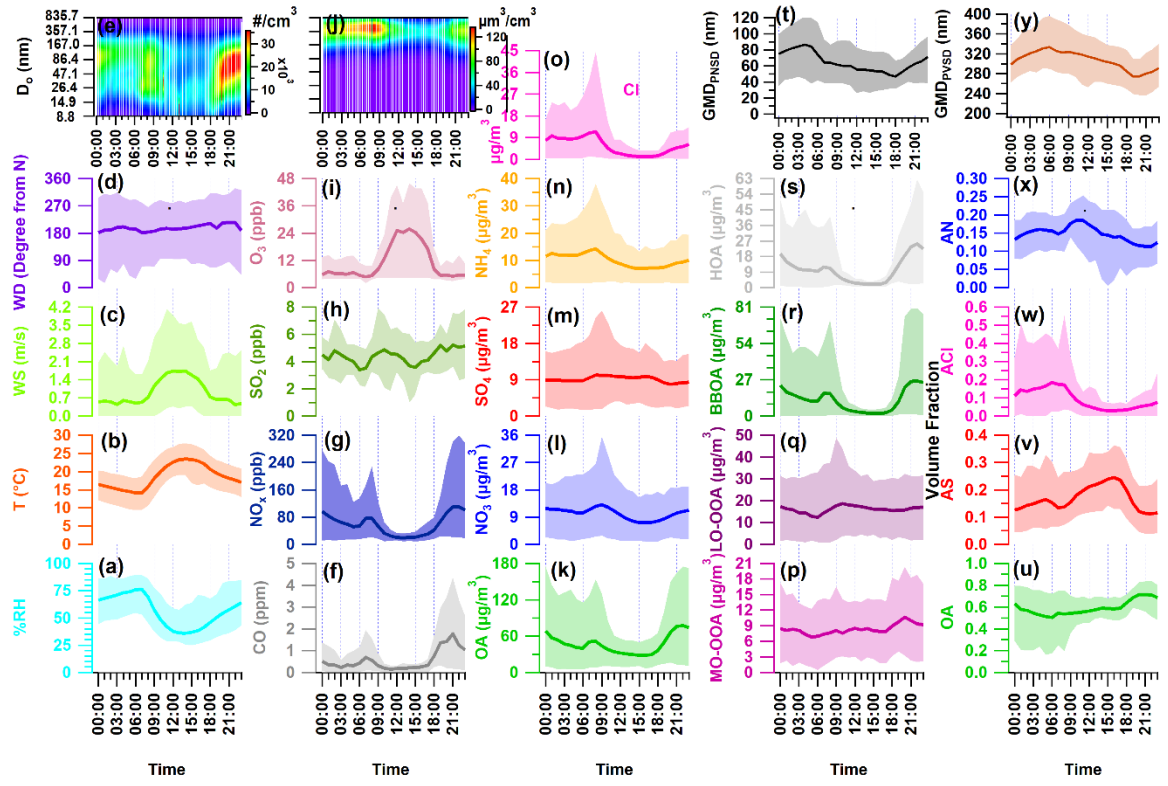

Figure 3: Diurnal variation of ambient meteorological parameters (a) % ambient relative humidity (RH), (b) temperature (T), (c) wind speed (WS), (d) wind direction (WD), and (e) particle number size distribution (PNSD), mass concentration of ambient trace gases (f) carbon mono-oxide (CO), (g) nitrogen oxides (NOx), (h) sulfur dioxide (SO_2), and (i) ozone (O_3), (j) particle volume size distribution (PVSD), mass concentration of aerosol constituents (k) organic aerosol (OA), (l) nitrate (NO_3), (m) sulfate (SO4), (n) ammonia (NH_4), and (o) chloride (Cl), mass concentration of organic aerosol species (p) more oxidized-oxygenated OA (MO-OOA), (q) less oxidized-oxygenated OA (LO-OOA), (r) biomass burning OA (BBOA), and (s) hydrocarbon like-OA (HOA), (t) geometric mean diameter of particle number size distribution (GMDPNSD) and volume fractional contribution of (u) organic aerosol (OA), (v) ammonium sulfate (AS), (w) ammonium chloride (ACl), and (x) ammonium nitrate (AN) in PM_1, and (y) geometric mean diameter of particle volume size distribution (GMD_{PVSD}). Upper and lower boundary of shaded area represents the 95th and 5th percentile values of respective species.

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.

Response:

The authors thank the reviewer for the comment. The wind directions predominantly varied from 20 to 330 degrees from the north; therefore, Fig. 2d shows the statistically average WD of nearly 180 degrees. However, in a real scenario, southerly WD has a negligible fraction.

4) Figure 3: line 332, is Fig. 3d for 150 nm?

Response:

Thanks for pointing it out. As suggested, this suggestion has been incorporated in the revised highlights.

(Figure 4d: Line 352-353) "(d) 150 nm ($\kappa_{150nm\_90\%}$), and (e) 200 nm ($\kappa_{200nm\_90\%}$)."

5) Figure S5: Use a consistent unit, ppb or µg m-3.

Response:

The authors thank the reviewer for the comment. Fig. S5 has been revised in the revised supplementary.

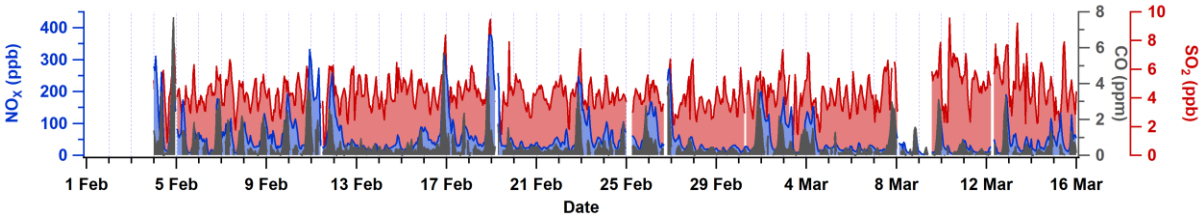

Figure S5: Temporal variability in atmospheric NO_x, CO, and SO_2 gases concentrations.

6) Figure 8: Add a legend for the pie chart here.

Response:

Thanks. We modified the plot and add color legends in the plot to accommodate your comment.

(Line 603-609)

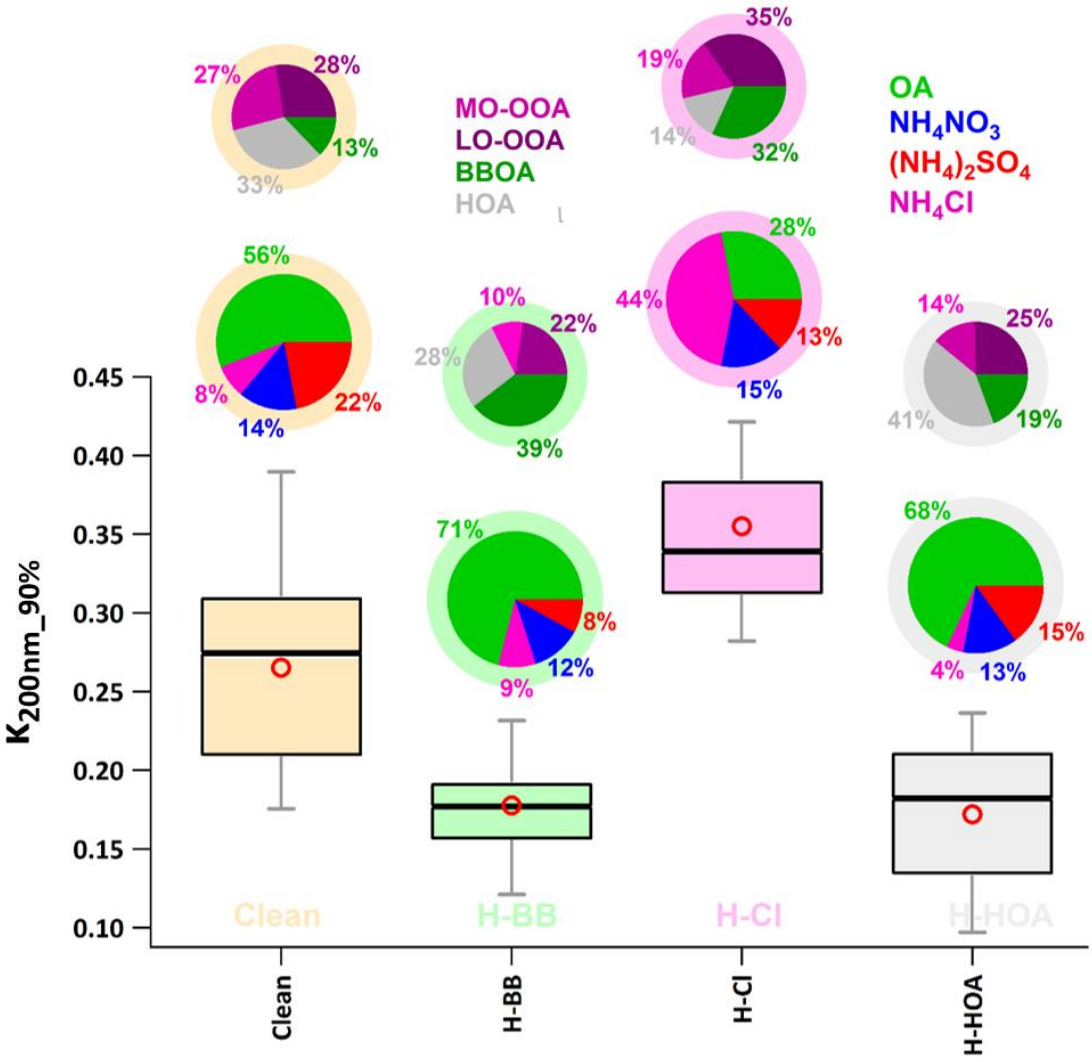

Figure 9: Box plot showing variation in H-TDMA measured hygroscopic parameter of 200 nm size particles $\kappa_{H\text{-}TDMA}$ ($\kappa_{200nm\_90\%}$) in high biomass burning (H-BB), high-chloride (H-Cl), and high-hydrocarbon like organic aerosol (H-HOA) events. Different colors represent respective events in the plot. A bigger pie chart represents the overall average volume fractional contribution of various aerosol species indicated by color-coding. In addition, minor pie charts described the event average mass fractional contribution of different OA species in OA. Diffused ring color of the pie chart displays the respective event.

Response to the Referee (RC) 2

The manuscript "Measurement report: Hygroscopicity of Size-Selective Aerosol Particles at Heavily Polluted Urban Atmosphere of Delhi: Impacts of Chloride Aerosol" revealed the wintertime chloride emission in the Delhi region governing the enhancement of aerosol hygroscopicity and aerosol-bound liquid water that trigger Delhi's fog episodes. The manuscript is written well and within the interest of the scientific communities. However, there are many gaps in the quality of presentation and lack of clarity in the manuscript.

Major Comments

- The author did not present the schematic of the experimental design. Therefore, it is difficult to understand the different instruments used in the study.

Response:

Thank you for your constructive comments. Your suggestion seems very legitimate. As suggested, this comment has been addressed in the revised manuscript. The schematic diagram of sampling instruments was added in the revised manuscript.

(Line 83-88 and 119-121) "Real-time atmospheric aerosol measurements were conducted simultaneously using Hygroscopic-Tandem Differential Mobility Analyzer (H-TDMA), Mobility Particle Size Spectrometer (MPSS), and Aerodyne Aerosol Chemical Speciation Monitor (ACSM, Aerodyne Research, Billerica MA) during winter (1st February 2020 to 16th March 2020) at the Indian Institute of Technology (IIT) Delhi in Block 5, at the height of nearly 15 m as shown in Fig. 1. The lab-2 is situated at the height of 15 m above the ground level and lab-1 is 50 m apart from lab-1."

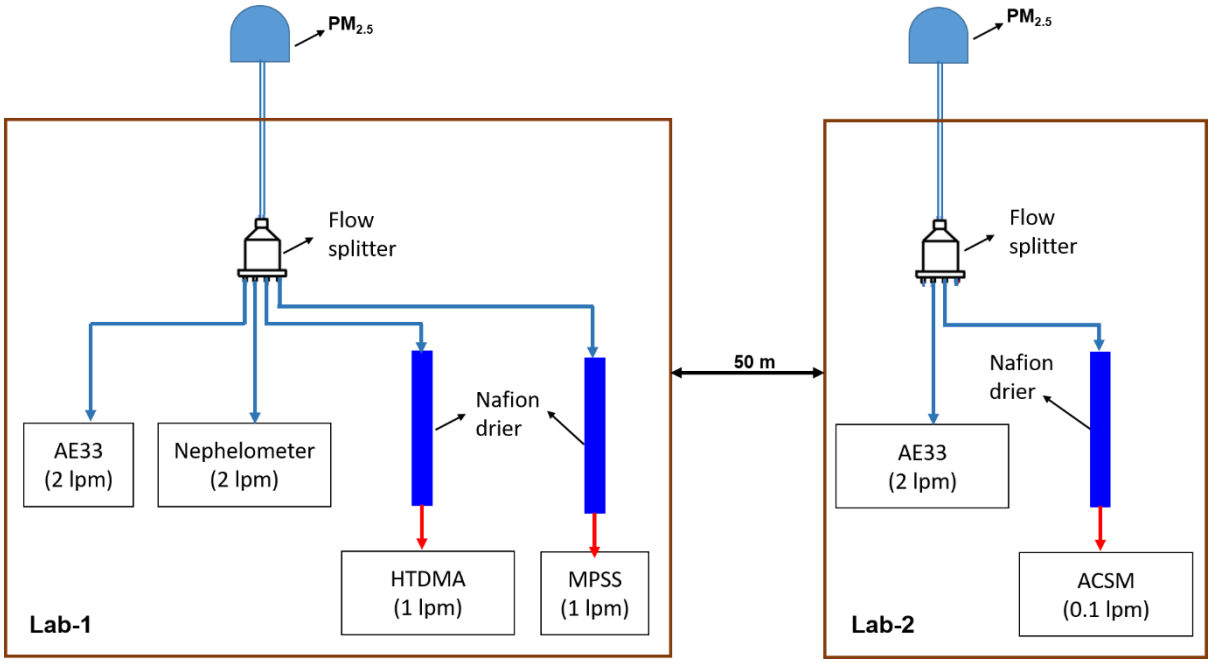

Figure 1: Schematic diagram of the inlet systems for aerosol sampling instruments. The blue and red sampling lines indicate the ambient air and dehumidified (RH<25%) ambient air, respectively.

- There is a lack of clarity on the classification of different events e.g., H-BB, H-HOA, H-Cl and clean. For example, how was the event classification made based on the aerosol chemical compositions? There is missing information about these events in figure 1 caption. It is recommended that the author should add a table to the text to discuss the event classification explicitly.

Response:

We sincerely thank the reviewer for pointing it out. Instead of a table, we add statements explaining these events' classification.

(Line 170-175) "Furthermore, based on the significant mass concentration peaks of BBOA, HOA, and Cl in the temporal variation, respectively, three different events were characterized: 1) High-residential or biomass burning (H-BB), 2) High-hydrocarbon-like OA (H-HOA), and 3) High-chloride (H-Cl) period. In addition, the "Clean Period" was defined where PM_1 loading was less than 25 percentiles (≤ 38.7 μgm^{-3}) of the sampling period. The starting and end time of the event was defined by the starting the increment in the concentration and reaching the starting value while the concentration decreased."

- The mathematical equations used in the text should be cross verified.

Response:

Thank you for your correction. We modified the mathematical equations used in the text in the revised manuscript

(Line 182-199)

Case-1 $\boldsymbol{R_{SO_4}(NH_4\ to\ SO_4)\ \leq 1}$

$$SA = 98.0795 \times max(0, (n_S - n_A))$$

$$ABS = 115.11 \times n_A$$

$$AS = 0$$

$$AN = 0$$

$$ACl = 0$$

Case-2 $\boldsymbol{1 < R_{SO_4} < 2}$

$$SA = 0$$

$$ABS = 115.11 \times ((2 \times n_S) - n_A)$$

$$AS = 132.1405 \times (n_S - n_A)$$

$$AN = 0$$

$$ACl = 0$$

Case-3 $\boldsymbol{R_{SO_4} \geq 2}$

$$SA = 0$$

$$ABS = 0$$

$$AS = 132.1405 \times n_S$$

$$AN = \left(min\left(\left(n_A - \left(\frac{ABS}{115.11} \right) - \left(\frac{(2 \times AS)}{132.1405} \right) \right), n_N \right) \right) \times 80.0434$$

$$ACl = \left(min\left(n_C, \left(n_A - \left(\frac{ABS}{115.11} \right) - \left(\frac{2 \times AS}{132.1405} \right) - \left(\frac{AN}{80.0434} \right) \right) \right) \right) \times 53.54$$

Minor comments

Page 1 and Line 27: Expand HTDMA

Response:

As suggested, this comments have been incorporated in the revised manuscript.

(Line 18-22) "In this study, we present the measurement results of bulk aerosol composition of non-refractory PM_1 from ACSM and size-resolved (Nucleation, Aitken, and Accumulated mode particles) hygroscopic growth factor and associated hygroscopicity parameter at 90% relative humidity (RH) measured using H-TDMA (Hygroscopic-Tandem Differential Mobility Analyser) at Delhi Aerosol Supersite (DAS) for the first time."

Page 1 and Line 33: Expand OA

Response:

As suggested, this suggestion has been incorporated in the revised manuscript.

(Line 31-32) "Additionally, the high chloride content in aerosols appears to counteract the negative effects of high organic aerosol (OA) levels on cloud condensation nuclei (CCN) activity."

Page 3 and Line 67: Expand IGP

Response:

Thank you for your correction. We modified the sentence in the revised manuscript.

(Line 64-65) "In past decades, fast economic growth and industrialization in the Indo Gangetic Plain (IGP) led to severe air quality during wintertime (Wester et al., 2019)."

Page 5 and Line 137: This is a repeated sentence.

Response:

The text '*The humidity sensor of DMA2 was automatically calibrated with 100 nm ammonium sulfate particles after each scan cycle.*' was removed as suggested.

Page 6 and Line 159: The equation is not correct.

Response: We sincerely thank the reviewer for pointing it out. As suggested, the equation (1) has been corrected in the revised manuscript.

(Line 152)

$$\kappa_{H-TDMA\_90\%} = (HGF\_90\%^3 - 1)\left[\frac{1}{RH}\, exp\left(\frac{4\sigma M_w}{RT\rho_w D_o HGF_{90}\%}\right) - 1\right] \quad (1)$$

Page 6 and Line 169: It is not clear the modified ion pairing scheme: what is the difference between SA and AS

Response: Thank you for your correction. We modified the sentence in the revised manuscript.

(Line 178-181) "However, Gysel et al. (2007) did not include NH_4Cl in their ion-pairing scheme; therefore, we elaborated this scheme and made some modifications in this scheme to include ammonium chloride (ACl) in the calculation. Hence, our modified ion-pairing scheme includes NH_4Cl (ACl), NH_4NO_3 (AN), $(NH_4)_2SO_4$ (AS), NH_4HSO_4 (ABS), and H_2SO_4 (SA) are shown below:"

Page 8 and Line 195: The author should discuss the source of the gas and meteorological data. At what height the met parameters were measured?

Response:

We sincerely thank the reviewer for pointing it out. As suggested, we have added the corresponding explanation sources of gas and meteorological data text in the revised manuscript.

(Line 123-128) "2.2 Meteorological and Gas Data

The gas data was taken from the location site R.K Puram -DPCC, a continuous ambient air quality monitoring station controlled by the central control room for air quality management (Delhi-NCR). The gas data were downloaded from the CPCB website (https://app.cpcbccr.com/ccr/#/caaqm-dashboard/caaqm-landing/data). R.K. Puram is located 3.5 km northwest of IIT Delhi. The wind speed (WS), wind direction (WD), temperature (T), and relative humidity (RH) were continuously measured using an automatic weather station (Watch Dog 2000 series). The weather station is mounted over the top of the 9th-floor building of the IITD."

Page 8 and Line 196: The author talked about PNSD. It is not clear how they measured it? Is it from the HTDMA or additionally a size spectrometer was used. A detailed schematic experimental design is needed.

Response:

Authors sincerely thank the reviewer. We think that what we discussed in the first comment's response can also be the response to this comment. However, in addition, we add statements explaining the PNSD and its measurement in the revised manuscript.

(Line 109-110) "Particle number size distributions (PNSDs) and particle volume-size distributions (PVSDs) were measured using a Mobility Particle Size Spectrometer (MPSS (TROPOS type)).

(Line 158-161) 2.3.2 MPSS

MPSS measures electrical mobility distribution, which is then converted to PNSD in the 8 to ~800 nm mobility diameter range by applying an inversion algorithm to correct for multiple charged aerosol particles (Wiedensohler, 1988; Pfieffer et al., 2014) and diffusional losses (Wiedensohler et al., 2012; 2018)."

Page 8 and Line 205-207: Reference is missing.

Response: We sincerely thank the reviewer for pointing it out. As suggested, we have added a reference in the text in the revised manuscript.

(Line 233-234) "This comparatively higher ambient temperature and O_3 peak concentration during noontime (Fig. 3i) indicate the daytime photo-oxidation process (Nelson et al., 2023)."

Page 8 and Line 213: It is not clear how the intensity of biomass burning activities was determined.

Response:

Authors sincerely thank the reviewer for the comment. We did not determine the intensity of the biomass-burning activities. The ambient trace gases NOx and CO are the markers of burning activities. Their concentration found a good correlation with the peak concentration of organic aerosol. Therefore, we imply that the peak in the concentration of CO and NOx indicates the local burning activities.

Page 9 and Line 220: Author should explain the nighttime peak of SO_2.

Response:

The authors thank the reviewer for the comment.

(Line 244-246) "In contrast, SO_2 follows a different trend, with dynamic variations ranging from 0.46 to 9.55 ppb (4.41 ± 1.20) and showing peaks in the morning (09:00-12:00 hours) and at midnight (21:00-02:00 hours) associated with the local industrial stack emissions."

Page 9 and Line 235: It is not clear about MPSS. Is it a separate instrument associated with the experimental design? If so, why was the MPSS data not presented in this study?

Response:

We sincerely thank the reviewer. We think that what we discussed in the first major comment's and 8th minor comment's response can also be the response to this comment. However, in addition, The MPSS time series data already have been shown in the manuscript in Fig. 1(c) in terms of PNSD.

Page 9 and Line 240: … average mass concentration 46.5 ±39.6 ug/m^3 consistent with 112 ug/m^3…. This is not clear.

Response:

Thank you for your constructive comments. We modified the statement for better explanation.

(Line 261-265) "The OA ranged between 1 and 293 (46.5 ± 39.6) µg/m^3 with the predominant fraction of PM_1, consistent with the range of 53.3 to 166 (112) µg/m^3 observed during winter

(December-February) at the present site (Gani et al., 2019). However, lower average OA concentration could be explained by the measuring period of February-March, as aerosol loading starts decreasing in February after reaching its peak in December-January (Gupta and Mandariya, 2013).”

Page 12: The y- axis of diel Cl plot is not clear.

Response:

We sincerely thank the reviewer for pointing it out. The plot has been corrected in the revised manuscript.

(Fig.3o: Line 303-314)

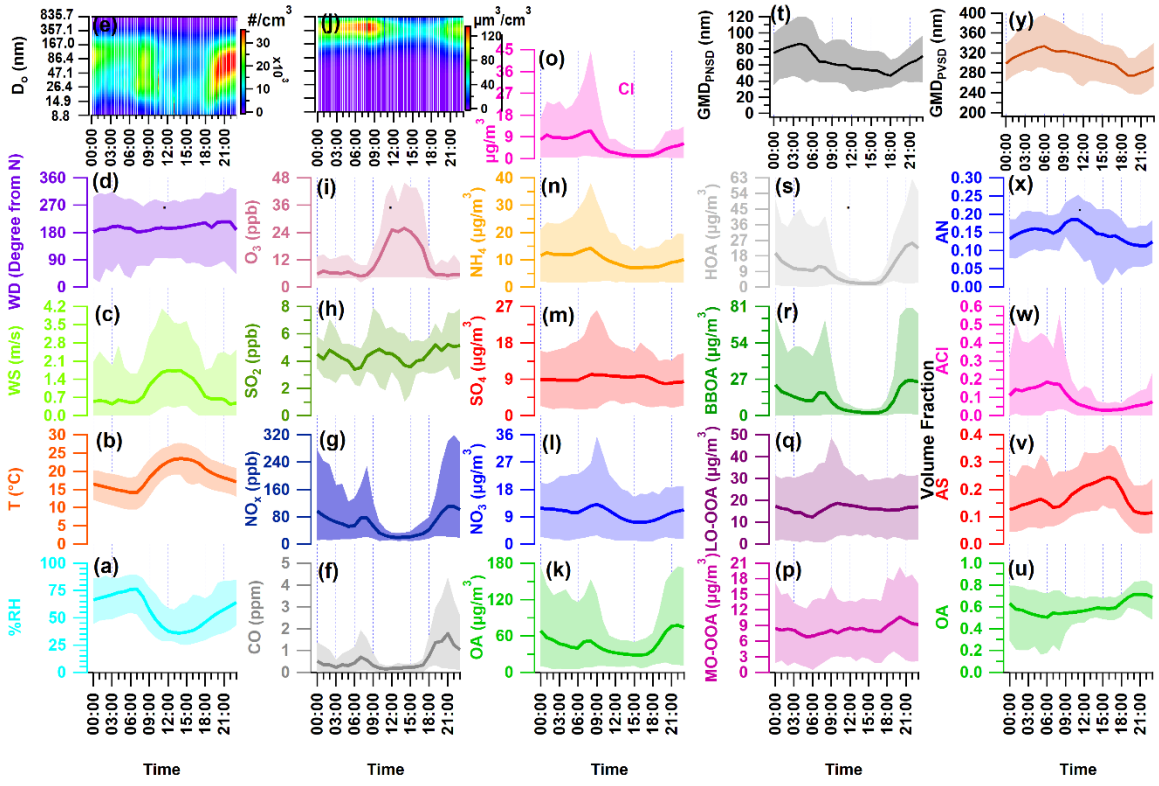

Figure 3: Diurnal variation of ambient meteorological parameters (a) % ambient relative humidity (RH), (b) temperature (T), (c) wind speed (WS), (d) wind direction (WD), and (e) particle number size distribution (PNSD), mass concentration of ambient trace gases (f) carbon mono-oxide (CO), (g) nitrogen oxides (NOx), (h) sulfur dioxide (SO_2), and (i) ozone (O_3), (j) particle volume size distribution (PVSD), mass concentration of aerosol constituents (k) organic aerosol (OA), (l) nitrate (NO_3), (m) sulfate (SO4), (n) ammonia (NH_4), and (o) chloride (Cl), mass concentration of organic aerosol species (p) more oxidized-oxygenated OA (MO-OOA), (q) less oxidized-oxygenated OA (LO-OOA), (r) biomass burning OA (BBOA), and (s) hydrocarbon like-OA (HOA), (t) geometric mean diameter of particle number size distribution (GMDPNSD) and volume fractional contribution of (u) organic aerosol (OA), (v) ammonium sulfate (AS), (w) ammonium chloride (ACl), and (x) ammonium nitrate (AN) in PM_1, and (y) geometric mean diameter of particle volume size distribution (GMD_{PVSD}). Upper and lower boundary of shaded area represents the 95th and 5th percentile values of respective species.

Page 17 and Line No.398: Author should explain why two linear regressions are drawn in the correlation plot (example Fig. 5a).

Response:

The light color regression lines and equations represent the correlation of all data points of $\kappa_{200nm\_90\%}$ with the volume and mass fractional contribution of ACl in PM_1. In contrast, the dark color regression lines and equations indicate the regression line of averaged $\kappa_{200nm\_90\%}$ over the 10% increment of ACl by volume. We add statements in the Figure caption that explain the regression lines.

(Line 438-444)

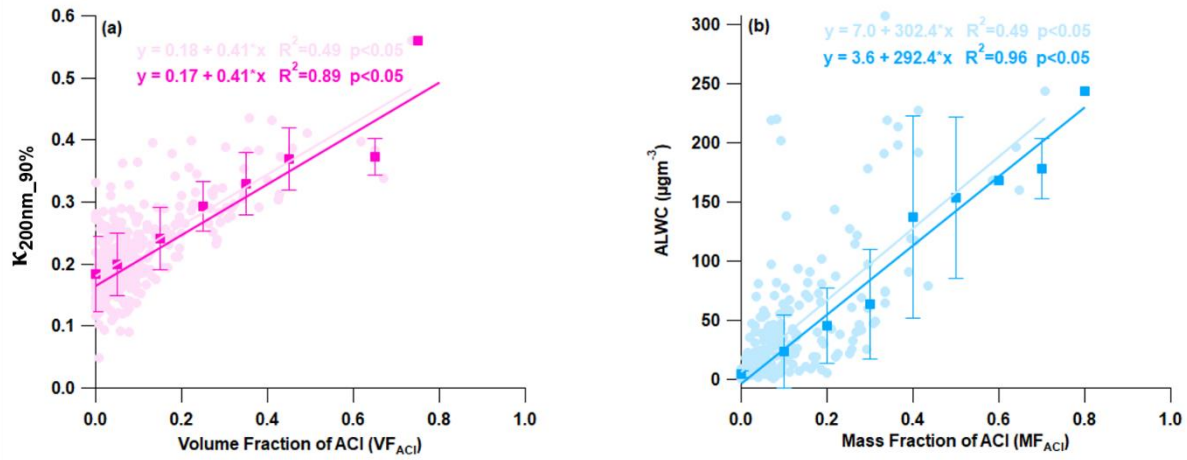

Figure 6: Correlation plot for (a) $\kappa_{200nm\_90\%}$ vs volume fraction of ammonium chloride aerosol (VF_{ACl}) and (b) aerosol liquid water content (ALWC) vs mass fraction of ammonium chloride (MF_{ACl}). The solid circle and square marker represent the individual data points and the average of 10% volume and mass fraction increment of ACl data points, respectively. The light and dark color regression lines and equations indicate the overall and average (10% volume and mass fraction increment) correlation, respectively. The error bars indicate the standard deviation of the data points within the 10% mass and volume fractional bins.

Page 18 and Line No.418: Author should provide the ALWC vs mass fraction of AN and AS in the supplement.

Response:

Thank you for your constructive comments. Your suggestion seems very legitimate. The plot (Fig. S8) has been incorporated in the revised manuscript.

(Supplement, Line 76-82)

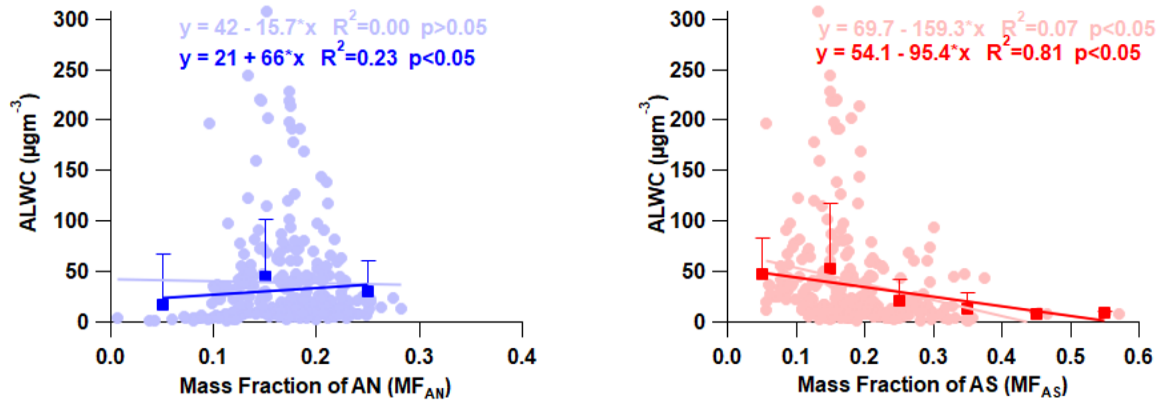

Figure S8: Correlation plot for (a) aerosol liquid water content (ALWC) vs mass fraction of ammonium nitrate (MF_{AN}) and (b) aerosol liquid water content (ALWC) vs mass fraction of ammonium sulfate (MF_{AS}). The solid circle and square marker represent the individual data points and the average of 10% mass fraction increment of data points, respectively. The light and dark color regression lines and equations indicate the overall and average (10% mass fraction increment) correlation, respectively. The positive error bar indicates the standard deviation of the data points within the 10% mass fractional bin.

Page 19 and Line No 434. The author should clearly mention the dates they consider for a relatively clean period.

Response:

Thank you for your legitimate comment. We add statements explaining the clean period's date and duration in the revised manuscript.

(Line 545-546) "The 24th and 25th of February and the 5th, 6th, and 7th of March were marked as Clean events. The night 21 hr to morning 11 hr duration was recorded as the clean duration."

Page 22 and Line No 505-507. Is it 39% of BBOA by mass? Figure 8 is not clear. The color coding should be clarified in the plot.

Response:

Thanks. We modified the plot and add color legends in the plot to accommodate your comment.

(Line 603-609)

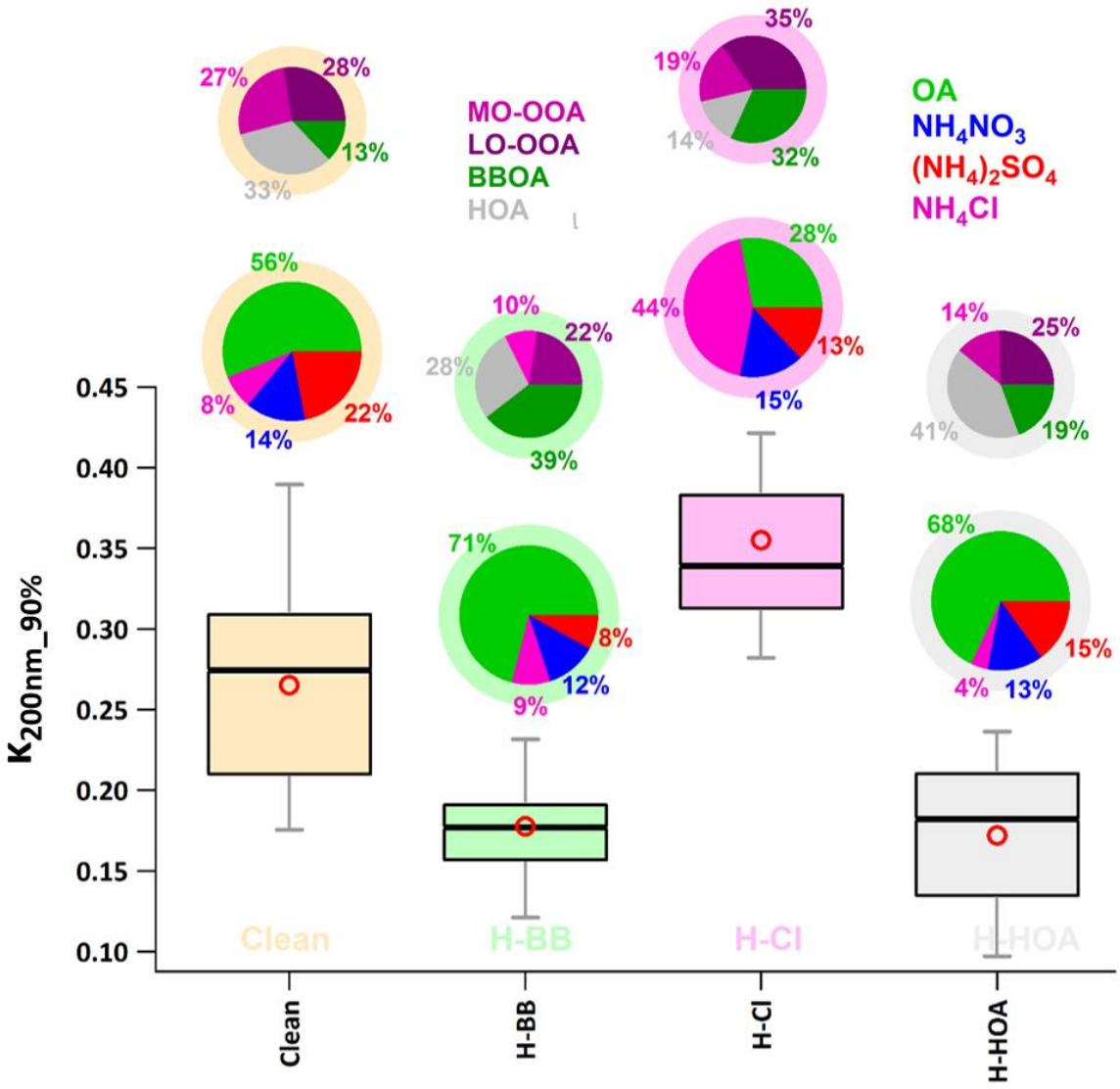

Figure 9: Box plot showing variation in H-TDMA measured hygroscopic parameter of 200 nm size particles $\kappa_{H\text{-}TDMA}$ ($\kappa_{200nm\_90\%}$) in high biomass burning (H-BB), high-chloride (H-Cl), and high-hydrocarbon like organic aerosol (H-HOA) events. Different colors represent respective events in the plot. A bigger pie chart represents the overall average volume fractional contribution of various aerosol species indicated by color-coding. In addition, minor pie charts described the event average mass fractional contribution of different OA species in OA. Diffused ring color of the pie chart displays the respective event.

Page 22 and Line No 505-507. The dates and times of the event should be clarified in the figure 1 caption.

Response:

Thanks. The dates and times of the events have been clarified from the Figure 1 caption. We add statements explaining the clean period's date and duration in the revised manuscript.

(Line 495-497) "High BB events were noted during the initial period (1-12 February) of the field campaign. However, H-BB events were generally captured either during the midnight (01:00 hr) to morning (08:00 hr) or evening (20:00 hr) to midnight (01:00 hr). Although, sometimes, it was continued from evening (21:00 hr) to morning (11:00 hr)."

Page 23 and Line 535: The x-axis label is missing.

Response:

We sincerely thank the reviewer for pointing it out. The plot has been corrected in the revised manuscript.

(Line 569-571)

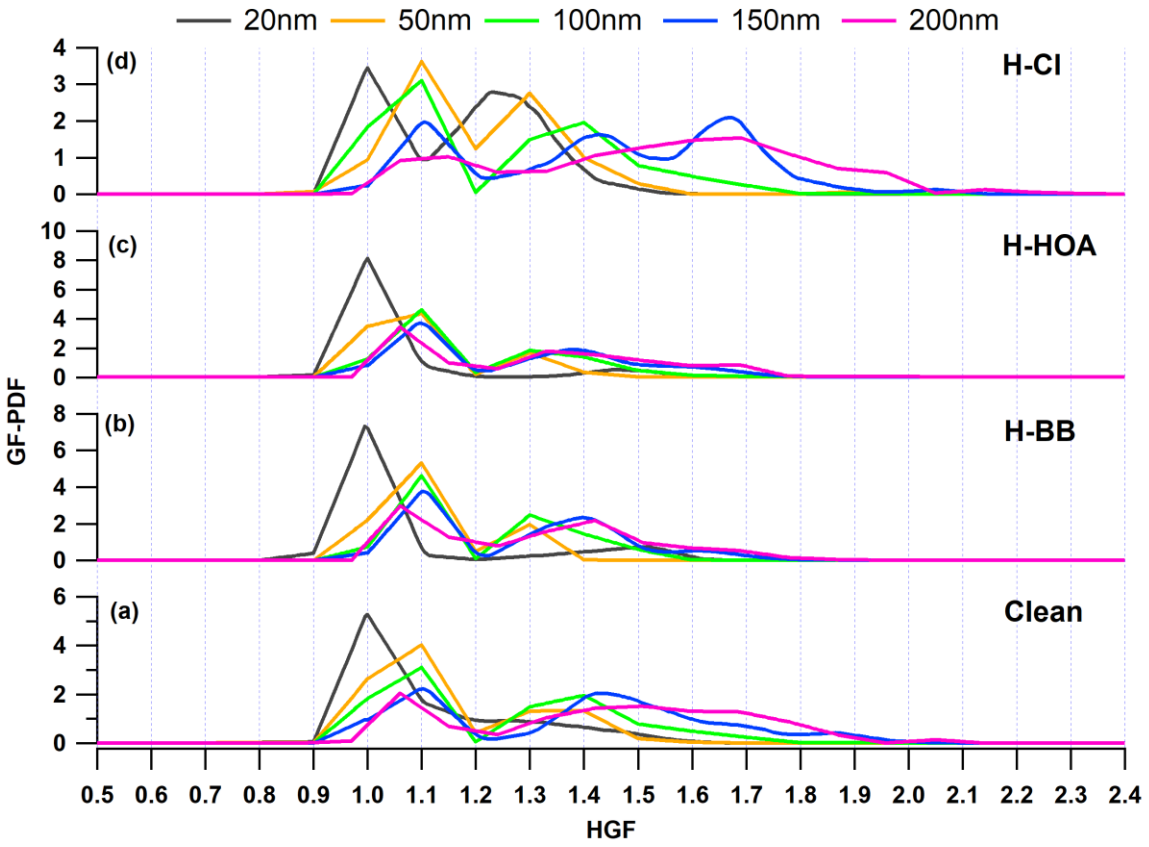

Figure 8: Growth Factor Probability Density Function (GF-PDF) of 20, 50, 100, 150, and 200 nm aerosol particles for the (a) clean, (b) H-BB, (c) H-HOA, and H-Cl periods.

Page 25 and Line 583: However,…time in India…This statement is not true.

Response:

Thanks. We modified the text to justify our previous statement.

(Line 616-618) "However, we reported hygroscopicity of nucleation and Aitken mode particles using HTDMA for the first time in India."

Supplements

Page 2 and Line 23: Author should present the time series data of MPSS during the study period.

Response:

Thanks. The MPSS time series data already have been shown in the manuscript in Fig. 1(c) in terms of PNSD.

Page 7 and Line 80: I don't see any difference in the probability distributions of BBOA, HOA and ACL. The Author should clarify it.

Response:

Thanks. Yes, the probability of potential BBOA, HOA, and ACl sources is similar. Therefore, we conclude that during H-BB events, the receptor site was influenced by air mass from some parts of Uttar Pradesh, Punjab, and Haryana comprising BBOA, HOA, and ACl aerosol.