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."

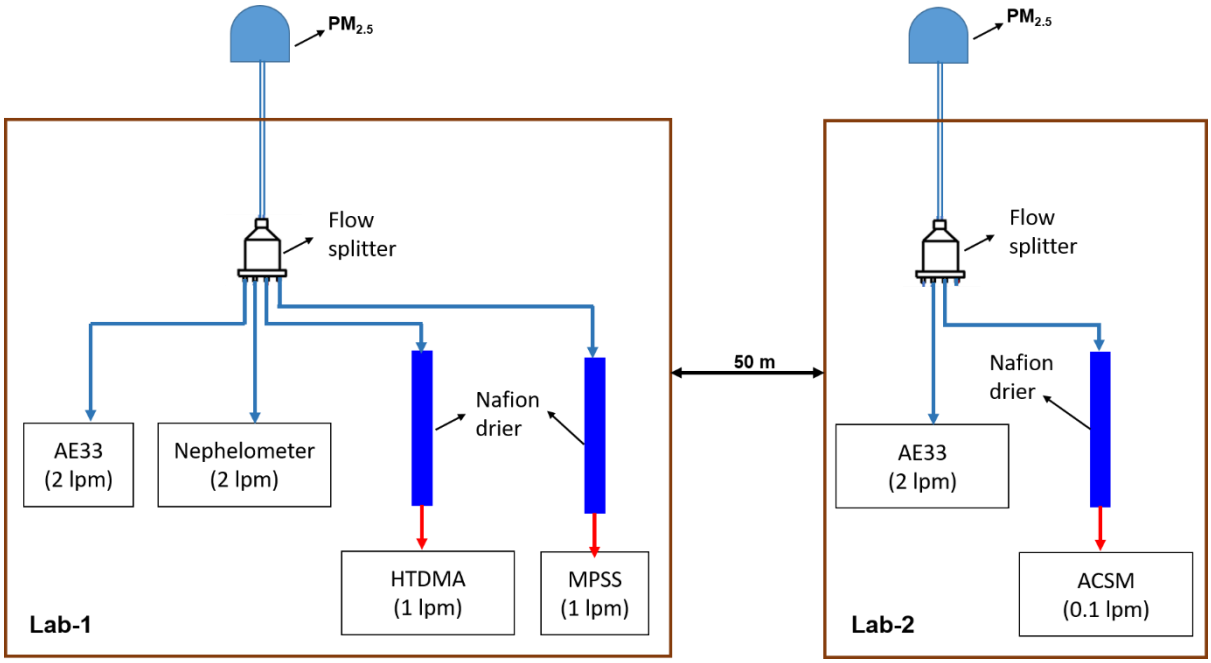

Figure 1: Schematic diagram of the inlet systems for aerosol sampling instruments. The blue and red sampling lines indicate the ambient air and dehumidified (RH<25%) ambient air, respectively.

- 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)

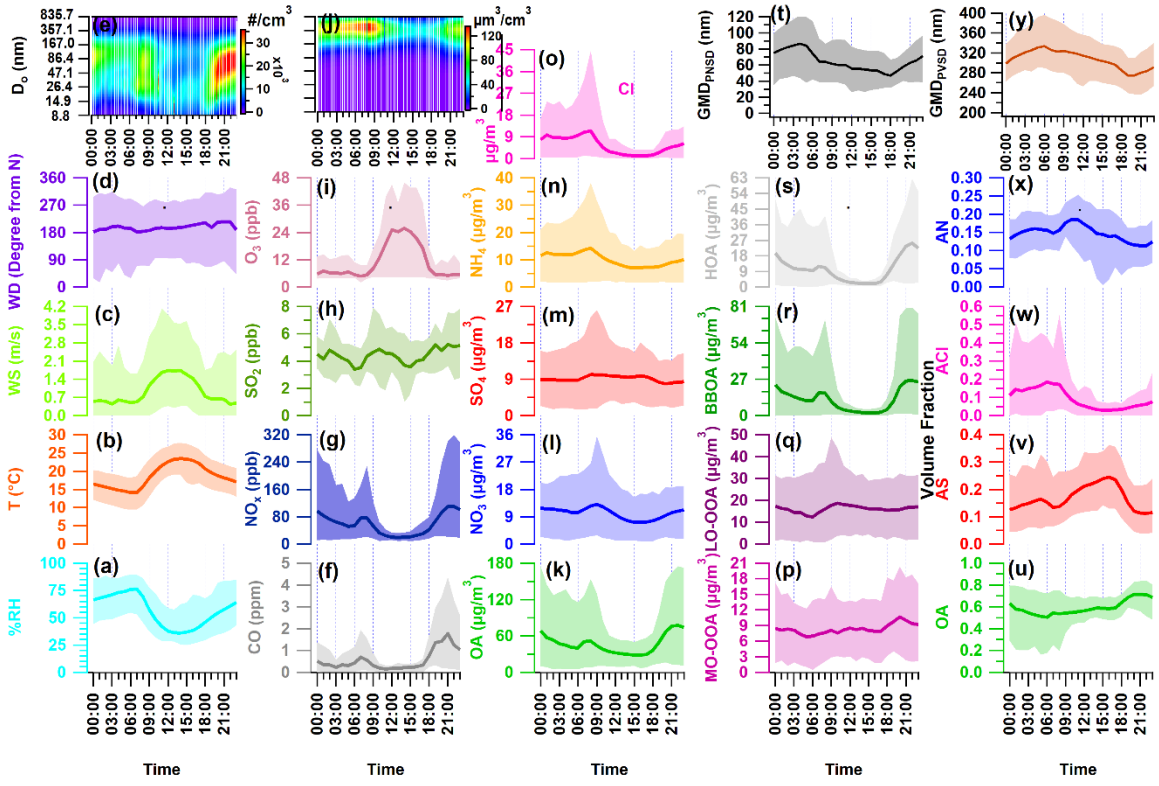

Figure 3: Diurnal variation of ambient meteorological parameters (a) % ambient relative humidity (RH), (b) temperature (T), (c) wind speed (WS), (d) wind direction (WD), and (e) particle number size distribution (PNSD), mass concentration of ambient trace gases (f) carbon mono-oxide (CO), (g) nitrogen 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)

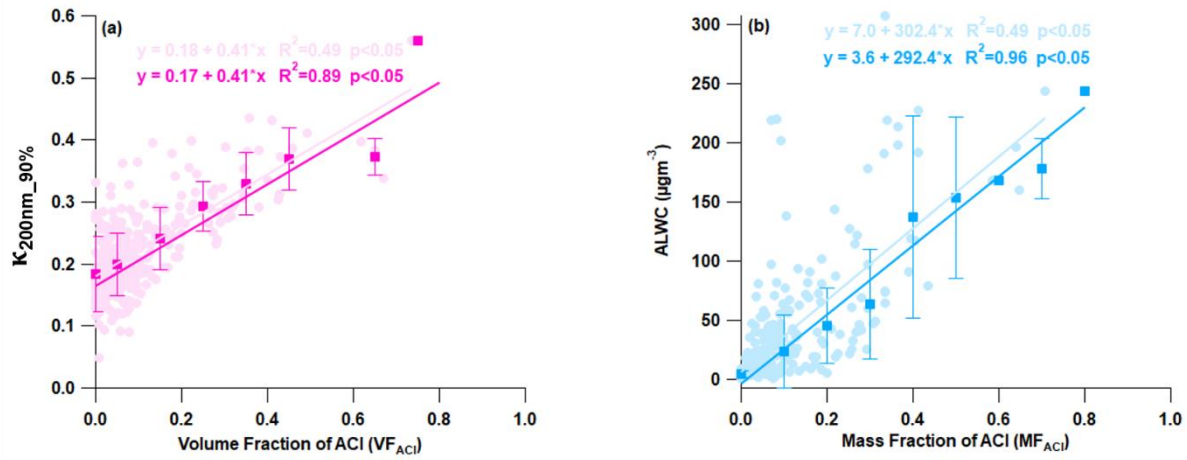

Figure 6: Correlation plot for (a) $\kappa_{200nm\_90\%}$ vs volume fraction of ammonium chloride aerosol (VF_{ACl}) and (b) aerosol liquid water content (ALWC) vs mass fraction of ammonium chloride (MF_{ACl}). The solid circle and square marker represent the individual data points and the average of 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)

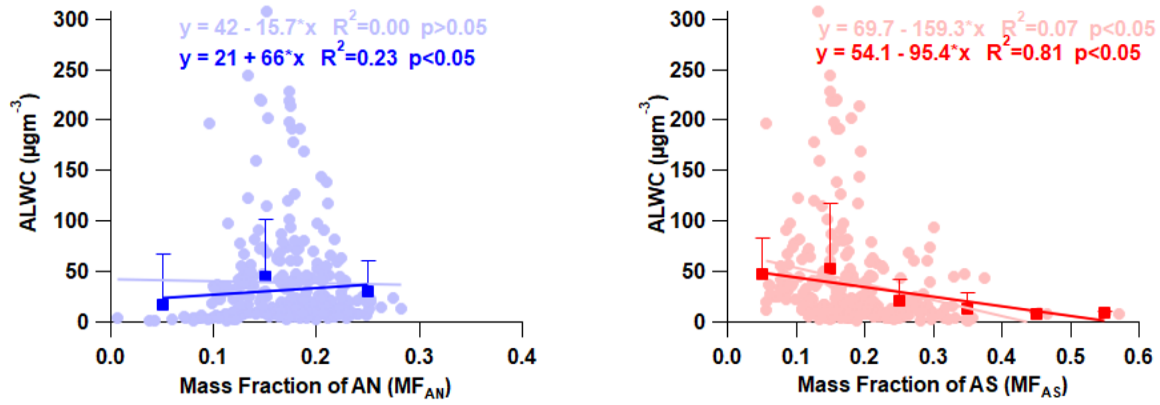

Figure S8: Correlation plot for (a) aerosol liquid water content (ALWC) vs mass fraction of ammonium nitrate (MF_{AN}) and (b) aerosol liquid water content (ALWC) vs mass fraction of ammonium sulfate (MF_{AS}). The solid circle and square marker represent the individual data points and the average of 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)

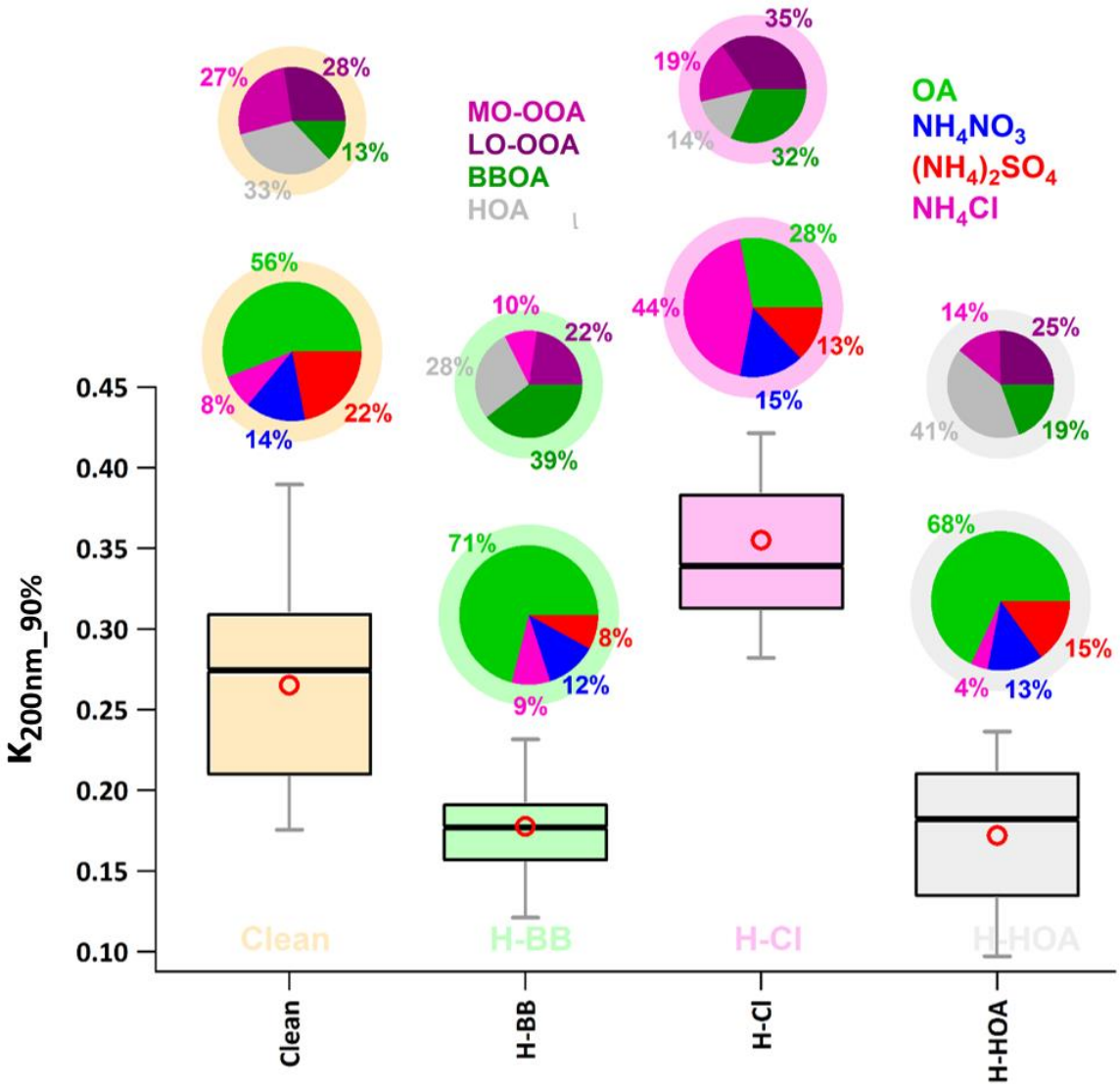

Figure 9: Box plot showing variation in H-TDMA measured hygroscopic parameter of 200 nm size particles $\kappa_{H\text{-}TDMA}$ ($\kappa_{200nm\_90\%}$) in high biomass burning (H-BB), high-chloride (H-Cl), and high-hydrocarbon like organic aerosol (H-HOA) events. Different colors represent respective 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)

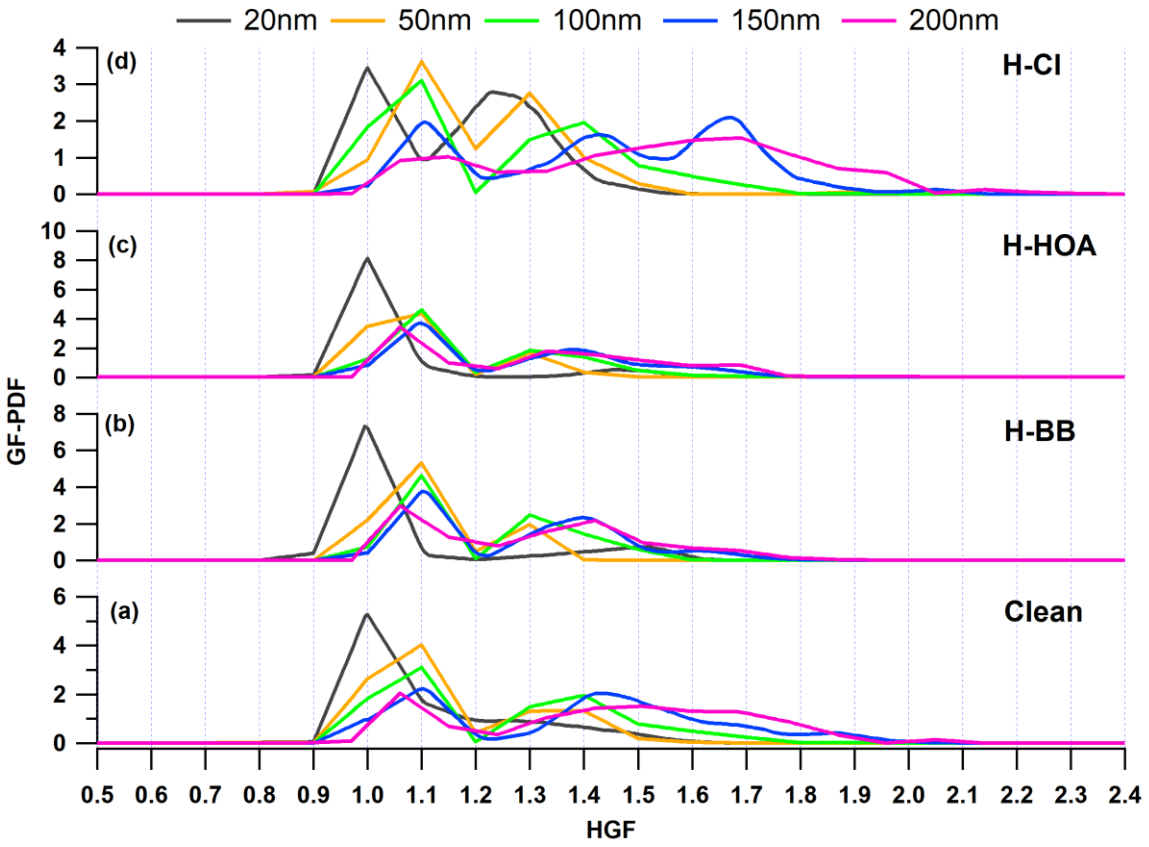

Figure 8: Growth Factor Probability Density Function (GF-PDF) of 20, 50, 100, 150, and 200 nm aerosol particles for the (a) clean, (b) H-BB, (c) H-HOA, and H-Cl periods.

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.