Dear editor, we would like to express our gratitude to you and the two referees for providing thoughtful comments and valuable suggestions that have significantly improved the quality of our manuscript. Below, you will find our point-by-point responses to the individual comments of the two referees and editor.

Response to the Referee (RC) 2

Minor comments:

1. Fig. 2 caption: I thought the pink boxes indicated the high-CL events. Please double check.

Response:

Authors thank the reviewer for highlighting this point. Yes, the pink boxes indicate the H-Cl events. We have corrected the related text. Please see the changes in the revised manuscript (MS).

(Line 299-308) "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, grey, and pink 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. Fig. 3d: The wind direction should be averaged arithmetically. Vector averaging should be used to do so. Check the following link for instructions: https://math.stackexchange.com/questions/44621/calculate-average-wind-direction

Response:

Authors thank the reviewer for the constructive comments. Your suggestion looks very legitimate. Authors revised the calculation as per suggestion and revised the plot. The plot has been corrected in the revised manuscript.

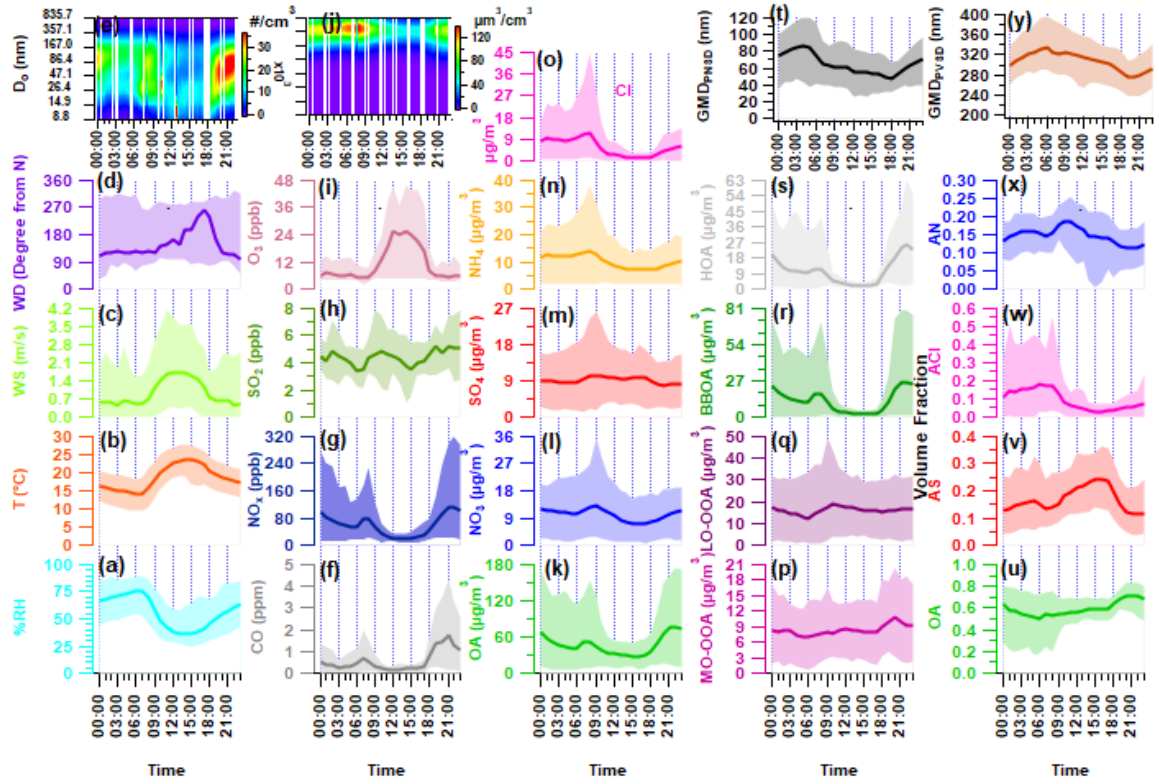

Figure 3: Diurnal variation of ambient meteorological parameters (a) % ambient relative humidity (RH), (b) temperature (T), (c) wind speed (WS), (d) wind direction (WD), and (e) particle number size distribution (PNSD), mass concentration of ambient trace gases (f) carbon mono-oxide (CO), (g) nitrogen 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 (SO_4), (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 (GMD_{PNSD}) 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. No x-axis label for Fig. 8 in the revised manuscript.

Response:

The plot has been corrected in the revised manuscript.

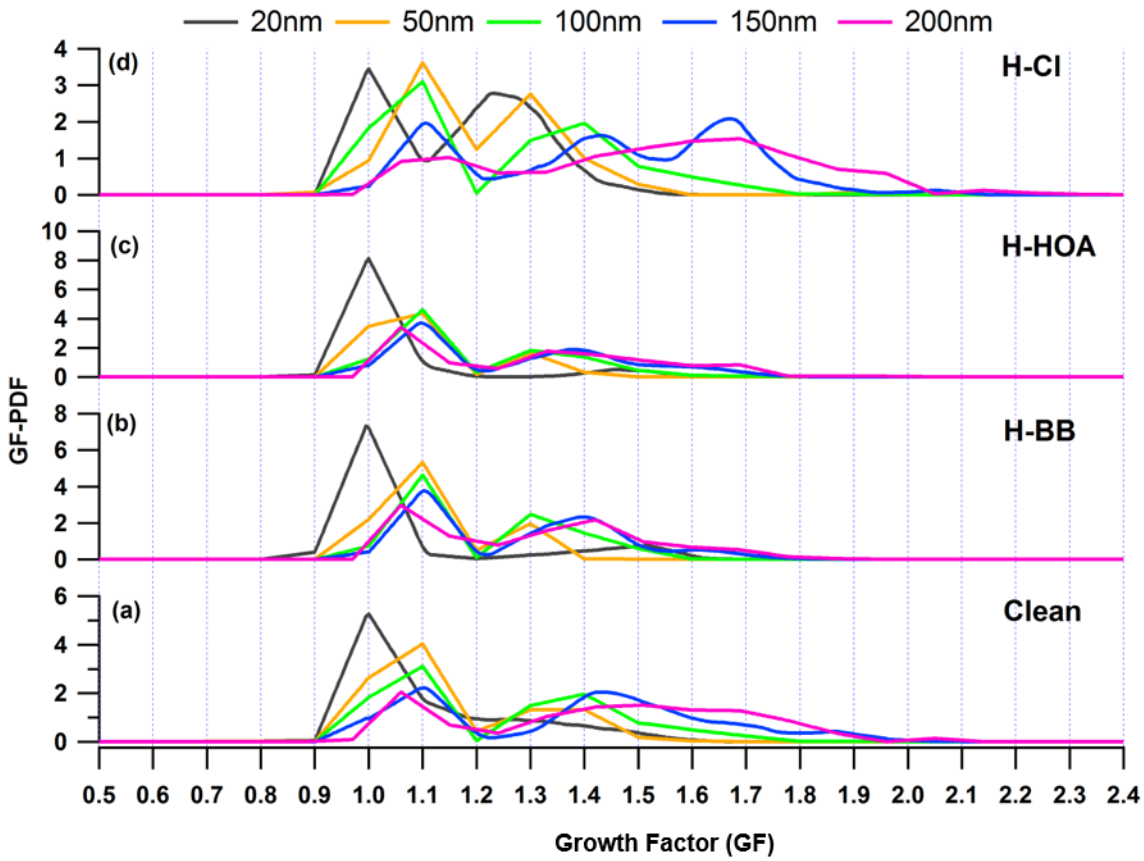

Figure 8: Growth Factor Probability Density Function (GF-PDF) of 20, 50, 100, 150, and 200 nm aerosol particles for the (a) clean, (b) H-BB, (c) H-HOA, and H-Cl periods.

Response to the Referee (RC) 3

There is a technical error still in the revised version and need to be fixed before publication. The figure 3 (o) left axis is not well adjusted.

Authors thank the reviewer for highlighting this point. Please see the changes in the revised manuscript (MS).

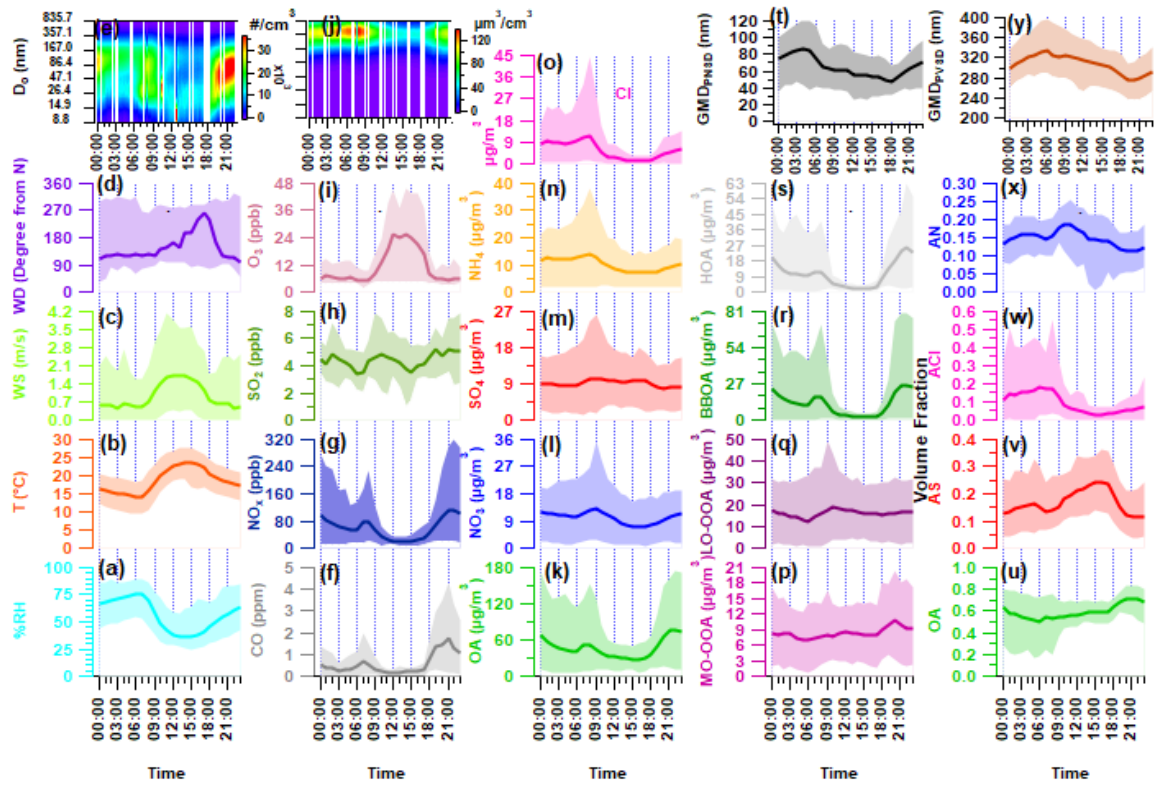

Figure 3: Diurnal variation of ambient meteorological parameters (a) % ambient relative humidity (RH), (b) temperature (T), (c) wind speed (WS), (d) wind direction (WD), and (e) particle number size distribution (PNSD), mass concentration of ambient trace gases (f) carbon mono-oxide (CO), (g) nitrogen 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 (SO_4), (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 (GMD_{PNSD}) 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.

Response to the Editor

1. The readability and the grammar of the manuscript need to be deeply improved. It is highly recommended that the manuscript is reviewed by a native English speaker.

Authors thank the reviewer for highlighting this point. Please see the changes in the revised manuscript (MS).

2. In the abstract several terms are defined; however, they are only used once. Please only use an abbreviation when a specific term is going to be used several times. For example, there is no need to use the following abbreviations in the abstract: ACSM, DAS, H-TDMA, CCN, and OA.

Authors thank the reviewer for highlighting this point. Please see the changes in the revised manuscript (MS).

*"**Abstract.** Recent research has revealed the crucial role of winter-time, episodic high chloride (H-Cl) emissions in the Delhi region, which significantly impact aerosol hygroscopicity and aerosol-bound liquid water, thus contributing to the initiation of Delhi fog episodes. However, these findings have primarily relied on modeled aerosol hygroscopicity, necessitating validation through direct hygroscopicity measurements. This study presents the measurements of non-refractory bulk aerosol composition of PM_1 from an Aerodyne aerosol chemical speciation monitor and for first-time size-resolved hygroscopic growth factors (Nucleation, Aitken, and Accumulated mode particles) along with their associated hygroscopicity parameters at 90% relative humidity using a hygroscopic-tandem differential mobility analyzer at the Delhi Aerosol Supersite. Our observations demonstrate that the hygroscopicity parameter for aerosol particles varies from 0.00 to 0.11 (with an average of 0.03 ± 0.02) for 20 nm particles, 0.05 to 0.22 (0.11 ± 0.03) for 50 nm particles, 0.05 to 0.30 (0.14 ± 0.04) for 100 nm particles, 0.05 to 0.41 (0.18 ± 0.06) for 150 nm particles, and 0.05 to 0.56 (0.22 ± 0.07) for 200 nm particles. Surprisingly, our findings demonstrate that the period with H-Cl emissions displays notably greater hygroscopicity (0.35 ± 0.06) in comparison to spans marked by high biomass burning (0.18 ± 0.04), high hydrocarbon-like organic aerosol (0.17 ± 0.05), and relatively cleaner periods (0.27 ± 0.07). This research presents initial observational proof that ammonium chloride is the main factor behind aerosol hygroscopic growth and aerosol-bound liquid water content in Delhi. The finding emphasizes, ammonium chloride's role in aerosol-water interaction and related haze/fog development. Moreover, the high chloride levels in aerosols seem to prevent the adverse impact of high organic aerosol concentrations on cloud condensation nuclei activity."*

3. On the other hand, the authors define the same term several times along the manuscript. It is recommended that a term is defined the first time it is used and later on only the abbreviation is used. For example, H-Cl should be defined in L15 and not in L26.

Thank you for pointing out it. Authors modified the text throughout the manuscript. Please see the changes in the revised manuscript (MS).

4. Figures S3 and S4 are not called in the main text.

Thank you for pointing out it. Please see the changes in the revised manuscript (MS).

(Line 206-207) "Our results shows a strong correlation and nearly unit slope (0.9999) between the calculated and modeled inorganic salts, as presented in Fig. S3."

(Line 264-266) "Additionally, we observed a high correlation (r^2 = 0.83, $p<0.05$) between PM_1 measured by ACSM and MPSS, assuming an effective aerosol density of 1.6 g/cm^3 (refer to Fig. S4)."

5. Figures S5 and S6 cannot be called before Figures S2, S3, and S4.

Thank you for pointing out it. Please see the changes in the revised manuscript (MS).

L29-30: "Ammonium Chloride" should be "ammonium chloride"

Thank you for pointing out it. Please see the changes in the revised manuscript (MS).

(Line 28-30) "This research presents initial observational proof that ammonium chloride is the main factor behind aerosol hygroscopic growth and aerosol-bound liquid water content in Delhi."

L35: Add a reference after "budgets"

Thank you for pointing out it. Please see the changes in the revised manuscript (MS).

(Line 34-36) "The Intergovernmental Panel on Climate Change (IPCC) (Intergovernmental Panel on Climate Change, 2023) reported that the interaction between aerosols and clouds is not completely comprehended, and there are significant uncertainties in gauging global radiative budgets."

Reference:

Intergovernmental Panel on Climate Change: Climate Change 2023 – The Physical Science Basis, Cambridge University Press., 2023.

L39: "is crucial to predict the" should be "is crucial to better predict the"

Thank you for pointing out it. The txt has been modified in the revised manuscript. Please see the changes in the revised manuscript (MS)

(Line 40-42) "Its comprehension is vital to better predicting the aerosol size distribution and scattering properties with more accuracy in global models under varying atmospheric humidity (RH) conditions (Randall et al., 2007)."

L41: Delete "atmospheric conditions"

Thank you for pointing out it. The text has been modified in the revised manuscript. Please see the changes in the revised manuscript (MS).

(Line 42-44)" Hygroscopicity at higher RH results in an increase in the cross-sectional area of the aerosol, leading to efficient light scattering by the aerosol particles (Tang and Munkelwitz, 1994)."

L56: It seems that "counter" is misplaced.

Thank you for pointing out it. The text has been modified in the revised manuscript. Please see the changes in the revised manuscript (MS).

(Line 55-59)" Over the past few decades, researchers have extensively measured aerosol hygroscopicity using a hygroscopic tandem differential mobility analyzer (H-TDMA) (Massling et al., 2005; Gysel et al., 2007; Mandariya et al., 2020; Swietlicki et al., 2008; Yeung et al., 2014; Kecorius et al., 2019) and a CCN counter (Bhattu and Tripathi, 2015; Gunthe et al., 2011; Massoli et al., 2010; Ogawa et al., 2016) under sub- and supersaturated conditions, respectively."

L58-61. Improve the grammar.

Thank you for pointing out it. The text has been modified in the revised manuscript. Please see the changes in the revised manuscript (MS).

(Line 55-65)" Over the past few decades, researchers have extensively measured aerosol hygroscopicity using a hygroscopic tandem differential mobility analyzer (H-TDMA) (Massling et al., 2005; Gysel et al., 2007; Mandariya et al., 2020; Swietlicki et al., 2008; Yeung et al., 2014; Kecorius et al., 2019) and a CCN counter (Bhattu and Tripathi, 2015; Gunthe et al., 2011; Massoli et al., 2010; Ogawa et al., 2016) under sub- and supersaturated conditions, respectively. Petters and Kreidenweis (2007) introduced the hygroscopicity parameter, kappa (κ), to correlate aerosol hygroscopicity with its chemical composition. Hygroscopicity of OA may differ according to their chemical properties such as solubility, extent of dissociation in aerosol water, and surface activity, which can pose challenges in quantifying OA hygroscopicity (Hallquist et al., 2009; Jimenez et al., 2009). As a result, this introduces further discrepancies in predicted and measured aerosol hygroscopicity. Therefore, there is a requirement to investigate the measurement-based aerosol hygroscopicity of Delhi's atmosphere to gain a better understanding of the recurring occurrences of haze and cloud formations."

L62: “Delhi’s atmosphere to understand” should be “Delhi’s atmosphere to better understand”

Thank you for pointing out it. The text has been modified in the revised manuscript. Please see the changes in the revised manuscript (MS).

(Line 62-65) “As a result, this introduces further discrepancies in predicted and measured aerosol hygroscopicity. Therefore, there is a requirement to investigate the measurement-based aerosol hygroscopicity of Delhi’s atmosphere to gain a better understanding of the recurring occurrences of haze and cloud formations.”

L64-65: “to severe air quality”. Do the authors mean “to severely poor/low air quality”?

Thank you for pointing out it. The text has been modified in the revised manuscript. Please see the changes in the revised manuscript (MS).

(Line 66-67) “In recent decades, rapid economic growth and industrialization in the Indo-Gangetic Plain (IGP) have resulted in significantly poor air quality during the winter season (Wester et al., 2019).”

L70-71: Improve the grammar.

Thank you for pointing out it. The text has been modified in the revised manuscript. Please see the changes in the revised manuscript (MS).

(Line 72-73) “A recent study conducted in Delhi revealed that frequent high chloride events promote high levels of aerosol liquid water content under elevated humid conditions.”

L79-80: “hygroscopicity. Hence, it is essential to measure size-resolved aerosol hygroscopicity in Delhi's atmosphere and investigate its role in the context of high chloride”. This is redundant, therefore, I suggest deleting it.

Thank you for pointing out it. The text has been DELETED in the revised manuscript. Please see the changes in the revised manuscript (MS).

L83-84: H-TDMA was already defined.

Thank you for pointing out it. The text has been modified in the revised manuscript. Please see the changes in the revised manuscript (MS).

(Line 85-89) “Real-time measurements of atmospheric aerosols were conducted during winter (February 1, 2020 to March 20, 2020) at Indian Institute of Technology (IIT) Delhi, Block 5. H-TDMA, TROPOS-type Mobility Particle Size Spectrometer (MPSS), and Aerodyne Aerosol Chemical Speciation Monitor (ACSM) from Aerodyne Research in Billerica, MA were used simultaneously at a height of approximately 15 meters above ground level (a.g.l.) as depicted in Fig. 1. Lab-2 is located 50 meters away from Lab-1.”

L84: Add the model and manufacturer of the MPSS.

MPSS is built by TROPOS itself, so it does not have a model number as it is not a commercial product. It is called TROPOS type. The text has been changed in the revised manuscript. Please see the changes in the revised manuscript (MS).

(Line 85-89) "Real-time measurements of atmospheric aerosols were conducted during winter (February 1, 2020 to March 20, 2020) at Indian Institute of Technology (IIT) Delhi, Block 5. H-TDMA, TROPOS-type Mobility Particle Size Spectrometer (MPSS), and Aerodyne Aerosol Chemical Speciation Monitor (ACSM) from Aerodyne Research in Billerica, MA were used simultaneously at a height of approximately 15 meters above ground level (a.g.l.) as depicted in Fig. 1. Lab-2 is located 50 meters away from Lab-1."

L87: "The lab-2 is situated at the height of 15 m a.g.l. and lab-2 is 50 m apart" should be "The lab-2 is situated at the height of 15 m a.g.l. and is 50 m apart"

The text has been changed in the revised manuscript. Please see the changes in the revised manuscript (MS).

(Line 85-89) "Real-time measurements of atmospheric aerosols were conducted during winter (February 1, 2020 to March 20, 2020) at Indian Institute of Technology (IIT) Delhi, Block 5. H-TDMA, TROPOS-type Mobility Particle Size Spectrometer (MPSS), and Aerodyne Aerosol Chemical Speciation Monitor (ACSM) from Aerodyne Research in Billerica, MA were used simultaneously at a height of approximately 15 meters above ground level (a.g.l.) as depicted in Fig. 1. Lab-2 is located 50 meters away from Lab-1."

L99: "for aerosol and sheath respectively" should be "for aerosol flow and sheath flow, respectively."

The text has been changed in the revised manuscript. Please see the changes in the revised manuscript (MS).

(Line 102-103) "There are two humidity sensors (Vaisala) in the system for aerosol flow and sheath flow respectively."

L103: "Both the DMAs were". Fix it.

The text has been changed in the revised manuscript. Please see the changes in the revised manuscript (MS).

(Line 107-108) "Size calibration for both DMAs involved the application of Latex particles of standard size, 200 nm prior to measurement."

L110: MPSS was already defined.

The text has been changed in the revised manuscript. Please see the changes in the revised manuscript (MS).

(Line 113-114) "Particle number size distributions (PNSDs) and particle volume-size distributions (PVSDs) were measured using a MPSS."

L111: “A Detailed description” should be “A detailed description”

The text has been changed in the revised manuscript. Please see the changes in the revised manuscript (MS).

(Line 165) “For a detailed account of the ACSM setup, please refer to Arub et al. (2020).”

L111: “ACSM was” should be “The ACSM was”

The text has been changed in the revised manuscript. Please see the changes in the revised manuscript (MS).

(Line 165-166) “The ACSM operated in a temperature-controlled laboratory at almost 0.1 lpm and 1-minute time resolution.”

L113: “ratio (m/z) m/z 10 to m/z 140” should be “ratio (m/z) 10 to m/z 140.”

The text has been changed in the revised manuscript. Please see the changes in the revised manuscript (MS).

(Line 166-167) “It was set to measure mass-to-charge ratio (m/z) from 10 to 140.”

Lines 111-118 need to be combined with Lines 165-175 in one single section.

The text has been changed in the revised manuscript. Please see the changes in the revised manuscript (MS).

(Line 165-181) “For a detailed account of the ACSM setup, please refer to Arub et al. (2020). The ACSM operated in a temperature-controlled laboratory at almost 0.1 lpm and 1-minute time resolution. It was set to measure mass-to-charge ratio (m/z) from 10 to 140. The PM_1 aerosol beam was concentrated and directed towards the vaporizer at 600 °C. The flash-vaporized compounds were then ionized through impact ionization at 70 eV electrons and detected using a quadrupole mass spectrometer (Ng et al., 2011). The study employed a 200 millisecond amu^{-1} scan speed and a pause setting of 125 for a sampling duration of 64 seconds to collect aerosol mass spectra using the ACSM technique. Refer to Gani et al. (2019) for comprehensive guidance on the ACSM operational procedures. For ACSM calibration and data processing, please refer to Patel et al. (2021). Positive matrix factorization (PMF) was conducted on the data, resulting in a four-factor solution: hydrocarbon-like OA (HOA), biomass burning OA (BBO), less-oxidized OA (LO-OOA), and more-oxidized OA (MO-OOA), as shown in Fig. S1. More information regarding PMF analysis can be found in section S.1 and Fig. S2 of the

Supplementary information. Three different events were identified based on the temporal variation of mass concentration peaks of BBOA, HOA, and Cl (see Fig. 2): 1) a high-residential or biomass burning period (H-BB); 2) a high-hydrocarbon-like OA period (H-HOA); and 3) a high-chloride period (H-Cl). Additionally, the "Clean Period" was defined as a period where the PM_1 loading was less than the 25th percentiles (≤ 38.7 μg m^{-3}) for the sampling period. The event's starting and ending times were determined by the initial increase in concentration and subsequent return to the starting values as the concentration decreased."

L167: "Patel et al., 2021." Fix it.

The text has been changed in the revised manuscript. Please see the changes in the revised manuscript (MS).

(Line 171-172) "For ACSM calibration and data processing, please refer to Patel et al. (2021)."

L220-223: RH, T, WD, WS, PNSD, and OA were already defined.

The text has been changed in the revised manuscript. Please see the changes in the revised manuscript (MS).

(Line 225-227) "Fig. 2 depicts the hourly-resolved temporal changes of various meteorological parameters, including RH, T, WD, and WS, PNSD, PVSD, principal components of non-refractory PM_1, and OA with their corresponding fractional mass contributions."

L226. RH and T were already defined.

The text has been changed in the revised manuscript. Please see the changes in the revised manuscript (MS).

(Line 229-231) "The ambient RH and T vary within the range of 24.2% to 96.6% and 9.0 °C to 28.5 °C, respectively. The average values of RH and T are 56.0% ± 18.2% and 18.7 °C ± 4.2 °C, respectively."

L243: WS was already defined.

The text has been changed in the revised manuscript. Please see the changes in the revised manuscript (MS).

From here on, I stopped checking the grammar, spelling, and punctuation. Please check out the rest of the manuscript.

The text has been changed in the revised manuscript. Please see the changes in the revised manuscript (MS).