Measurement report: Hygroscopicity of Size-Selective Aerosol Particles at Heavily Polluted Urban Atmosphere of Delhi: Impacts of Chloride Aerosol

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Abstract. Recent research has unveiled the pivotal role of wintertime episodic high chloride emissions in the Delhi region, which significantly influence aerosol hygroscopicity and aerosol-bound liquid water, thereby contributing to the initiation of fog episodes in Delhi. However, these findings have primarily relied on modeled aerosol hygroscopicity, necessitating validation through direct hygroscopicity measurements. 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. Our observations indicate that the hygroscopicity parameter ($\kappa_{H\text{-}TDMA\_90\%}$) ranges from 0.00 to 0.11 (with an average of 0.03 ± 0.02) for 20 nm aerosol 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. Remarkably, our results reveal that the period characterized by high chloride (H-Cl) emissions exhibits significantly higher hygroscopicity (0.35 ± 0.06) compared to periods marked by high biomass burning (H-BB) (0.18 ± 0.04), high hydrocarbon-like organic aerosol (H-HOA) (0.17 ± 0.05), and relatively cleaner periods (0.27 ± 0.07). This study provides first 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/fog development. 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.

1. Introduction

The Intergovernmental Panel on Climate Change (IPCC) reported that aerosol-cloud interaction is still not fully understood and has significant uncertainties in quantifying global radiative budgets. Aerosol hygroscopicity plays a pivotal role in overcoming and explaining these uncertainties. Hygroscopicity is crucial to understand how the aerosol particles act as cloud condensation nuclei (CCN) and forms fog droplets/haze at sub-saturated/nearly saturation and cloud droplets at atmospheric supersaturation levels (McFiggans et al., 2006; Topping and McFiggans, 2012). Its understanding is crucial to predict the aerosol size distribution and scattering properties better in global models under varying atmospheric humidity (RH) conditions (Randall et al., 2007). Hygroscopicity at higher RH atmospheric conditions leads to an enhanced aerosol cross-sectional area, resulting in efficient light scattering by the aerosol particles (Tang and Munkelwitz, 1994). It mainly depends on particle size and chemical composition. Generally, the inorganic salts such as ammonium salts of sulfate, nitrate, and chloride, are highly hygroscopic (Hu et al., 2011; Petters and Kreidenweis, 2007), organic aerosol (OA) are comparatively less hygroscopic (Jimenez et al., 2009; Kroll et al., 2011), while dust particles and black/elemental carbon particles are stated as hydrophobic (Seinfeld and Pandis, 2006). Further, the elevated atmospheric RH during winter and monsoon favour the formation of more oxidized secondary organic aerosol (SOA) via aqueous-phase (Ervens et al., 2011) and heterogeneous reactions (McNeill, 2015), leads to enhancement in organic aerosol hygroscopicity (Jimenez et al., 2009; Mei et al., 2013) which adversely impact on the local visibility (Li et al., 2016; Liu et al., 2012). However, aerosol loading inversely affects aerosol hygroscopicity (Mandariya et al., 2020a). Apart from it, aerosol loading is also a critical factor in deciding the lifetime of cloud, which affects the region's rain quantitatively (Albrecht, 1989; Lohmann and Feichter, 2005).

Over the past decades, aerosol hygroscopicity has been intensively measured using 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 CCN (Bhattu and Tripathi, 2015; Gunthe et al., 2011; Massoli et al., 2010; Ogawa et al., 2016) counter under sub- and supersaturation levels, respectively. Petters and Kreidenweis (2007) introduced a hygroscopicity parameter, kappa (κ), to associate aerosol hygroscopicity with its chemical composition. Furthermore, hygroscopicity associated with OA potentially varies with OA chemical properties like solubility, the extent of dissociation in aerosol water, and surface activity (Hallquist et al., 2009; Jimenez et al., 2009), leads to difficulty in the quantification of OA hygroscopicity, result in introducing more discrepancies in predicted and measured aerosol hygroscopicity. Hence, there is a need to

62 explore measurement-based aerosol hygroscopicity for Delhi's atmosphere to understand the frequent haze/cloud

63 formations better.

64 In past decades, fast economic growth and industrialization in the Indo Gangetic Plain (IGP) led to severe air

65 quality during wintertime (Wester et al., 2019). Delhi is potentially affected by local and regional air pollution

66 problems in wintertime (Arub et al., 2020; Bhandari et al., 2020; Gani et al., 2019; Prakash et al., 2018). Recent

67 studies (Gani et al., 2019; Rai et al., 2020) have shown chloride is one of the predominant factors to degrade the

68 air quality in the Delhi region and significantly favour the haze/fog formation during winter (Gunthe et al., 2021).

69 Trash and biomass burning for heating and waste degradation are among the main contributors to chloride in Delhi

70 (Rai et al., 2020). A recent study conducted in Delhi reported that frequent high chloride events promotes high

71 aerosol liquid water content under elevated humid condition leads to haze and poor visibility in the city (Chen et

72 al., 2022). In addition, Gunthe et al. (2021) showed higher chloride also facilitates enhancement in aerosol

73 hygroscopicity, however, this study was based on theoretical hygrocopicity. Therefore, it is essential to investigate

74 the impacts of chloride on aerosol hygroscopicity and its potential to enhance aerosol-bound liquid water based

75 on field measurements. Moreover, the hygroscopicity of the aerosol particles in the heavily polluted urban

76 atmosphere, which confines to highly complex composition, is extremely limited, like Delhi, situated at Indo

77 Gangetic Plain (IGP), India, where air quality severely degrades during haze/fog-dominated. To the author's best

78 knowledge, the current study is first in Delhi, India, exploring a complex atmosphere of IGP using H-TDMA-

79 measured aerosol hygroscopicity. Hence, it is essential to measure size-resolved aerosol hygroscopicity in Delhi's

80 atmosphere and investigate its role in the context of high chloride.

81 **2. Experimental Methods**

82 **2.1 Aerosol Measurements**

83 Real-time atmospheric aerosol measurements were conducted simultaneously using Hygroscopic-Tandem

84 Differential Mobility Analyzer (H-TDMA), Mobility Particle Size Spectrometer (MPSS), and Aerodyne Aerosol

85 Chemical Speciation Monitor (ACSM, Aerodyne Research, Billerica MA) during winter (1st February 2020 to

86 16th March 2020) at the Indian Institute of Technology (IIT) Delhi in Block 5, at the height of nearly 15 m above

87 ground level (a.g.l.) as shown in Fig. 1. The lab-2 is situated at the height of 15 m a.g.l. and lab-2 is 50 m apart

88 from lab-1.

89 In this study, the HTDMA system was used to investigate the hygroscopic growth of size-resolved particles at 90

90 % RH. The HTDMA system has been previously used in many field campaigns (Massling et al., 2007; Wu et al.,

91 2013b; Zhang et al., 2016). The HTDMA system (TROPOS, Germany) is comprised of two Differential Mobility

92 Analyzers (DMAs, type Hauke-median, TROPOS, Germany), a Condensation Particle Counter (CPC, Model

93 3772, TSI Inc., USA) along with a humidifier system located between the two DMAs. The role of first DMA is

94 to select the quasi-monodisperse particles at a dry diameter (D_p, dry) with 30% RH. After that, the size-selected

95 particles pass through a humidity conditioner, which can be adjusted from 30% to 90% RH by regulating the

96 aerosol and sheath air flow by mixing dry air with RH<5% and humid air with ∼95% RH (Maßling et al., 2003).

97 The uncertainties associated with RH measurement at 90% RH is 1.0%. The particle hygroscopic growth

98 distribution at dry size (D_p, dry) at a certain humidity can be easily determined with CPC. There are two humidity

99 sensors (Vaisala) in the system for aerosol and sheath respectively. The humidity sensors positioned in the second

100 DMA were calibrated automatically with 100 nm ammonium sulfate (($NH_4)_2SO_4$) particles every 30 min at 90%

101 RH to analyze the stability at high RH. The measurement error of the HTDMA mainly depends on the uncertainty

102 in measuring and controlling the RH within the system (Su et al., 2010). Therefore, all RH sensors were calibrated

103 using the Vaisala salt kit comprising LiCl, NaCl, KCl etc. prior the measurement campaign. Both the DMAs were

104 size calibrated by applying the Latex particles with the standard size of 200 nm before the start of the measurement.

105 The number concentration peak occurred at 203 nm, referring to accuracy of DMAs size selection at 1.5%.

106 HTDMA system was operated at 90% RH to measure the hygroscopic growth factors (HGFs) for particles with

107 D_p, dry of five different sizes i.e. 20, 50, 100, 150 and 200 nm. The time resolution of the full scan covering the

108 five sizes was about 30 min.

109 Particle number size distributions (PNSDs) and particle volume-size distributions (PVSDs) were measured using

110 a Mobility Particle Size Spectrometer (MPSS (TROPOS type)).

111 A Detailed description of ACSM setup can be found in Arub et al. (2020). ACSM was operated at nearly 0.1 lpm

112 at 1 min time resolution in a temperature-controlled laboratory. ACSM was set to run to measure mass-to-charge

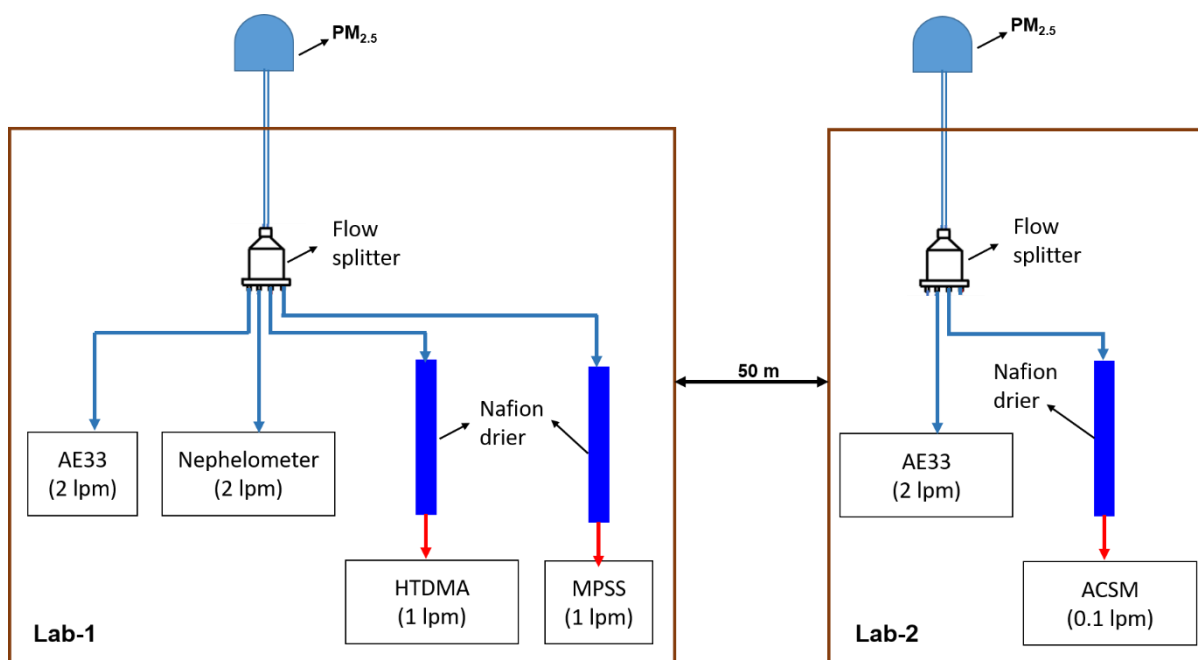
113 ratio (m/z) m/z 10 to m/z 140. The ACSM measures non-refractory particulate matter less than 1μm (NR-PM_1).

114 The concentrate PM_1 aerosol beam was impacted on the vaporizer at 600 °C and flash-vaporized compounds were

115 subsequently ionized through impact ionization at 70 eV electron and detected with a quadrupole mass

116 spectrometer (Ng et al., 2011). The 200 ms amu^{-1} scan speed and pause setting at 125 for a sampling time (64 s)

117 were set to acquire aerosol mass spectra in ACSM. Detailed operational procedures for the ACSM are explained

118 elsewhere in Gani et al. (2019).

119

120 **Figure 1: Schematic diagram of the inlet systems for aerosol sampling instruments. The blue and red**

121 **sampling lines indicate the ambient air and dehumidified (RH<25%) ambient air, respectively.**

122 **2.2 Meteorological and Gas Data**

123 The gas data was taken from the location site R.K Puram -DPCC, a continuous ambient air quality monitoring

124 station controlled by the central control room for air quality management (Delhi-NCR). The gas data were

125 downloaded from the CPCB website (https://app.cpcbccr.com/ccr/#/caaqm-dashboard/caaqm-landing/data). R.K.

126 Puram is located 3.5 km northwest of IIT Delhi. The wind speed (WS), wind direction (WD), temperature (T),

127 and relative humidity (RH) were continuously measured using an automatic weather station (Watch Dog 2000

128 series). The weather station is mounted over the top of the 9th-floor building of the IITD.

129 **2.3 Data Analysis**

130 **2.3.1 H-TDMA**

131 Overall, we recorded 1483 H-TDMA scans cycles. Afterward, the % difference between measured and theoretical

132 growth factors (Δq) was calculated after each scan cycle for 100 nm ammonium sulfate particles. Those scan

133 cycles came between $\Delta q \leq \pm 5\%$, were only carried out for further data treatment, and the rest scans cycles were

134 discarded (Kecorius et al., 2019). Thus, we had 1102 H-TDMA scan cycles following this data quality check.

135 Regarding good scan cycles, we had 1449, 1431, 1438, 1470, and 1420 good H-TDMA scans for 20, 50, 100,

136 150, and 200 nm particles, respectively, to further analyze. Afterward that, a piecewise linear TDMAinv

137 algorithm, namely TDMAinv Toolkit, written in IgorPro and developed by Gysel et al. (2009), was used to do

138 post-data treatment on the raw HGF. Because the measured distribution function is a skewed and smoothed

139 integral transform of the actual growth factor probability density functions (GF-PDFs). A detailed description of

140 the raw data processing in the TDMAinv toolkit to measure real HGFs is described in Gysel et al. (2009). The

141 TDMAinv toolkit was successfully used in various studies around the globe (Gysel et al., 2007; Liu et al., 2012;

142 Sjogren et al., 2007; Wang et al., 2018a) and at Kanpur, India (Mandariya et al., 2020a). Besides, the RH in the

143 DMA2 generally achieved the set value of 90% and remained stable within ±1%, although occasionally, it faced

144 a more considerable drift. All growth factors measured between 88 and 92% RH were corrected to a target value

145 of 90% (HGF_90%) (Gysel et al., 2007) using the kappa-model suggested in the TDMAinv toolkit (Gysel et al.,

146 2009) to minimize this DMA2 RH drifts. After it, 979, 957, 972, 969, and 966 scans were found corrected at target

147 RH for 20, 50, 100, 150, and 200 nm aerosol particles, respectively, which further averaged for 60 min time

148 resolution and finally, these numbers reached to 425, 429, 419, 424, and 417, respectively.

149 Further, size-resolved hygroscopicity factors (kappa, κ, say $\kappa_{H\text{-TDMA}\_90\%}$) were calculated from the respective size-

150 resolved target RH corrected HGFs using equation (1) kappa-Köhler theory (Mandariya et al., 2020a; Petters and

151 Kreidenweis, 2007).

152 $$\kappa_{H-TDMA\_90\%} = (HGF\_90\%^3 - 1)\left[\frac{1}{RH} \, exp\left(\frac{4\sigma M_w}{RT\rho_w D_o HGF_{90}\%}\right) - 1\right], \tag{1}$$

153 Where, $\kappa_{H\text{-TDMA}\_90\%}$ is the hygroscopicity factor at 90% RH, HGF_90% is the size-resolve HGF at 90% RH, RH

154 is the atmospheric relative humidity in fraction, σ is the surface tension of the aerosol liquid droplet-air interface

155 at the droplet surface in N/m and can be assumed nearly to pure water, R is the universal gas constant in J K^{-1} mol^{-1}

156 ^{1}, M_w is the molecular mass of water, T is the ambient temperature in Kelvin (K), ρ_w is the density of water in

157 kg/m^3, and D_o is the dry mobility diameter of the particle in m.

158 **2.3.2 MPSS**

159 MPSS measures electrical mobility distribution, which is then converted to PNSD in the 8 to ~800 nm mobility

160 diameter range by applying an inversion algorithm to correct for multiple charged aerosol particles (Wiedensohler,

161 1988; Pfieffer et al., 2014) and diffusional losses (Wiedensohler et al., 2012; 2018).

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163

2.3.3 ACSM

An Aerodyne Aerosol Chemical Speciation Monitor (ACSM, Aerodyne Research, Billerica MA) provided mass concentrations of organic aerosol (OA), ammonium (NH_4), sulfate (SO_4), nitrate (NO_3), and chloride (Cl). Details on ACSM calibration and data processing are in Patel et al., 2021. We conducted Positive matrix factorization (PMF) on the data and found a four-factor solution (hydrocarbon-like OA, "HOA"; biomass burning OA, "BBOA"; less-oxidized OA, "LO-OOA"; more-oxidized OA "MO-OOA) to best represent the data set. Further details about PMF analysis are in section S.1 of the SI. Furthermore, based on the mass concentration peaks of BBOA, HOA, and Cl in the temporal variation (Fig. 1), 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 ($\leq$ 38.7 $\mu g\ m^{-3}$) 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.

2.3.4 Derived Secondary Inorganic Salts

The ACSM mainly measures OA, NO_3, SO_4, NH_4, and Cl. Therefore, we adopted a simplified ion-pairing scheme reported by Gysel et al. (2007). 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:

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

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

$$ABS = 115.11 \times n_A$$

$$AS = 0$$

$$AN = 0$$

$$ACl = 0$$

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

189    $SA = 0$

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

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

192    $AN = 0$

193    $ACl = 0$

194   **Case-3 $R_{SO_4} \geq 2$**

195    $SA = 0$

196    $ABS = 0$

197    $AS = 132.1405 \times n_S$

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

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

200   Here, n denotes the number of moles, whereas A, N, S, and C denotes the NH_4, NO_3, SO_4, and Cl species. We

201   also predicted these inorganic salts concentrations from the ISORROPIA v2.1 model using NH_4, SO_4, NO_3, and

202   Cl. We found a strong correlation and nearly unit slope (0.9999) between the calculated and modelled inorganic

203   salts as presented in Fig. S1, which strongly justifies the new ion-pairing scheme adopted in this study.

2.3.5 Windrose and Potential Source Contribution Function (PSCF)

205   The windrose plot was plotted by openair in R package (http://www.r-project.org, http://www.openair-

206   project.org). The 48-hour back trajectory of air masses reaching Delhi super site (DSL) at 500 m above the ground

207   at every hour for the entire study period was estimated by an offline based Hybrid Single-Particle Lagrangian

208   Integrated Trajectory (HYSPLIT4) model developed by NOAA/Air Resources Laboratory (ARL)) (Draxler and

209   Hess, 1997). The input meteorological data for back trajectories were taken from the Global Data Assimilation

210 System (GDAS 0.5 degree) archive maintained by ARL (http://ready.arl. noaa.gov/archives.php). Further,

211 utilizing these estimated back trajectories as input combined with the measured mass fraction of chemical species

212 of bulk aerosol, Potential Source Contribution Function (PSCF) analysis was carried out with the help of a tool

213 called Zefir (V 3.7) written in Igor Pro (WaveMetrics). Detail description regarding Zefir tool can be found

214 elsewhere (Petit et al., 2017). In addition, box plots reported in the subsequent section were also plotted with the

215 help of this tool. The aerosol liquid water content (ALWC) as a function of inorganic species mass concentration,

216 ambient temperature (T), and ambient relative humidity (RH), was calculated by ISORROPIA-II model

217 (Fountoukis and Nenes, 2007).

3. Result and Discussions

3.1 Overview of meteorology, trace gases, and aerosol characterization

220 Fig. 2 illustrates the hourly-resolved temporal variability of meteorological parameters, including relative

221 humidity (RH), temperature (T), wind direction (WD), and wind speed (WS), as well as the particle number size

222 distribution (PNSD), particle volume size distribution (PVSD), principal components in non-refractory PM_1, and

223 organic aerosol (OA) with their fractional mass contributions. Additionally, Fig. S5 displays the temporal

224 variability of atmospheric gases, such as nitrogen oxides (NOx), carbon monoxide (CO), and sulfur dioxide (SO_2).

225 Delhi's winter climate is primarily influenced by a depression generated by Western Disturbances, resulting in

226 cold waves in the region. The ambient relative humidity (RH) and temperature (T) exhibit variability in the range

227 of 24.2% to 96.6% and 9.0 °C to 28.5 °C, respectively, with average values of 56.0% $\pm$ 18.2% and 18.7 °C $\pm$ 4.2

228 °C. These fluctuations indicate that Delhi's atmosphere transitions from being wet and cold in February to dry and

229 relatively warm in March. Notably, nighttime conditions tend to be cooler and more humid compared to daytime

230 throughout the sampling period. Ambient RH exhibits a diurnal pattern, with a peak in the early morning (06:00-

231 07:00 hours) and a valley around midday (13:00-15:00 hours). In contrast, ambient temperature follows an

232 opposing trend, rising during midday, which can be correlated with higher solar radiation during those hours (see

233 Fig. 3a & b). The higher ambient temperature and peak O_3 concentration during midday (Fig. 3(i)) suggest the

234 presence of daytime photo-oxidation processes (Nelson et al., 2023). The wind speed (WS) and wind direction

235 (WD) varied from 0.0 to 5.6 (1.0 $\pm$ 1.0) m/s and 4.0 to 345.7 (197.1 $\pm$ 84.4) degrees from the North, respectively,

236 as shown in Fig. S6. Predominant wind directions were WNW-WSW and E-ESE. These patterns suggest that the

237 atmosphere remains relatively stagnant during the study period, and the measured aerosols likely represent

238 emissions and local aerosol chemistry in Delhi.

239   Additionally, ambient trace gases NOx and CO exhibit significant variability throughout the sampling period,

240   peaking during local burning activities. Ambient NOx levels reach a maximum of 421.2 ppb (58.4 ± 61.9) during

241   intense biomass burning activities. CO concentrations also peak during similar periods as NOx and vary from 0.0

242   to 7.66 ppm (0.58 ± 0.79), as shown in Fig. S5. The diurnal variation of these trace gases is presented in Fig. 3 (f,

243   g, h, and i), with two peaks (06:00-08:00 and 17:00-20:00 hours) associated with morning local biomass/trash

244   burning emissions and nighttime traffic rush hours. In contrast, SO_2 follows a different trend, with dynamic

245   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

246   at midnight (21:00-02:00 hours) associated with the local industrial stack emissions.

247   The PM_1 particle number concentration ranges from 408 to 29,845 /cm^3 (11319 ± 5552). High particle number

248   concentrations are typically associated with local burning events. The particle concentration increases in the

249   evening (at 18:00 hours) and reaches its maximum value at midnight, suggesting the resumption of residential

250   burning activity and traffic emissions. These activities likely contribute to the lower geometric mean diameter

251   (GMD) of the particle number size distribution (PNSD, approximately 47 nm), which increases to nearly 87 nm,

252   as shown in Fig. 3(t), indicating nighttime aging of organic aerosol. The hourly averaged mean diurnal GMD of

253   PVSD varies from approximately 274 to 324 nm (Fig. 3(y)), with a mean value of 309.1 ± 33.1 nm, which is close

254   to this study's higher-end particle size of 200 nm hygroscopicity measurement. Therefore, ACSM bulk aerosol

255   composition is the most suitable choice for discussing the hygroscopicity of these particles.

256   The hourly time-resolved $NR-PM_1$ (say hereafter PM_1) concentration varied from 9.0 to 357.9 μg/m^3, averaging

257   81.2 ± 56.6 μg/m^3. This observation lies well within the boundary of 12.7-392 μg/m^3 ($NR-PM_1$), reported by Gani

258   et al. (2019) for the same sampling site. Prakash et al. (2018) reported that PM_1 mass concentration is 83% of

259   $PM_{2.5}$, representing the dominance of combustion-based particles. Further, we observed that ACSM measured

260   PM_1 was highly correlated ($r^2 = 0.83$, p<0.05) with MPSS measured PM_1, assuming an effective aerosol density

261   1.6 g/cm^3 (Fig. S2). The OA ranged between 1 and 293 (46.5 ± 39.6) μg/m^3 with the predominant fraction of PM_1,

262   consistent with the range of 53.3 to 166 (112) μg/m^3 observed during winter (December-February) at the same

263   site (Gani et al., 2019). However, lower average OA concentration could be explained by the measuring period of

264   February-March, as aerosol loading starts decreasing in February after reaching its peak in December-January

265   (Gupta and Mandariya, 2013). The campaign average fractional contribution OA to PM_1 was 56%, ranging from

266   1 to 84%. This high OA contribution in PM_1 is consistent with other studies conducted in IGP (Chakraborty et al.,

267   2016a; Gani et al., 2019; Mandariya et al., 2019) and worldwide (Jimenez et al., 2009; Zhang et al., 2007). Peaked

268   OA mass concentrations were noted between 21:00-23:00 hr (figure 2(k)), consistent with previous studies

269 conducted at the current site (Gani et al., 2019; Rai et al., 2020). Campaign average mass concentration of NO_3

270 was 10.1 ± 7.0 μg/m^3 and showed diurnal variation with a peak in the morning and midnight (Fig. 3(l)). Besides,

271 SO_4 showed slight enhancement at 08:00 hr and remained nearly constant from noon to 17:00 hr (Fig. 3(m)).

272 However, Cl varied between 0.13 to 77.83 μg/m^3, and higher concentrations of Cl were found episodic throughout

273 the campaign. The Cl$^-$ concentration was found consistent with Gani et al., 2019's previously reported value of

274 0.1-66.6 μg/m^3 at the same site. The temporal variation of various OA factors is presented in Fig. 2(g and h).

275 Biomass burning organic aerosol (BBOA) mass concentration peaks during the night and morning hours (Fig.

276 3(r)). Low-volatility oxygenated organic aerosol (LO-OOA) exhibits a peak in the morning and remains relatively

277 constant at noontime, suggesting steady formation. Meanwhile, moderately oxygenated organic aerosol (MO-

278 OOA) shows a slight increase around noontime, indicating formation through daytime photooxidation (Mandariya

279 et al., 2019; Sun et al., 2016). Overall, oxygenated organic aerosol (OOA) was the dominant fraction of OA during

280 the sampling period. The H-BB events displayed a dynamic variation in BBOA mass concentration, ranging from

281 16.3 to 134.7 (averaging 50.7 ± 24.0) μg/m^3. Concurrently, these events also exhibited a higher concentration of

282 HOA (ranging from 9.6 to 109.4 μg/m^3), suggesting a potential similarity in the sources of HOA during this

283 occurrence. However, during H-HOA events, a higher concentration of HOA (ranging from 4.8 to 58.9 μg/m^3)

284 was observed, although these concentrations were notably lower than those observed during H-BB events.

285 Nevertheless, fractional mass contribution of HOA to OA was largest among all OA species. Furthermore, H-Cl

286 events exhibited elevated concentrations of both primary organic aerosol HOA and BBOA. BBOA contributed

287 approximately 40.0%, 21.1%, 32.5%, and 13.1% to OA during H-BB, H-HOA, H-Cl, and relatively clean events,

288 respectively, suggesting diverse sources of BBOA. Moreover, during the H-HOA event, HOA's average

289 contribution was the highest among all events, at 41.6%. Additionally, Cl's fractional mass contribution in PM_1

290 reached up to 44.9% during the H-Cl event, in contrast to 21.2% in H-BB events and 7.3% in H-HOA events.

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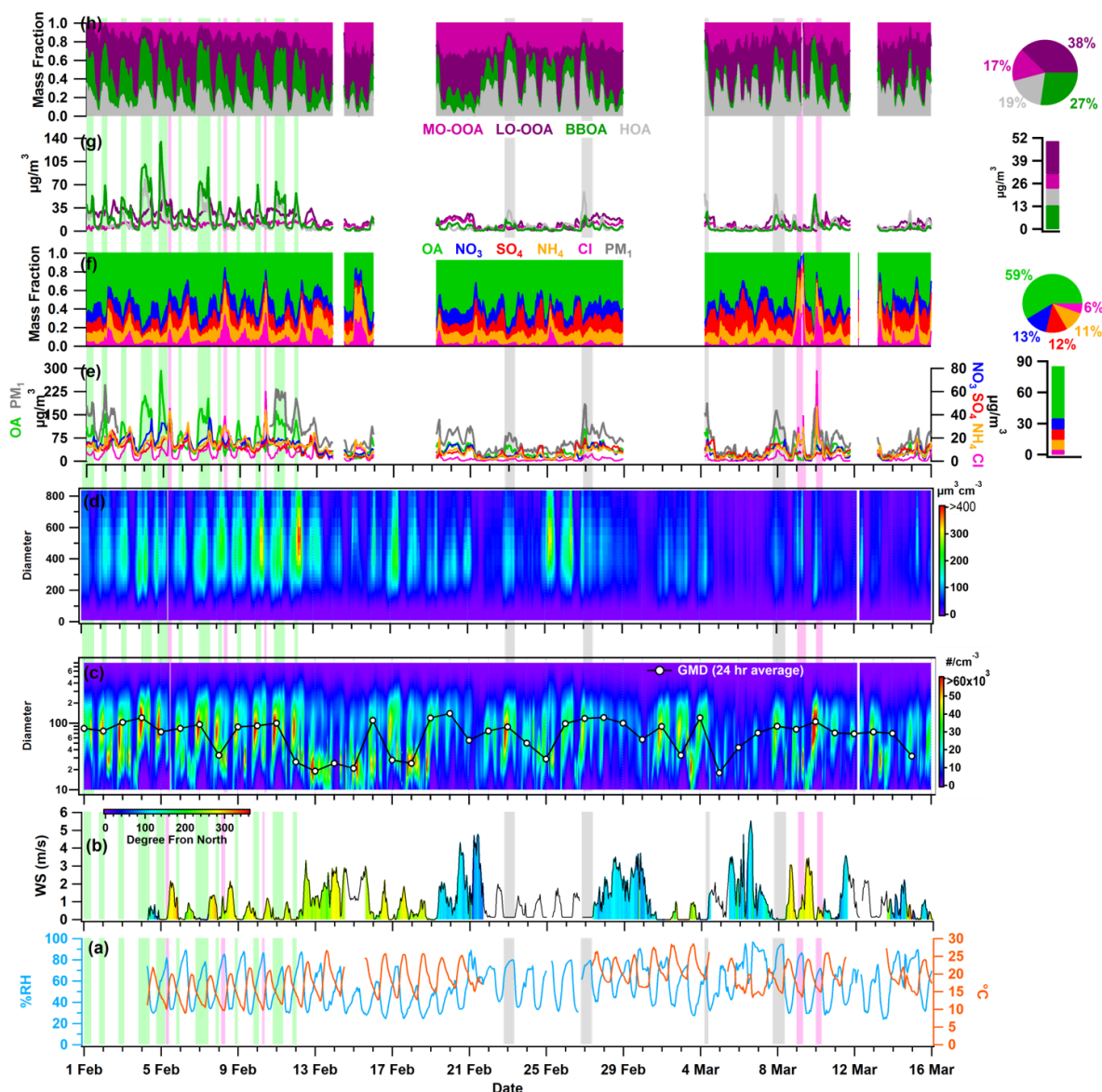
Figure 2: Temporal variability of ambient (a) relative humidity (RH), temperature (T), (b) wind speed (WS), wind direction (WD), (c) particle number-size distribution (PNSD), 24-average geometric mean diameter (GMD), (d) particle volume-size distribution (PVSD), (e) particulate matter (PM₁), organic aerosol (OA), nitrate (NO₃), sulfate (SO₄), ammonium (NH₄), chloride (Cl), (f) fractional contribution of OA, NO₃, SO₄, NH₄, and Cl in PM₁, (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.

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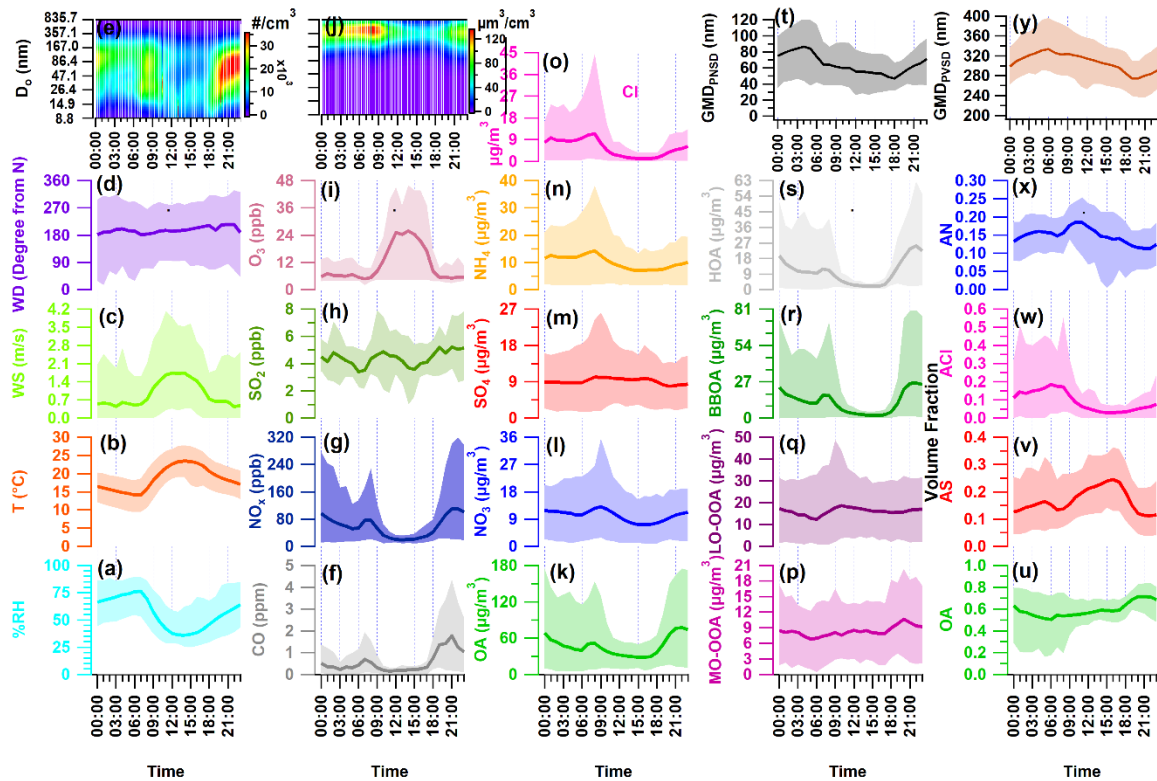
Figure 3: Diurnal variation of ambient meteorological parameters (a) % ambient relative humidity (RH), (b) temperature (T), (c) wind speed (WS), (d) wind direction (WD), and (e) particle number size distribution (PNSD), mass concentration of ambient trace gases (f) carbon mono-oxide (CO), (g) nitrogen oxides (NOx), (h) sulfur dioxide (SO₂), and (i) ozone (O₃), (j) particle volume size distribution (PVSD), mass concentration of aerosol constituents (k) organic aerosol (OA), (l) nitrate (NO₃), (m) sulfate (SO₄), (n) ammonia (NH₄), 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 PM₁, 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.

3.2 Hygroscopicity of Nucleation, Aitken, and Accumulation Mode Particles

3.2.1 Temporal variability

Fig. 4 shows the dynamic variability in the hourly averaged HGF$_{90\%}$ and hygroscopicity parameter ($\kappa_{H\text{-}TDMA\_90\%}$, kappa) of Nucleation, Aitken, and Accumulation mode aerosol particles at 90% ambient relative humidity. 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. These mean hygroscopic growth factors were noted as statistically ($p < 0.05$) different from each other. In addition, the hygroscopicity ($\kappa_{20nm\_90\%}$ and $\kappa_{50nm\_90\%}$) of 20 and 50 nm aerosol particles varied between 0.00-0.11 and 0.02-0.25, with an average of 0.03 ± 0.02 and 0.09 ± 0.03, respectively. Nucleation mode particles were observed, mainly monomodal GF-PDF (Fig. 4(a)), comprising nearly 74±24% nearly hydrophobic particles (HGF <1.2). However, this contribution was raised to 100%, which was observed to have a good association with night-time local burning activities, as shown in the Fig. 4(a). The nucleation mode particles ($\kappa_{20nm\_90\%}$) showed significantly ($p < 0.05$) lower hygroscopicity than Aitken mode particles ($\kappa_{50nm\_90\%}$). Hong et al. (2015) reported that nucleation mode particles are more sensitive to condensable vapors like fresh VOCs, H_2SO_4 and HCl. However, the present study did not measure these species. The κ of Aitken size particles were comparable with 0.24 ± 0.08 of 52.6 ± 6.9 size particles reported by Gunthe et al. (2011) for Beijing. Beijing is also one of the most polluted urban locations like Delhi, which could justify the comparison. However, Gunthe et al. (2011) performed this study using CCN on supersaturation levels. The campaign average hygroscopicity parameter (kappa, $\kappa_{90\%}$) increased significantly ($p < 0.05$) with particle size, which can be attributed to the kelvin effect (Wang et al., 2018a). In the accumulation size range (100, 150, and 200 nm), $\kappa_{90\%}$ increased to ∼0.56. The overall sampling average values of $\kappa_{100nm\_90\%}$, $\kappa_{150nm\_90\%}$, and $\kappa_{200nm\_90\%}$ were 0.14 ± 0.04, 0.18 ± 0.06, and 0.22 ± 0.07, respectively. The $\kappa_{200nm\_90\%}$ varied between 0.05 and 0.56. The similar kind of variation in κ with particle size has been demonstrated in Kanpur, situated at the center of IGP, India (Mandariya et al., 2020a) and worldwide studies (Cerully et al., 2015; Enroth et al., 2018; Fan et al., 2020; Kawana et al., 2016; Kim et al., 2020; Kitamori et al., 2009; Ogawa et al., 2016; Sjogren et al., 2012; Wang et al., 2018a). Moreover, this was attributed to the predominant increment in inorganic to OA fraction in particles with increment in size. Furthermore, $\kappa_{H\text{-}TDMA\_90\%}$ was found approximately in the 0.13-0.77, reported by Arub et al. (2020) at Delhi for PM$_1$ without considering BC. Although, Arub et al. (2020) theoretically predicted particles' hygroscopicity by considering a particle's chemical composition. They found a decrease in κ calculation by 10% when BC was considered in aerosol chemical composition. Also, $\kappa_{H\text{-}TDMA\_90\%}$ measured in the current study were found in line with the global average value of 0.27 ± 0.21 for continental aerosols (Petters and Kreidenweis, 2007; Pringle et al., 2010). Further, to understand the impact of a particle's chemical composition, local meteorology, and air mass trajectories on $\kappa_{H\text{-}TDMA\_90\%}$ for accumulation mode particle discussed in subsequent sections.

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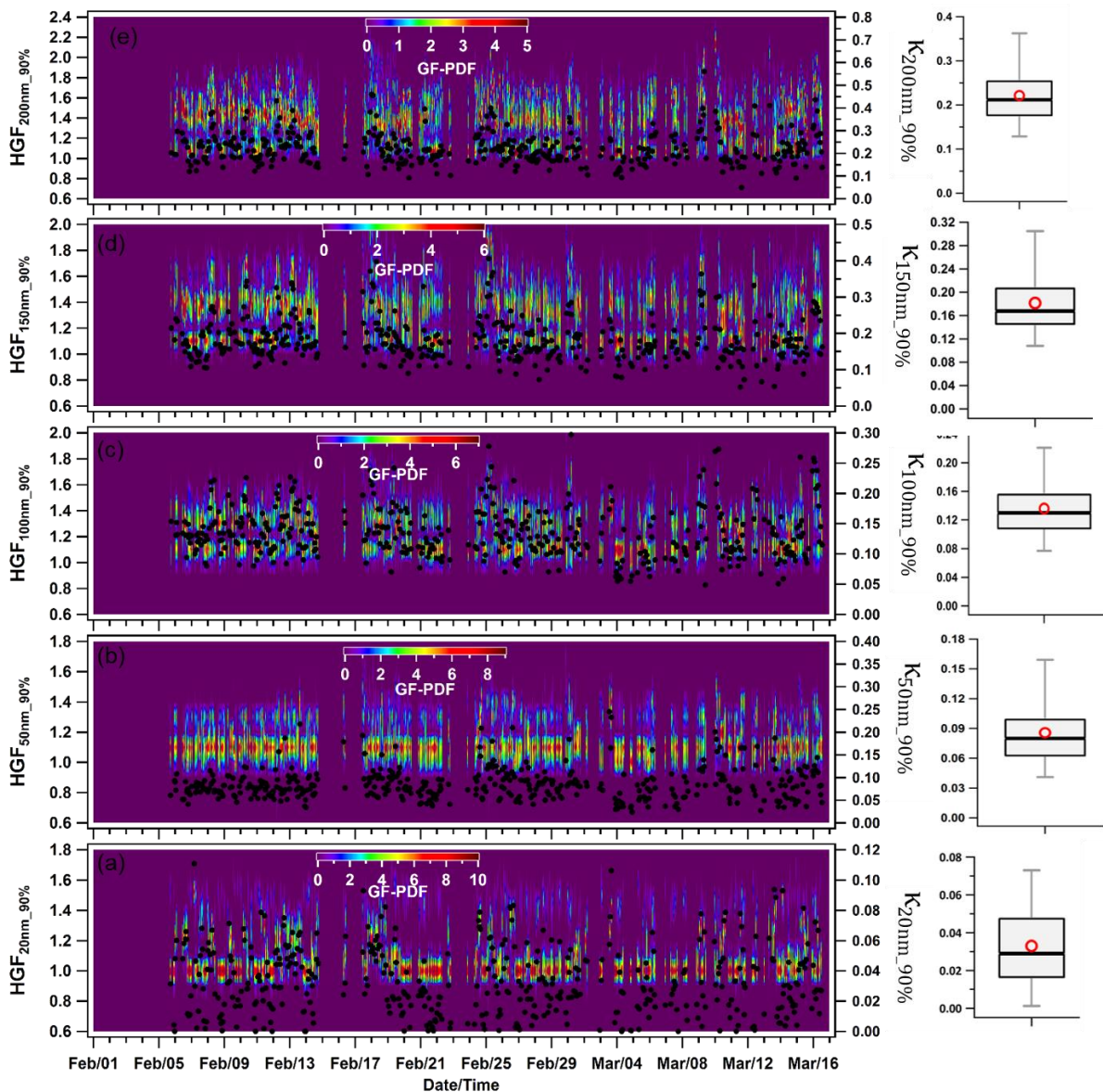
Figure 4: Temporal variability in hygroscopic parameter kappa (κ) of nucleation mode particles (a) 20 nm ($\kappa_{20nm\_90\%}$), Aitken mode particles (b) 50 nm ($\kappa_{50nm\_90\%}$), and Accumulation mode particles (c) 100 nm ($\kappa_{100nm\_90\%}$), (d) 150 nm ($\kappa_{150nm\_90\%}$), and (e) 200 nm ($\kappa_{200nm\_90\%}$). The box plots represent the variability in the hygroscopicity of respective sizes of particles in which low and high whisker traces represent the 5 and 95 percentile, respectively. The red marker indicates the average of the data, whereas the upper and lower sides of the boxes indicate the 75 and 25 percentile of the data, respectively.

3.2.2 Diurnal variability

358   The diurnal variability in $\kappa_{\text{H-TDMA\_90\%}}$ was found different for nucleation ($\kappa_{20nm\_90\%}$), Aitken ($\kappa_{50nm\_90\%}$), and

359   Accumulation ($\kappa_{100nm\_90\%}$, $\kappa_{150nm\_90\%}$, and $\kappa_{200nm\_90\%}$) mode particles. Fig. 5 displayed a diel variation of an average

360   of hourly-resolved κ for each size. The bigger size particles exhibited higher values of κ than smaller size particles,

361   which is a similar trend reported at Kanpur, India (Mandariya et al., 2020a) and other worldwide locations (Fan

362   et al., 2020; Hong et al., 2015). In general, it was observed that all size particles exhibited late-night hump (02:00-

363   05:00 hr) in $\kappa_{\text{H-TDMA\_90\%}}$. Besides, only $\kappa_{20nm\_90\%}$ demonstrated a clear diurnal variability with two peaks, one late

364   night (02:00-04:00 hr) and the other in noontime (14:00-16:00 hr), and two valleys during the morning (07:00-

365   10:00 hr) and night (19:00-22:00 hr). These valleys reflects the strong impacts of local burning and traffic

366   activities (Pringle et al., 2010). In addition, nucleation size particles were potentially contributed by nearly

367   hydrophobic particles (HGF<1.2) from evening to midnight. They showed mono modal GF-PDF around unit

368   hygroscopic growth factor, possibly indicating local emission generated particles. The 20 nm particles are small

369   enough and lie on the boundary of nucleation mode particles. Achtert et al. (2009) reported a similar diurnal trend

370   of Nucleation and Aitken mode particles, attributed the lower values to the emission of hydrophobic aerosol

371   particles during the local burning emissions. Daytime hump is attributed to the intense photochemical oxidation

372   process, which causes the enhancement of more oxidized species on the aerosol particle. Furthermore, their

373   chemical composition is dominantly controlled by the gaseous condensation of H_2SO_4, HNO_3, and VOCs (Hong

374   et al., 2015). The aerosol's chemical composition can address this variability of $\kappa_{\text{H-TDMA\_90\%}}$. However, $\kappa_{50nm\_90\%}$

375   also follows a similar diurnal variability as $\kappa_{20nm\_90\%}$, although it showed less variability. Further, as the dry size

376   of the aerosol particles increased to accumulated mode, diurnal variation shifted toward nearly steady for the rest

377   of the day. Hong et al. (2018) also observed no obvious diurnal pattern for 100 and 150 nm particles of organic-

378   dominated aerosols over the Pearl River Delta region in China.

379   Furthermore, the diurnal cycles of aerosol physicochemical properties also reflect the dynamic diurnal variation

380   in the planetary boundary layer (PBL) that leads to the accumulation of particles during night-time. Although this

381   study did not quantify size-resolved chemical composition, so, this study used bulk-aerosol composition to address

382   the trend variability only. However, daily average aerosol PNSD varied between 18.0-140.0 nm with a mean of

383   73.1 ± 33.8 nm. And, the mode of PVSD changed approximately around 300-600 nm. Therefore, it could be an

384   excellent approximation to discuss $\kappa_{200nm\_90\%}$ variability with aerosol's bulk chemical properties. The midnight to

385   early morning hump in hygroscopicity of accumulation mode particles can be attributed to the high rise in the

386   ratio of inorganic volume fraction to OA volume fraction (Fan et al., 2020), as illustrated in figure 2 (r, s t, u, and

387   v). Moreover, during mid-night and early morning in the winter, water-soluble organic and inorganic gases are

388   partitioned and/or coagulated/condensed on the surface of the pre-existing particles. Further, in the presence of

389   high RH and lower temperature, primary and secondary less oxidized organic aerosol participated in the aging

390   process, which leads to enhancement their oxidation via aqueous/heterogeneous reaction, according to it increase

391   the particle's hygroscopicity (Jimenez et al., 2009; Wu et al., 2016). Similar results were observed by Fan et al.

392   (2020) during winter in urban Beijing, and they attributed it with the enhancement of more hygroscopic particles

393   due to the aqueous-oxidation and/or condensation process on the pre-existing particles. In general, higher

394   noontime solar radiation favours more intense photooxidation processes. It supports the partitioning of relatively

395   more oxidized and less volatile organics on the particulate surface, enhancing the hygroscopicity of accumulation

396   mode particles (Duplissy et al., 2011; Massoli et al., 2010; Tritscher et al., 2011). However, interestingly, we

397   observed a noontime flatten pattern of $\kappa_{H\text{-}TDMA\_90\%}$, and it could be attributed to the mix of the positive and negative

398   impact of an enhancement in the volume fraction of OA and more hygroscopic ammonium sulfate and decrement

399   in ACl, and AN's volume fraction. Lower volume fractional contribution of highly volatile ACl could be the

400   potential factor that modulates accumulation mode particle's hygroscopicity. This can be supported by the strong

401   correlation of $\kappa_{H\text{-}TDMA\_90\%}$ and volume fraction of ACl in that size particles (ε_{ACl}).
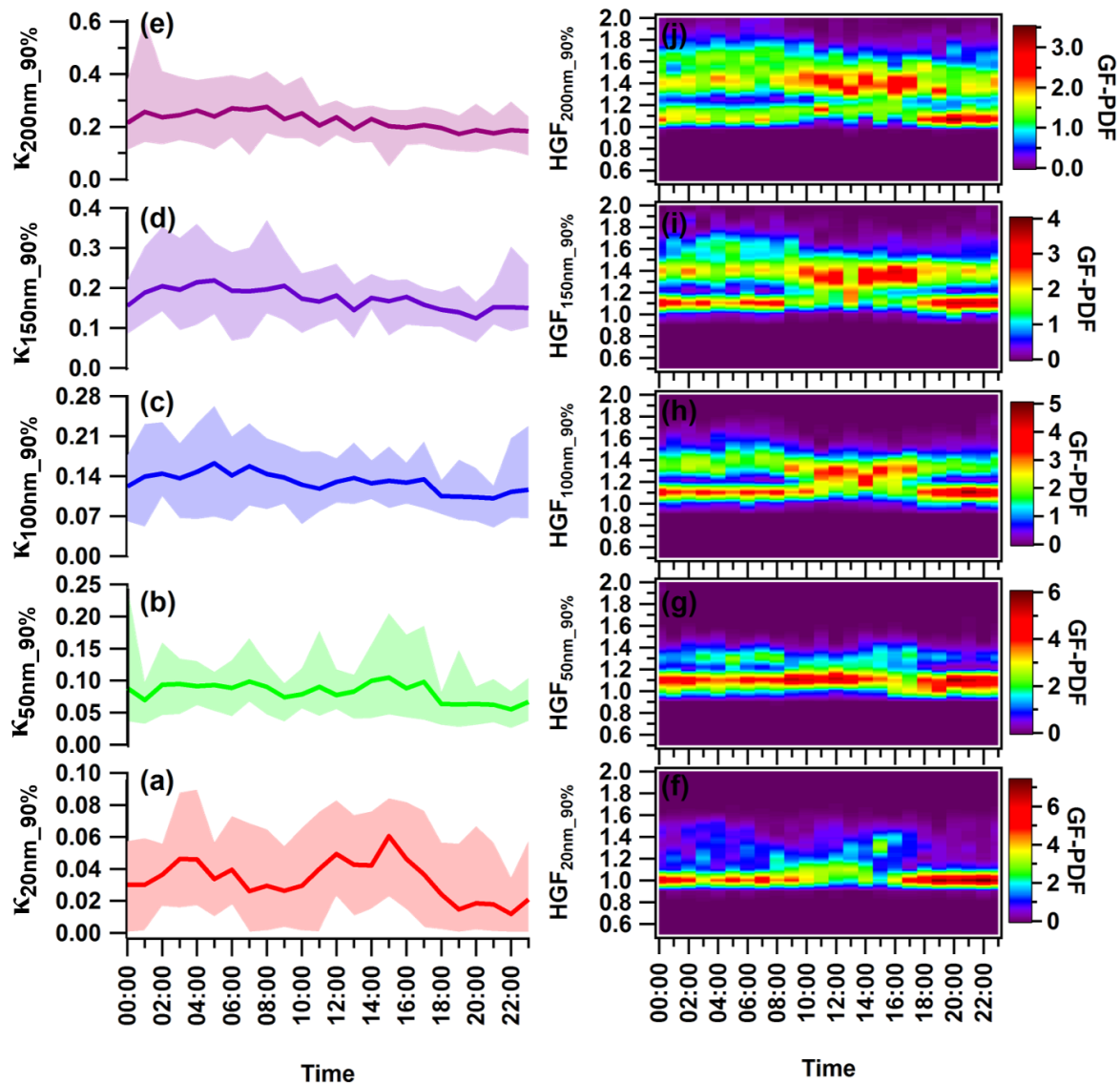
Figure 5: Diurnal variance in the hygroscopic parameter kappa (κ) of nucleation mode particles (a) 20 nm ($\kappa_{20nm\_90\%}$), Aitken mode particles (b) 50 nm ($\kappa_{50nm\_90\%}$), and Accumulation mode particles (c) 100 nm ($\kappa_{100nm\_90\%}$), (d) 150 nm ($\kappa_{150nm\_90\%}$), and (e) 200 nm ($\kappa_{200nm\_90\%}$) and hygroscopic growth factor of (f) 20 nm (HGF$_{20nm\_90\%}$), (g) 50 nm (HGF$_{50nm\_90\%}$), (h) 100 nm (HGF$_{200nm\_90\%}$), (i) 150 nm (HGF$_{150nm\_90\%}$), and 200 nm ($\kappa_{200nm\_90\%}$) aerosol particles. The solid line represents diurnal average values, and the upper and lower shaded area represents 95 and 5 percentile values of corresponding average values. Different color coding has been used to represent various size-specific kappa values. The color scale represents the growth factor probability density function of hygroscopic growth factor.

3.2.3 Driving Factor of Hygroscopicity

A correlation analysis was carried out between measured chemical species and aerosol to explore the factors governing aerosol hygroscopicity, as shown in the Fig. 6. Organic aerosol was observed negatively impact κ,

413 explained by a negative correlation (Fig. S7(a)). This negative correlation of OA with κ is also observed in India

414 (Bhattu et al., 2016; Mandariya et al., 2020b) and worldwide (Enroth et al., 2018; Hong et al., 2014; Kawana et

415 al., 2016; Kitamori et al., 2009; Wang et al., 2018a; Wu et al., 2013a). This result indicates that primary

416 constituents dominated the OA during high loading, considered nearly hydrophobic or less hygroscopic. In

417 addition, the current study observed that an enhancement of 10% of OA by volume in 200 nm aerosol particles

418 would be responsible for a 4% decrement in its hygroscopicity (Fig. S7(a)). Interestingly, ammonium sulfate and

419 nitrate showed a positive but poor correlation with hygroscopicity (Fig. 7(b and c). It could be due to sulfate and

420 nitrate aerosol dominating the bigger particles (>200nm). However, a 10% enhancement of AS by volume was

421 found to be responsible for the enhancement of hygroscopicity only by 1.6%. But if AS contribution increased in

422 the aerosol composition, aerosol water-bound capacity was negatively impacted (Fig. S8b) Besides, Fig. 6(a)

423 shown an increasing volume fraction of ACl in PM_1 with an increase in aerosol hygroscopicity, and this strong

424 positive correlation is responsible for an enhancement in kappa by 4.2% over the increment of 10% ACl by

425 volume, which was the highest among all chemical species. Further, ammonium chloride has a more significant

426 water uptake potential (Chen et al., 2022; Zhao et al., 2020), which can be justified by the solid correlation of

427 aerosol liquid water content (ALWC) with a mass fraction of ACl in PM_1 as shown in Fig. 6(b). This indicates

428 that particles with a more considerable ammonium chloride fraction uptake more water vapor, leading to higher

429 hygroscopic aerosol particles. It is clear that the increases in ammonium chloride fraction enhanced aerosol liquid

430 water content and led to higher hygroscopicity of aerosol particles. A Recent study in Delhi by Chen et al. (2022)

431 unveils that ammonium chloride fraction in PM_1 aerosol enormously enhances during the higher relative humidity

432 conditions during the winter season due to the co-condensation of semivolatile ammonium chloride with water

433 vapor on the particles and leads to enhance water uptake and lead severe winter haze in Delhi. The very high

434 volume fractions (>30%) of ACl in atmospheric PM_1 were observed episodic, suggesting a high fraction of Cl in

435 the particle phase is strongly dependent on excess ammonia in the atmosphere. These results indicate that ammonia

436 is the controlling factor for chloride partitioning in the particle phase, resulting in high aerosol water content under

437 high RH and lower temperature conditions. As the ACl is strongly dependent on the RH and temperature.
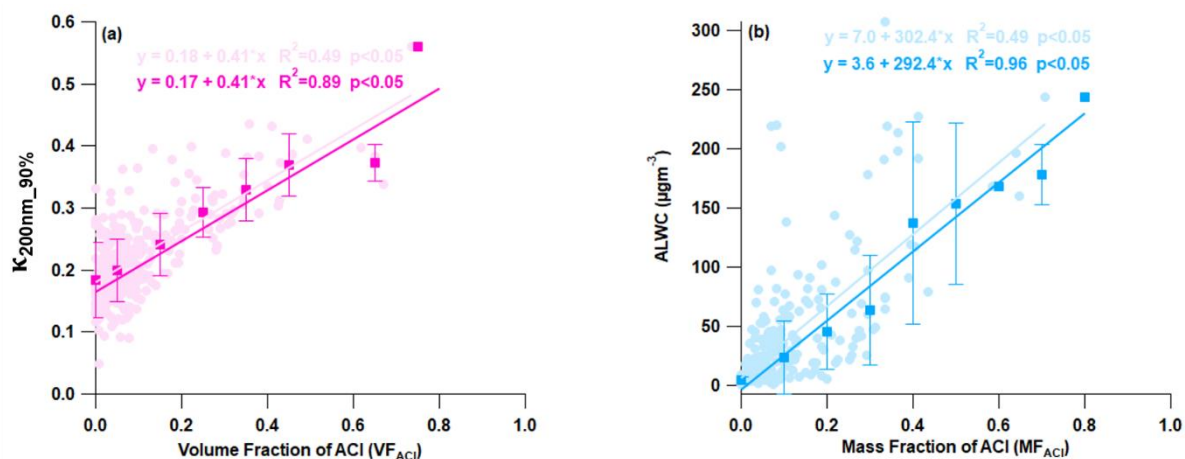
Figure 6: Correlation plot for (a) κ₂₀₀nm_90% vs volume fraction of ammonium chloride aerosol (VF_ACl) and (b) aerosol liquid water content (ALWC) vs mass fraction of ammonium chloride (MF_ACl). The solid circle and square marker represent the individual data points and the average of 10% volume and 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.

3.2.4 Hygroscopicity during high biomass burning (H-BB), high-hydrocarbon like OA (H-HOA), high-Cl (H-Cl), and relatively Clean Periods

Delhi's atmosphere is a complex array of chloride and organic aerosol sources like combustion (crop residue, agriculture waste, medical waste, municipal waste, plastic, etc., burning) and industrial sources. Therefore, all episodic events were classified into three to investigate the impact of chloride and OA on aerosol hygroscopicity. First, high biomass burning (H-BB) event; second, high-hydrocarbon like OA (H-HOA) event; and third, high-Cl (H-Cl) events. Further, aerosol chemical composition data were filtered according to hygroscopic parameter data for further analysis. By performing so, data information that is characteristic of the local emission and atmospheric chemistry in question and the effects of various potential transported air mass types can be retrieved. It is valuable to extract any possible information about aerosol sources and transformation process evaluation to interpret its influence on the aerosol's hygroscopicity.

3.2.4.1 High-Cl (H-Cl) events

H-Cl events, representing the substantial loading of ACl on the receptor site, were chosen mainly due to the significant jump (>20%) in fractional volume contribution NH_4Cl (ε_{ACl}) in the PM₁ aerosol. This period observed apparent surface wind from W-direction, although WNW, WSW, and SE winds also influence the site, as shown

in Fig. 7b. The average GMD of the PNSD was nearly 64 nm, indicating that local fresh emissions sources influence the particles. In addition, the mean SO_2, NOx, and CO concentrations were at 3.6 ppb, 51.9 ppb, and 0.4 ppm, respectively. BBOA and HOA are potentially contributed from the WNW and SE directions, as explained in the bipolar plot Fig. 7(c & d), and seem to come from a similar local source. Among inorganic species, ACl observed excellent association with ambient RH, as shown in Fig. 7e and f, indicating the atmospheric gaseous HCl neutralized with NH_3 gas in the presence of atmospheric water content. HCl sources could be coal power plants, trash burnings in solid waste dumping sites, and other industries located in the W-WSW direction (Gani et al., 2019), as shown in the map in Fig. 7a. Atmospheric high Cl events are potentially dominated by trash burning in Delhi during winter (Shukla et al., 2021; Tobler et al., 2020). Moreover, bipolar plots (figure 6(e and f)) suggest that ACl formation under high RH conditions associated with a relatively calm atmosphere trigger particles' hygroscopicity. This hypothesis can be supported with a good association of aerosol liquid water content (ALWC) as discussed in previous section. Furthermore, GF-PDF of all size particle marked relatively more fractionaly contribution of secondary mode particles as showed in Fig. 8d. Overall more hygroscopic (HGF90%>1.2) particles were marked by 42, 47, 50, 74, and 83% contributions in the 20, 50, 100, 150, and 200 nm size particles, respectively. Hence, ACl is a critical factor to enhance aerosol hygroscopicity to trigger fog/haze formation under higher RH and colder atmospheric conditions as discussed in the previous section.

Similarly, Gunthe et al. (2021) observed that high local emission of hydrochloric acid in Delhi during February-March gets partitions into aerosol liquid water under high humid conditions, enhancing the water uptake capacity of aerosol sustain particle's hygroscopic growth, result in fog/haze formation. Moreover, worldwide studies on size-resolved hygroscopicity observed Cl less than 1%, so they omitted ACl as an aerosol constituent into the discussion. In addition, the current study did not find any strong correlation of κ with AS and AN. It could be due to their association with larger particle sizes. Besides, ACl could be associated with comparatively lower size particles ($\leq$ 200 nm). Furthermore, in context to look influence of air mass trajectories, we further mapped aerosol's constituents in the association of air mass back trajectories in PSCF to see the potential area source contribution influencing the aerosol evaluation processes, ultimately aerosol's hygroscopicity. However, we did not find any back trajectory influencing the receptor site, as all trajectory endpoints were observed above the planetary boundary layer height.

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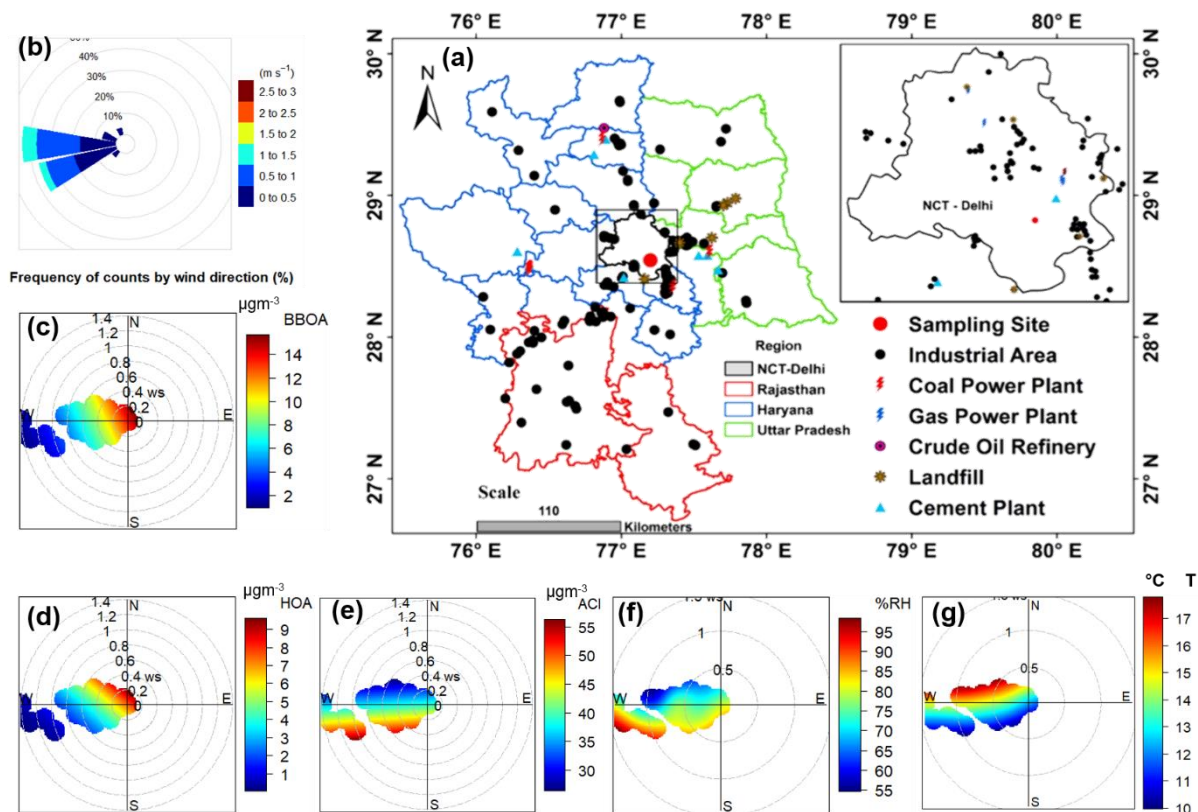


Figure 7 Map of (a) Delhi showing various types of industries located in the region and nearby locations, (b) wind rose diagram and conditional bi-polar plots showing variation in mass concentration of (c) biomass burning OA (BBOA), (d) hydrocarbon like OA (HOA), (e) ammonium chloride (ACl), (f) % ambient relative humidity (RH), and (g) ambient temperature (T), with wind direction (WD) and wind speed (WS) during H-Cl events. A background map showing various industrial locations was adapted from Rai et al. (2020).

3.2.4.2 High biomass burning (H-BB) Events

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 hours) to morning (08:00 hours) or evening (20:00 hours) to midnight (01:00 hours). Although, sometimes, it was continued from evening (21:00 hours) to morning (11:00 hours). The predominant surface wind circulations were from W, W-WNW, and W-WSW directions (Fig. S9b). The aerosol was dominated by local emissions, as aerosol constituents are mainly associated with slower wind circulations from landfill sites, industrial areas, and coal power plants, as shown in Fig. S9a. Further, it could justify the potential source contribution function (PSCF) analysis considering 48-hr air mass back trajectories, as shown in Fig. S10. The average GMD of the PNSD was nearly 87 nm. In addition, the mean SO_2, NOx, and CO concentrations were at 4.7 ppb, 124.1 ppb, and 1.5 ppm, respectively. Therefore, BBOA possibly contributed from the open local biomass burning activities at landfill sites or others. Organic aerosol in the H-BB event confined

the most considerable fraction, 39%, of BBOA, following HOA, 28%. Figure S9 (b, c, and d) clearly shows that BBOA and HOA have similar local source profiles but differ from the ACl source. Moreover, ACl was not found to have a good association with ambient RH and but was associated with nearby coal power plant's emissions. However, 48 h air mass back trajectories indicated that the current city was also influenced by air mass from some parts of Uttar Pradesh, Punjab, and Haryana. These states are the potential hub of crop residue burning, industrial activities and brick kilns. These cities have a substantial fraction of OA in PM_1 and OA mainly affected by biomass activities during winter. The H-BB event captured a considerable volume fraction, 71% of OA in PM_1 and BBOA contributed almost 39%, as illustrated in the Fig. 9. So, lower inorganic to OA ratio was a potential factor in decreasing the aerosol hygroscopicity in H-BB events. Further, a primary organic aerosol contribution was enhanced during this event and, on average, raised to 67%. OA loading inversely affects the aerosol's hygroscopicity. Mandariya et al. (2020) reported a similar observation in Kanpur, and the authors suggested that the contribution of primary biomass burning (BBOA) and hydrocarbon-like OA adversely affects aerosol hygroscopicity. BBOA showed a good negative correlation with the hygroscopicity of 200 nm particles, supporting the following conclusion. Apart from this, the Nucleation size particle (20 nm) showed 0.02 ± 0.02 hygroscopicity parameter with mono mode GF-PDF with the unit mode (Fig. 8b) and confined 83.7 ± 18.6 % nearly hydrophobic particles. Furthermore, as aerosol size increased, hygroscopicity parameter ($\kappa_{H-TDMA\_90\%}$) enhanced significantly ($p < 0.05$) as the contribution of relatively secondary aerosol particles (GF>1.2) increased with aerosol size. Accumulation size aerosol, 100 nm contributed approximately 54% by nearly hydrophobic (GF<1.2) and 46% by more hygroscopic (GF>1.2) particles.

3.2.4.3 High-HOA (H-HOA) Events

H-HOA events were identified based on the considerable mass concentration and fraction of HOA in the organic aerosol. These periods were noted generally 19:00 hr to Morning 09:00 hr during 22-23 and 26-27 February and 4, and 7-8 March as indicated in Fig. 2. The average GMD of the PNSD was nearly 80 nm. In addition, the mean SO_2, NOx, and CO concentrations were at 4.3 ppb, 136.7 ppb, and 1.1 ppm, respectively. PSCF explore the probability of impacts of long-range transported aerosol. Interestingly, it was observed that air masses over Delhi, Haryana, and Uttar Pradesh were potentially associated with hydrocarbon-like OA (Fig. S11). BBOA also followed a similar path as HOA. However, the potential area source of ACl was the nearby region of Delhi and Haryana. The HOA loading was significantly ($p < 0.05$) higher than in H-BB, H-Cl and Clean periods. However, emission sources were different during both H-HOA and H-BB periods. As HOA was the potential contributor to OA, it is likely the critical constituent to modulate aerosol hygroscopicity in the region during these events. HOA

is mainly considered hydrophobic (Duplissy et al., 2011). Therefore, elevated HOA contribution (41%) in OA could be responsible for lower κ in these events. The overall hygroscopicity of 20, 50, 100, 150, and 200 nm size particles was recorded as 0.01 ± 0.01, 0.06 ± 0.03, 0.11 ± 0.03, 0.14 ± 0.04, and 0.17 ± 0.05, respectively. The predominant fractional contribution of primary aerosol particles (GF<1.2) seems to be a reason for this lower hygroscopicity of particles, as shown in figure 7(c). Overall, OA predominantly constitutes the fraction in the PM_1, and primary OA contributed approximately 60% in OA. However, relative increment in the contribution of other more hygroscopic constituents like secondary organic aerosol (LO-OOA and MO-OOA), ACl and ammonium sulfate (AS) in the aerosol possible tried to balance the negative impact high-HOA on which limited κ.

3.2.4.5 Relatively Clean Period

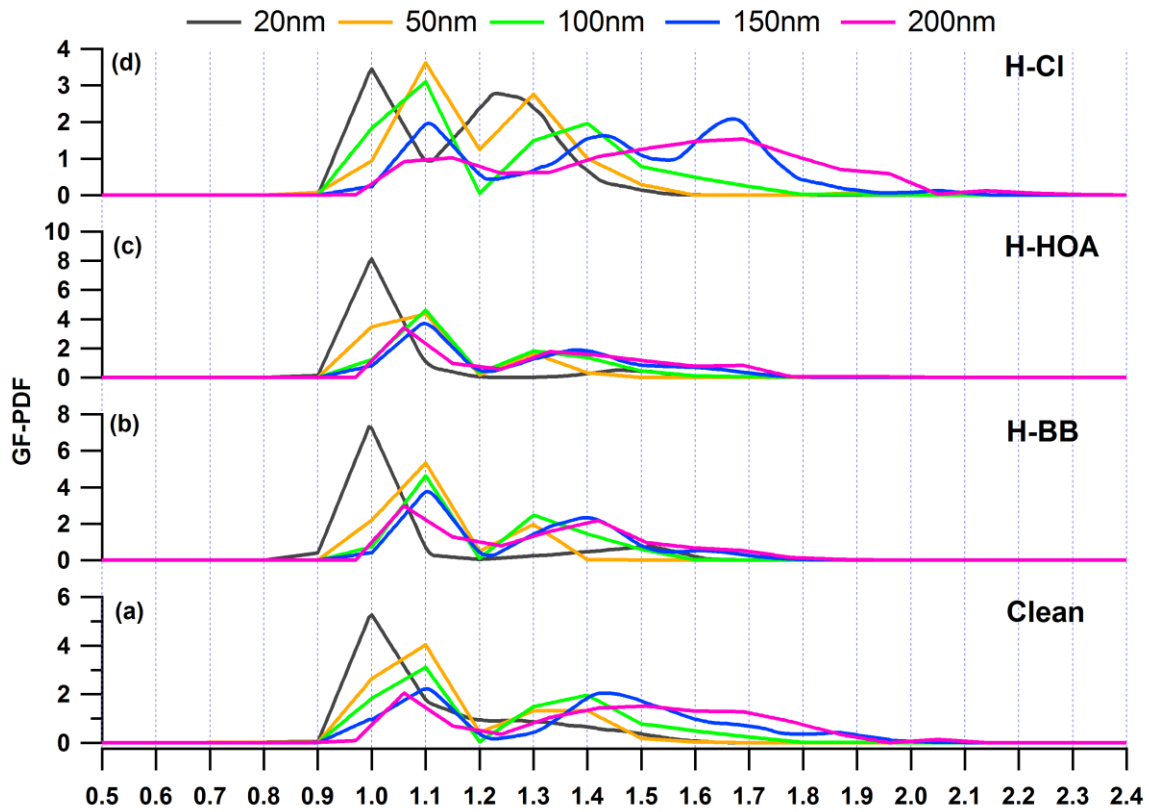
The 24th and 25th of February and the 5th, 6th, and 7th of March were marked as clean events. The night 21 hour to morning 11-hour duration was recorded as the clean duration. The relatively clean period was predominantly dominated by E, S-E winds; however, pollution was associated with calm winds, as illustrated in Fig. S9. The average GMD of the PNSD was nearly 54 nm. In addition, the mean SO_2, NOx, and CO concentrations were at 4.2 ppb, 43.2 ppb, and 0.4 ppm, respectively. All BBOA, HOA, and ACl were observed to be associated with similar sources and found an excellent association with ambient relative humidity. The mean concentration of organic aerosol, ACl, AN, and AS was observed at 11.0 ± 6.4, 1.4 ± 1.1, 3.0 ± 1.5, and 4.4 ± 2.2 μgm^{-3}, respectively. These mass concentrations were significantly lower than in other specified periods. However, OA was still the dominant species, with 56% by volume in the PM_1, as indicated in Fig. 9. Among all OA factors, HOA was predominantly dominated in OA with 33%, although secondary organic aerosol confined the overall 54.4% of OA. Secondary OA is defined with relatively higher oxidized OA, and the oxidation state of OA positively impacts OA hygroscopicity (Kim et al., 2017; Richard et al., 2011; Wu et al., 2013a). The Clean period's mean hygroscopicity of 20, 50, 100, 150, and 200 nm particles were observed at 0.03 ± 0.02, 0.09 ± 0.04, 0.14 ± 0.06, 0.22 ± 0.09, and 0.27 ± 0.07, respectively, significantly ($p<0.05$) different to each other. However, the accumulation particle's (200 nm) hygroscopicity was not significantly ($p>0.05$) higher than the 150 nm particles. The hygroscopicity increment with size from 20 to 200 nm can also be explained by the fractional increment of more hygroscopic (GF>1.2) particles relative to nearly hydrophobic or less hygroscopic particles (GF<1.2). Nucleation particles, 20 nm was dominated mostly by less hygroscopic particles ($76.8 \pm 21.7\%$), indicates influence by fresh emission sources, whereas, Aitken (50 nm) and Accumulation (200 nm) size aerosol were

564 confined with 69.3 ± 14.7 and 25.4 ± 10.8 less hygroscopic particles, respectively. These results point out that

565 accumulation-size aerosols dominated secondary aerosols, which can also support their GF-PDF as shown in Fig.

566 8(a). Nucleation size aerosol particles (20nm) showed nearly mono modal GF-PDF with the mode of unit growth

567 factor. In contrast, the mode shifted towards the higher end as aerosol size increased and GF-PDF shifted from

568 unit to multi-mode.

569

570 **Figure 8: Growth Factor Probability Density Function (GF-PDF) of 20, 50, 100, 150, and 200 nm aerosol particles for**

571 **the (a) clean, (b) H-BB, (c) H-HOA, and H-Cl periods.**

572 **3.2.4.6 Comparison of κ for different events**

573 We considered a 200 nm accumulation particle size particle representing the bulk aerosol chemical composition

574 to compare the aerosol hygroscopicity among various periods. Further, in the present study, the mode of particle-

575 volume size distribution varied from 400 nm to 600 nm particle dry mobility diameter. Therefore, 200 nm size

576 accumulation particles are the best choice to compare hygroscopicity parameters among different periods

577 considering bulk aerosol composition in various mentioned periods. In addition, a good Pearson's r value, 0.76,

578   was found among $\kappa_{200nm\_90\%}$ and $\kappa_{chem\_90\%}$, derived from the dry PM_1 particle's chemical composition measured

579   from the ACSM based on the ZSR mixing rule (Stokes and Robinson, 1966), which justifies our choice.

580   The H-Cl event noted the highest value (0.36 ± 0.06) of $\kappa_{200nm\_90\%}$ against H-BB (0.18 ± 0.04), H-HOA ($0.17 \pm$

581   0.05), and Clean (0.27 ± 0.07) events, as illustrated in Fig. 9. The H-Cl event observed that the average $\kappa_{200nm\_90\%}$

582   value was significantly ($p < 0.05$) higher than those observed in other events. It means that a substantial increment

583   in Cl emission in the Delhi region could significantly enhance the aerosol liquid water content leading to higher

584   aerosol hygroscopicity, which can further strengthen cloud condensation nuclei formation, possibly triggering

585   haze/fog events in Delhi NCR (Gunthe et al., 2021). These results suggest that controlling the open trash/waste

586   burning in the region could help control Cl emission, which leads to minimizing the haze/fog formation possibility

587   during high atmospheric conditions. However, the difference in $\kappa_{200nm\_90\%}$ values between H-BB and H-HOA

588   events was not observed significantly ($p > 0.05$), possibly due to the relative changes in primary, secondary OA,

589   and inorganic species. In the H-HOA events, the negative effect of a significantly higher fractional (41%)

590   contribution of HOA to OA possibly balances with a positive impact of a 7% increment in secondary OA relative

591   to H-BB. Worldwide studies (Jimenez et al., 2009; Mandariya et al., 2019; Sun et al., 2013) reported secondary

592   organic aerosol associated with a higher O/C ratio, and several studies reported that the O/C ratio positively

593   correlated to κ (Jimenez et al., 2009; Kim et al., 2020) as described in the earlier text. Furthermore, impacts of

594   5% decrement in ACl during H-HOA event concerning H-BB event possibly managed by 7% increment in AS

595   fraction. Overall, these relative changes in aerosol constituents worked to insignificant changes in κ during H-BB

596   and H-HOA periods. Nevertheless, H-BB and H-HOA events witnessed significant ($p < 0.05$) lower hygroscopicity

597   compared to a relatively cleaner atmosphere. The aerosol associated with relatively cleaner events was with a

598   higher inorganic-to-organic ratio. In addition, the aerosol in clean periods comprised a significantly higher fraction

599   of secondary organic aerosol, which could be the reason for the higher hygroscopicity associated with organic

600   aerosol compared to other events. Worldwide (Aiken et al., 2008; Cerully et al., 2015b; Chakraborty et al., 2016b;

601   Mandariya et al., 2019) studies have reported that organic aerosol loading inversely impacts the oxidation/aging

602   process of OA. Overall, all these were responsible for higher hygroscopicity in relatively cleaner periods.

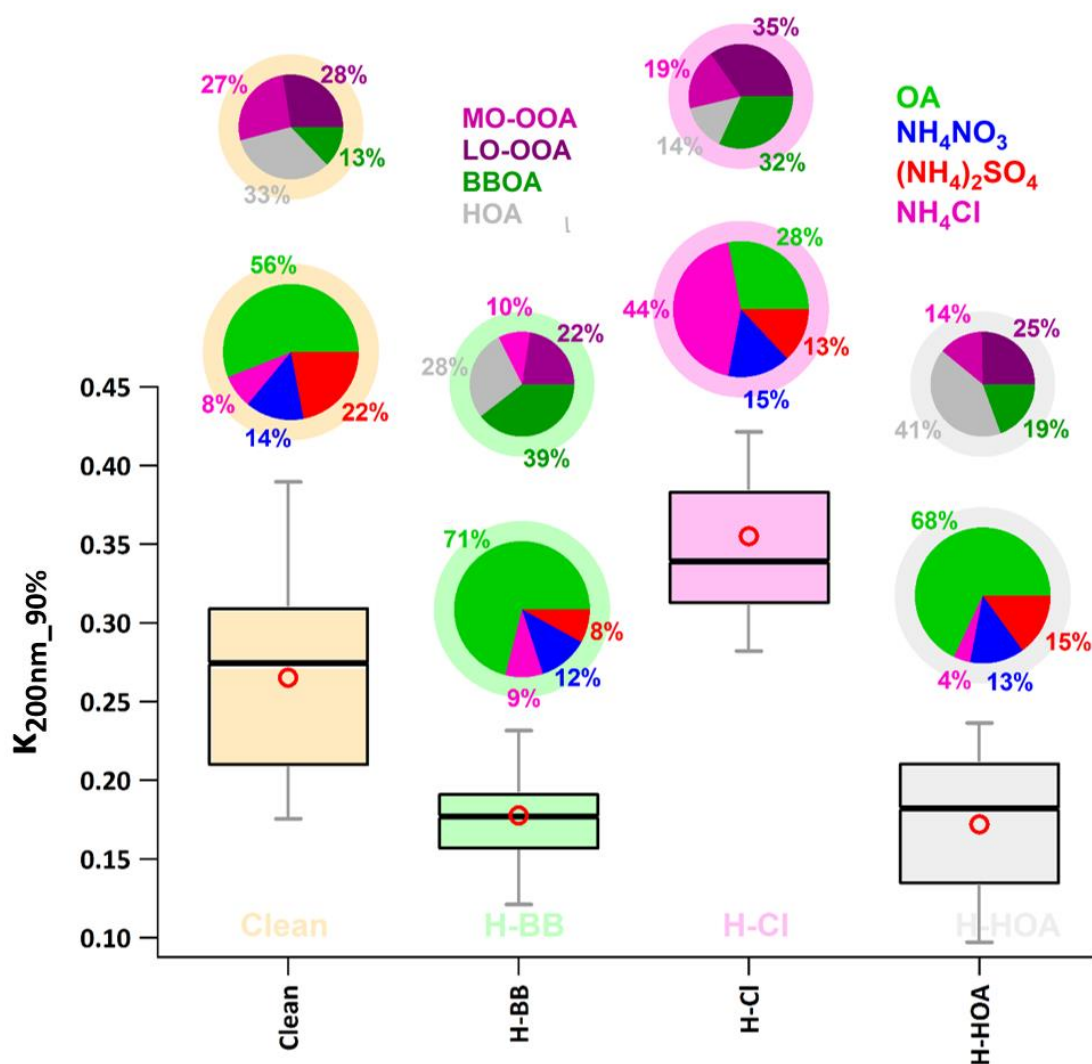



Figure 9: Box plot showing variation in H-TDMA measured hygroscopic parameter of 200 nm size particles $\kappa_{\text{H-TDMA}}$ ($\kappa_{\text{200nm\_90\%}}$) in high biomass burning (H-BB), high-chloride (H-Cl), and high-hydrocarbon like organic aerosol (H-HOA) events. Different colors represent respective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

4. Conclusions

The present study investigated the temporal variation of hygroscopicity in aerosol particles of different sizes, namely Nucleation (20 nm), Aitken (50 nm), and Accumulation (150 and 200 nm) modes in Delhi during the winter period of February-March 2020. This research also highlighted variations in hygroscopicity, specifically in aerosols with higher chloride, biomass burning, and hydrocarbon-like organic components. Delhi, known as

one of the most polluted cities, often experiences high levels of chloride pollution during winter haze and fog events. Consequently, this study reported the temporal variations in size-specific hygroscopic parameters ($\kappa_{H\text{-}TDMA\_90\%}$) under sub-saturated conditions (90% RH) in Delhi for the first time. Furthermore, it presented the hygroscopicity of nucleation and Aitken mode particles using HTDMA for the first time in India.

The observed $\kappa_{H\text{-}TDMA\_90\%}$ values ranged from 0.00 to 0.11 (with an average of 0.03 ± 0.02) for 20 nm aerosol 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. The average hygroscopicity parameter for the study period significantly increased with the size of the particles ($p<0.05$). $\kappa_{20nm\_90\%}$ and $\kappa_{50nm\_90\%}$ displayed dynamic diurnal variations, while larger accumulation mode particles exhibited a flatter diurnal pattern. This was attributed to the balancing positive and negative effects of changes in the volume fraction of NH_4Cl and organic aerosol (OA) in the aerosol with increasing particle size. Interestingly, the variation in $\kappa_{200nm\_90\%}$ was primarily associated with fluctuations in NH_4Cl and OA, rather than $(NH_4)_2SO_4$.

Furthermore, pollution episodes were predominantly linked to local biomass burning and industrial and waste-burning emissions in Delhi and nearby regions. The study primarily focused on highlighting the impacts of high biomass burning (H-BB), high hydrocarbon-like OA (H-HOA), and high chloride emissions (H-Cl) on aerosol hygroscopicity and compared them to cleaner periods. The H-Cl period exhibited significantly higher hygroscopicity (0.35 ± 0.06) compared to H-BB (0.18 ± 0.04), H-HOA (0.17 ± 0.05), and the relatively cleaner period (0.27 ± 0.07). However, H-BB and H-HOA showed no significant difference in hygroscopicity but displayed lower hygroscopicity compared to cleaner periods. This could be attributed to lower organic aerosol levels and a higher inorganic-to-organic aerosol ratio in the aerosol. The study also revealed that a 10% increase in chloride aerosol (ammonium chloride) in the aerosol significantly enhanced hygroscopicity, leading to approximately 3 $\mu g\ m^{-3}$ higher aerosol liquid water content during high chloride events. This 10% enhancement of a high-volume fraction of ammonium chloride in aerosol enhanced the aerosol hygroscopicity significantly ($p<0.05$) by 0.0041. Furthermore, the research suggested that chloride emissions were a significant concern in Delhi, enhancing aerosol hygroscopicity, promoting cloud formation during winter days, and contributing to fog and haze in the region. High chloride levels in aerosols counteracted the negative impact of high OA loading on cloud condensation nuclei (CCN) activity. Consequently, the results indicated that controlling open burning of waste materials could help reduce haze and fog events in Delhi during the winter months.

Supporting Information

644   Supplementary pieces of information are mentioned in the supplementary file.

645   **Data availability.** Data can be accessed at the following repository:

646   https://web.iitd.ac.in/~gazala/publications.html (Mandariya et al., 2023).

647   **Author contributions**

648   AH, MMVH, NAB, and GH operated aerosol instrumentation and collection of data on-board in Delhi. KP

649   analysed the ACSM data. AKM, AH, and GH conceptualized the structure of the manuscript. AKM analysed,

650   evaluated H-TDMA data, and wrote the manuscript. AH analysed MPSS data. AKM, AH, KP, JSA, LHR, AW,

651   and GH internally reviewed the manuscript and helped to write the manuscript.

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654   **Competing interests**

655   The authors declare that they have no conflict of interest.

656

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