



1 **Hyperlocal air quality monitoring and source apportionment of**  
2 **non-refractory PM<sub>2.5</sub> at three urban sites using stationary van-**  
3 **based measurements: A Lucknow case study**

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14 **Abstract.** The present study addresses a key gap in characterizing hyperlocal air quality across three contrasting land-  
15 use settings during the peak pollution season in a central city in the Indo-Gangetic Plain (IGP). We conducted ~744  
16 hours of hyperlocal measurements in Lucknow city, spanning the post-monsoon to winter season over 31 days, using  
17 a Van-Based Stationary Ambient Air Quality Monitoring Platform (VSAAQMP). Measurements were conducted at  
18 three contrasting land-use settings—a background Site (“Site 1”), a traffic corridor (“Site 2”), and a major industrial  
19 cluster (“Site 3”). High-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS) organic and inorganic  
20 mass spectra were subjected to Multilinear Engine-2, Positive Matrix Factorization (ME-2 PMF) source  
21 apportionment using a unified organic-inorganic aerosol (OA–IA) matrix. Across sites, NR-PM<sub>2.5</sub> was dominated by  
22 secondary organic aerosols (SOA), with biomass burning, traffic, and inorganic-associated organic factors (sulphate,  
23 nitrate-rich OA). Unlike Sites 1–2 (sulphate/low-volatility oxygenated organic aerosol (OOA)-dominant), Site 3  
24 showed heterogeneous OA from biomass burning, semi-volatile/nitrate OOA, solid fuel, and traffic. Particle growth  
25 events (PGEs) were predominantly nocturnal, occurring under stable boundary-layer and inversion conditions. Site 3  
26 exhibited ~5 times more nocturnal PGEs than the background, which was attributed to a higher condensation sink  
27 (CS). Stack vapors drive these PGEs (5–15 nm/h), enhancing the growth of oxidized biomass-burning organic aerosols  
28 (O-BBOA) and low-volatility OOA under inversions ( $r = 0.24$ ). These results highlight the potential of near-  
29 source stationary van-based measurements for resolving hyperlocal aerosol growth processes and source influences.

30



31 **Keywords:** Source Apportionment, Hyperlocal Monitoring, organic aerosols, Condensation sink, Particle growth  
32 events.

33

## 34 1. Introduction

35 Air pollution has emerged as a critical public health crisis in South Asia, contributing to millions of premature deaths  
36 each year. India bears one of the region's heaviest burdens, with approximately 1.67 million annual deaths linked to  
37 air pollution (Jaganathan et al., 2024). Persistent exposure to fine particulate matter—PM<sub>2.5</sub>, defined as particles smaller  
38 than 2.5 μm—remains widespread, as India ranks among the highest globally in population-weighted PM<sub>2.5</sub>  
39 concentrations (Xu et al., 2025). These challenges are especially pronounced in the Indo-Gangetic Plain (IGP),  
40 including the understudied Central IGP, home to some of the world's most polluted cities (Hao et al., 2025; IQAir,  
41 2023; Lakra et al., 2024).

42 Organic aerosols represent a substantial fraction of PM<sub>2.5</sub> across South Asia, as consistently demonstrated in recent  
43 observational studies (Hao et al., 2025; Lakra et al., 2024; Shukla et al., 2025). Globally, OA typically represents 20–  
44 60% of fine aerosol mass and can exceed 90% in biogenically dominated environments such as tropical forests  
45 (Aktypis et al., 2024). This widespread presence reflects not only their atmospheric abundance but also their active  
46 role in shaping aerosol behavior. Organic aerosols participate in processes such as new particle formation and  
47 subsequent growth (Kerminen et al., 2018), undergo complex aging, and influence atmospheric radiation and climate  
48 (Seinfeld and Pandis, 2006). Understanding where these aerosols come from, how they transform, and how they vary  
49 over time is therefore essential for interpreting PM<sub>2.5</sub> levels and assessing their broader environmental and health  
50 implications (Liu et al., 2024a). These processes are governed by contributions from both primary and secondary  
51 organic aerosol components. Primary organic aerosols (POA) are directly emitted into the atmosphere from sources  
52 like vehicular exhaust, biomass burning, and fuel combustion (Du et al., 2018). In contrast, secondary organic aerosols  
53 (SOA) form in the atmosphere when precursor gases such as sulfur dioxide (SO<sub>2</sub>), nitrogen oxides (NO<sub>x</sub>), and  
54 ammonia (NH<sub>3</sub>), and volatile organic compounds (VOCs)—undergo oxidation and produce low-volatility compounds,  
55 which then condense onto existing particles or nucleate to create new ones (Gu et al., 2023; Zhao et al., 2019). The  
56 types and amounts of emissions created can vary with ambient conditions. When VOCs undergo atmospheric  
57 oxidation, this process is closely linked to increases in particle diameter over time (Bhowmik et al., 2022; Jain et al.,  
58 2023). The resulting chemical transformations significantly influence the size and mass of PM<sub>2.5</sub> particles (Lalchandani  
59 et al., 2021).

60 Analyzing PM<sub>2.5</sub> composition reveals which species, such as sulfates, nitrates, ammonium, and organic compounds,  
61 contribute to hygroscopic and chemical growth (Kumar Kompalli et al., 2014). Variations in growth rates reflect the  
62 dynamic atmospheric processes that influence particle size distribution, toxicity, and health impacts (Schneider et al.,  
63 2015; Yue et al., 2009). Quantitative estimates of growth rates link chemical transformations with physical particle



64 evolution, aiding source identification, exposure assessment, and air quality management (Kompalli et al., 2020;  
65 Kumar Kompalli et al., 2014; Mishra et al., 2023).

66 Previous research has noted that air quality is a problem at different length scales (Roychowdhury et al., 2023). The  
67 conventional method of monitoring and managing air quality worldwide, including in India, relies on satellite  
68 observations and fixed-site Continuous Ambient Air Quality Monitoring Stations (CAAQMS)(Sathe et al., 2020).  
69 However, the approach falls short in capturing the disproportionate exposure caused by intense emissions near their  
70 sources, which occur on the neighborhood scale (Barmounakis et al., 2020). Consequently, it limits our understanding  
71 of intra-urban variability, fine spatial resolution, and hyperlocal gradients of various criteria pollutants, thereby  
72 hindering accurate health risk assessments within a region (Frederickson et al., 2023). There is a pressing need to  
73 conduct measurements at the hyperlocal level, characterizing particulate matter (PM) not only by mass concentration  
74 but also by its chemical composition, which critically influences its toxicity. Studies related to probing pathways of  
75 SOA formation and SOA were conducted earlier at Northern and CIGP Sites, primarily in Delhi (Bhattu et al., 2024;  
76 Bhowmik et al., 2022; Lalchandani et al., 2021, 2022; Shukla et al., 2021; Tobler et al., 2020). Furthermore, none of  
77 these studies used stationary van-based measurements to investigate hyperlocal air quality patterns across the city;  
78 only a few specifically analyzed intra-urban variabilities in NR-PM<sub>2.5</sub> composition. The stationary van-based  
79 measurements directly reflect air quality conditions along and near the roadways it surveys, providing localized,  
80 spatially resolved pollution data at the hyperlocal scale (Dhandapani et al., 2024; Lakra et al., 2025). The van used in  
81 the present study is equipped with highly sophisticated instruments, including an HR-TOF-AMS, an X-ray  
82 fluorescence-based metal monitor (Xact-625i), a Micro-Aethalometer, a scanning mobility particle sizer (SMPS), an  
83 optical particle sizer (OPS), and supporting instruments, such as pollutant gas analyzers and a Vaisala. Additionally,  
84 all measurements are stationary laboratory measurements with maximum characteristics at only four Sites in the CIGP,  
85 specifically in the city (Lakra et al., 2025). This study is the first of its kind in the CIGP, presenting source  
86 apportionment of NR-PM<sub>2.5</sub> at a hyperlocal scale across three Sites in Lucknow, India, from post-monsoon to winter  
87 using VSAAQMP. Additionally, stationary van-based monitoring efforts remain limited nationwide (DeWitt et al.,  
88 2015; Kerckhoffs et al., 2025; Maslouski et al., 2023). The motivation for conducting this research over Lucknow  
89 from the fact that, as one of India's fastest-growing economies and its most populous state, Uttar Pradesh's rapid  
90 development makes it essential to gain insights into its air quality (PRS Legislative Research, 2025). This study aims  
91 to (1) characterize the chemical composition of NR-PM<sub>2.5</sub> aerosols at the hyperlocal scale. (2) identify and quantify  
92 source contributions using a bilinear model, (3) analyze the diurnal and temporal variability of sources, and (4)  
93 investigate the association between particle growth events and aerosol composition dynamics in a polluted urban  
94 environment. The novelty of the present work lies in its hyperlocal characterization of urban aerosol populations  
95 within the city scale, demonstrating that particle number and mass are dominated by traffic-related and secondary  
96 sources, and that subsequent particle growth is strongly driven by the interplay of organic and inorganic vapors. This  
97 type of hyperlocal measurement helps strategically target air pollution sources rather than implementing bulk  
98 measures. Additionally, more coordinated and systematic actions are essential to address air pollution in highly  
99 impacted areas. The research integrates source apportionment, elemental ratios, and long-range transport assessments

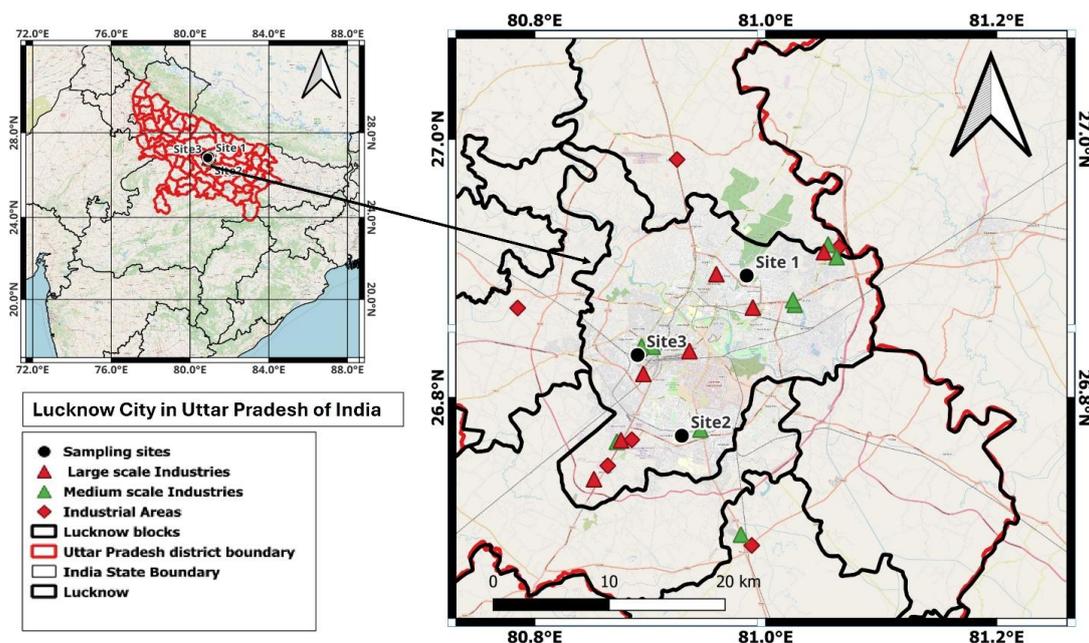


100 to understand pollution dynamics and spatial variability, providing insights essential for targeted mitigation and  
101 exposure reduction strategies in complex urban environments.

## 102 2. Study Site, Instrumentation, and Methodology:

### 103 2.1 Monitoring Sites for hyperlocal measurements.

104 Hyperlocal air quality monitoring using the VSAAQMP was conducted at three distinct locations in Lucknow: the  
105 Council of Scientific and Industrial Research–Central Institute of Medicinal and Aromatic Plants (CSIR-CIMAP)  
106 (26.89°N, 80.98°E), designated as the background Site (Lakra et al., 2025); Babasaheb Bhimrao Ambedkar University  
107 (BBAU) (26.77°N, 80.92°E), a traffic-influenced Site due to proximity to Shaheed Path; and Talkatora (26.83°N,  
108 80.88°E), classified as the industrial Site due to its proximity to the industrial area of Lucknow (Jain et al., 2024; Lakra  
109 et al., 2025). These designations were based on each Site's proximity to different pollution sources. The city  
110 experiences a continental climate, characterized by extremely hot summers with temperatures reaching up to 46 °C  
111 and cold winters with temperatures as low as 7 °C (IMD, 2012). According to recent projections, the population of  
112 Lucknow in 2025 is estimated to be around 4.13 million for the metropolitan area and about 4.12 million for the city  
113 itself (Jain et al., 2023; Lucknow, India Metro Area Population 1950-2025, 2025). Key pollutants, including  
114 particulate matter (PM<sub>2.5</sub> and PM<sub>10</sub>), carbon monoxide (CO), nitrogen dioxide (NO<sub>2</sub>), and sulfur dioxide (SO<sub>2</sub>),  
115 frequently exceed national ambient air quality standards in Lucknow (Jain et al., 2023; Pal et al., 2023). The first  
116 measurement Site, CSIR-CIMAP Lucknow, is located near the Kukrail forest in the northeastern part of the city,  
117 surrounded by a blend of urban development and green spaces. The Kukrail forest serves as a vital "green lung" for  
118 the area, significantly enhancing the local environment by absorbing carbon dioxide and pollutants through its dense  
119 tree cover, thereby helping reduce the urban heat island effect and combating climate change. The second  
120 measurement Site was BBAU (26.77°N, 80.92°E) in Lucknow, near Shaheed Path, a major expressway that  
121 significantly impacts the local environment. While this proximity improves accessibility and urban connectivity, it  
122 also increases air pollution from traffic in the campus vicinity. The area around BBAU features expanding residential  
123 neighborhoods, which further contribute to the urban landscape and affect local microclimate conditions. The presence  
124 of the Shaheed Path increases vehicular emissions, potentially compromising air quality near the university,  
125 particularly during peak traffic periods. When combined with emissions from surrounding residential areas, this  
126 contributes to moderate air pollution in the area. The third monitoring Site is the city's Talkatora Industrial Area. The  
127 VSAAQMP system has been deployed at the receptor location, situated downwind of the Site (26.83°N, 80.88°E).  
128 The Talkatora area of Lucknow, characterized by a concentration of small- and medium-sized industries, faces  
129 significant environmental challenges stemming from industrial operations. Its surroundings feature a mix of industrial  
130 estates and residential neighborhoods, forming a complex urban-industrial environment. Industrial activities in  
131 Talkatora are a major source of air pollution, often resulting in "severe" pollution levels. Hereafter, throughout this  
132 manuscript, CIMAP (26.89°N, 80.98°E), BBAU (26.77°N, 80.92°E), and Talkatora (26.83°N, 80.88°E) are referred  
133 to as Site 1, Site 2, and Site 3, respectively.



134

135 Figure 1. Showing the locations of the hyperlocal monitoring Sites in the city of Lucknow, Uttar Pradesh, India.

## 136 2.2 Sampling Period and Instrument Duration

137 The measurement was carried out from October to November 2023, during the post-monsoon season, across Lucknow,  
138 a major urban center in the CIGP. The measurement was carried out by deploying the VSAAQMP at a specific location  
139 for approximately 12-14 days, including 2-3 days for instrument calibration. However, the sampling duration has been  
140 extended in case of monitoring challenges, such as hardware failure, radio frequency interference to the AMS spectra,  
141 and van alignment with the road (Mushasha et al., 2024). Therefore, considering the above-mentioned challenges  
142 during the sampling period and filtering the noise from the measurement data, we have considered the measurement  
143 data for the CSIR-CIMAP for 6 days, i.e., from the 2<sup>nd</sup> of October 2023 to 7<sup>th</sup> October 2023, 11 days for 13<sup>th</sup> October  
144 2023 to 23<sup>rd</sup> October 2023, and 14 days for 6<sup>th</sup> November 2023 to 19<sup>th</sup> November 2023, respectively. All instruments  
145 were well installed, rack-mounted, stable, and positioned on a stand with an AMS over a three-degree-of-freedom  
146 spring stand system. For more details on the VSAAQMP, the reader can refer to our study on the source apportionment  
147 of elements across Lucknow city, using the VSAAQMP (Lakra et al., 2025). The van was equipped with an air  
148 conditioning system to maintain an optimal temperature, with thermally insulated walls to ensure proper functioning  
149 of all instruments. The subsequent section describes the specifications and working principles of each instrument  
150 utilized in the study.

151

### 152 2.2.1 HR-ToF-AMS



153 The Aerodyne Research Inc. HR-ToF-AMS was installed in a van to examine the chemical composition and size-  
154 resolved mass concentrations of NR-PM<sub>2.5</sub> particles at a 2-minute temporal resolution. The operational mechanisms of  
155 the HR-ToF-AMS are detailed in a research article (DeCarlo et al., 2006), and a brief overview is provided below.  
156 The inlet uses stainless-steel tubing (outer diameter ~8mm and inner diameter ~6 mm) to maintain a flow rate of 0.08  
157 L/min. It is connected to a silica gel diffusion dryer, which reduces moisture by 20% before the air enters the  
158 instrument's main sampling inlet (Bhattu et al., 2024; Chakraborty et al., 2016; Tobler et al., 2020). The silica gel  
159 beads are recharged every 5-6 days after each Ion Efficiency (IE) calibration at the start of sampling at each Site, to  
160 prevent humidity from impacting particle collection efficiency. After that, the ambient aerosols were passed through  
161 the critical orifice of ~100µm and entered the sizing chamber by an aerosol dynamic lens. In this chamber, a high-  
162 vacuum environment was maintained, and the particle size was determined by measuring the time it took for particles  
163 to travel a fixed distance. They were vaporized into a gaseous state. The resulting gas molecules are then ionized with  
164 an electron ionizer at 70 eV, and the ions are subsequently detected by a multi-channel plate (MCP) detector. Through  
165 the sampling period, the instrument is operated in the V-mode at all three Sites, characterized by the low resolving  
166 power( $m/\Delta m \approx 2500$  for  $m/z$  10-200,  $\Delta m$  is the full width half maximum, and  $m$  being the nominal  $m/z$  (DeCarlo et  
167 al., 2006) and high sensitivity (1-min detection limit of ~ 0.04 µgm<sup>-3</sup>). The instrument performed two cycles during  
168 the 2-minute sampling interval, each lasting 30 seconds. These cycles alternated between mass spectrum (MS) mode,  
169 which involved 15 seconds blocked and 15 seconds open, and particle time-of-flight (PToF) mode, which ran for 30  
170 seconds. Although we used MS data for our current analysis, we collected data in both MS and PToF modes to support  
171 potential future studies. The datasets obtained from the instrument from all three Sites were analyzed using  
172 SQUIRREL (SeQUential Igor data RetRiEvaL, version 1.66B) for Unit Mass Resolution (UMR) data and PIKA (Peak  
173 Integration by Key Analysis, version 1.26B) for high-resolution (HR) data. Both tools were implemented using IGOR  
174 Pro 9.0.5.4 (Wavemetrics, Inc., Portland, OR, USA), with a peak-fitting algorithm (DeCarlo et al., 2006) applied over  
175  $m/z$  ratios from 12 to 120. Air interference correction was performed by adjusting the fragmentation table at specific  
176 mass-to-charge ratios ( $m/z$ ) of 44, 34, 33, 32, 18, 16, and 12. This process utilized data from high-efficiency particulate  
177 air (HEPA) filters, which were frequently collected during the sampling period (Aiken et al., 2008; Allan et al., 2004).  
178 A collection efficiency (CE) value of 1 was used for the capture vaporizer, as recommended by Hu et al. (2017).  
179 Calibrations, including ionization efficiency (IE), were conducted at the beginning of each sampling Site. These  
180 calibrations followed the mass-based method using a condensation particle counter (CPC) and a scanning mobility  
181 particle analyzer (SMPS), as described by Jayne et al. (2000). The calibrations used 300 nm monodisperse particles  
182 of ammonium nitrate (NH<sub>4</sub>NO<sub>3</sub>) for IE calibration and ammonium sulfate (NH<sub>4</sub>(SO<sub>4</sub>)<sub>2</sub>) for RIE calibration with details  
183 provided in Table TS2 (a and b) of the supplementary information. Experimentally determined average relative  
184 ionization efficiencies (RIEs) for NH<sub>4</sub> and SO<sub>4</sub> at three Sites were 4.48 and 1.47, respectively. For chloride and  
185 organics, default RIE values of 1.3 and 1.4 were used in the present work. These defaults, established through  
186 laboratory calibrations with standards such as NH<sub>4</sub>Cl and organic compounds (e.g., levoglucosan), are widely used  
187 when Site-specific calibration is unavailable due to their robustness across ambient conditions and consistency with  
188 inter-instrument comparisons (Khare et al., 2025; Watson et al., 2020). They ensure the reliable quantification of non-



189 refractory species without overcomplicating the analysis, especially for low-concentration chlorides (often  $\text{NH}_4\text{Cl}$ ),  
190 where custom RIE determination may lack sufficient signal-to-noise (Nuaaman et al., 2015).

### 191 **2.2.2 Micro-Aethalometer:**

192 The real-time BC mass concentrations were measured using a laboratory-calibrated Micro-Aethalometer (AethLabs,  
193 model AE51), a self-contained device equipped with built-in pumps, a battery, an internal memory card for data  
194 storage, and flow control (AethLabs, 2016). This device is tailored for exposure monitoring and quantifies the rate of  
195 change in light absorption due to the continuous buildup of aerosols on a T60 Teflon-coated glass fiber filter.  
196 Measurements at 880nm are used as an indicator of atmospheric BC concentrations. The instrument was regularly  
197 monitored to ensure data accuracy. The maintenance routine included checking the flow rate, monitoring screen  
198 warnings, and replacing the AE51 filter every 4-5 hours to prevent aerosol overloading ( $\text{ATN} \leq 100$ ). This is important  
199 because high filter loading can cause BC levels to be underestimated (Weingartner et al., 2003). The instrument  
200 provides minute-by-minute high-resolution data, but we averaged over 30 minutes for analysis to reduce real-time  
201 data variability (Cheng and Lin, 2013). The laboratory-calibrated Micro-Aethalometer (AethLabs AE51) measures  
202 real-time BC mass with  $\pm 20\%$  accuracy,  $\pm 5\text{-}10\%$  precision, and  $\sim 1 \mu\text{g m}^{-3}$  detection limit (1-min average) (Alas et al.,  
203 2020). Uncertainties arise from filter loading (up to 70% bias without correction) and RH/T effects; apply by research  
204 (Virkkula et al., 2012) loading corrections for  $\pm 10\text{-}15\%$  Root mean square error (RMSE) (Good et al., 2018).  
205 Furthermore, the instrument was placed on a stable base to minimize vibrations, and sampling was conducted through  
206 a separate silicon tubing inlet, which was regularly cleaned with a foam spray to remove any blockages and maintain  
207 optimal sampling conditions. Moreover, previous studies have shown a robust correlation for both collocated  
208 measurements (Alas et al., 2019; Vernooij et al., 2022).

### 209 **2.2.3 SMPS**

210 The particle size distributions (12–672 nm) along with particle count were measured using a SMPS comprising a TSI  
211 model 3082 electrostatic classifier, a long Differential Mobility Analyzer (DMA), and a TSI model 3776 Condensation  
212 Particle Counter (CPC) (TSI Incorporated, 2006). More details can be found in the previous papers (Lakra et al., 2024;  
213 Mishra et al., 2023)

### 214 **2.2.4 Auxiliary Instruments and Parameters**

215 Data on external factors, including trace gases and meteorological parameters such as ambient temperature (AT) in  
216  $^{\circ}\text{C}$ , relative humidity (RH) in %, and wind speed (WS) in  $\text{ms}^{-1}$ , were measured using collocated operational instruments  
217 inside the van, including gas analyzers and Vaisala sensors. Measurements of trace gases, such as ozone ( $\text{O}_3$ ), sulfur  
218 dioxide ( $\text{SO}_2$ ), carbon monoxide (CO), and nitrogen dioxide ( $\text{NO}_2$ ), were measured using Thermo Scientific gas  
219 analyzers (models 49i, 43i, 48i, and 42i, respectively). All gas monitors were calibrated at the start of the campaign at  
220 each Site using a dynamic gas calibrator (model 146i). To validate the NR- $\text{PM}_{2.5}$  measurement data from the reference-  
221 grade  $\text{PM}_{2.5}$  monitor, Environment Beta Attenuation Monitor (E-BAM), manufactured by MetOne. The used monitor  
222 is USEPA-certified and uses the beta attenuation technique to measure  $\text{PM}_{2.5}$  particles (U.S. Environmental Protection



223 Agency, 2025). We have used the PM<sub>10</sub> inlet in conjunction with the PM<sub>2.5</sub> cyclone to robustly measure the PM<sub>2.5</sub>  
 224 mass concentration in μgm<sup>-3</sup>. The monitor used is a USEPA-certified device that employs the beta attenuation  
 225 technique to measure PM<sub>2.5</sub> mass concentration. The Planetary Boundary Layer Height (PBLH) data from MERRA-  
 226 2, with a spatial resolution of approximately 0.5° × 0.625° on a latitude-longitude grid and hourly temporal averaging  
 227 for the sampling period, have been extracted for the Sites of interest (Gelaro et al., 2017).

## 228 2.3 Source Apportionment Analysis (SAA)

### 229 2.3.1 SAA using PMF

230 Several receptor models are available for analyzing the characteristics of ambient aerosol measurements (Air Pollutant  
 231 Receptor Modeling, 2025; Watson et al., 2002). PMF with ME-2 engine was applied to the combined organic and  
 232 inorganic aerosol mass spectra obtained using a HR-ToF-AMS at all three Sites. The reason for using PMF coupled  
 233 with ME-2 was selected because it (i) uses an error-weighted, mass-conserving framework that incorporates individual  
 234 measurement uncertainties, (ii) enforces non-negativity on both source profiles and contributions, leading to  
 235 physically realistic factors, and (iii) allows the inclusion of a priori constraints in ME-2 to stabilize solutions and  
 236 separate closely collinear sources more robustly than traditional models such as chemical mass balance (CMB) or  
 237 unconstrained factor analysis (Paatero, 1999). The receptor model combines unconstrained factor exploration with a  
 238 priori profile constraints ( $\alpha = 0.02-0.1$ ), achieving >95% OA mass closure, 50-70% reduced rotational ambiguity, and  
 239 <20% uncertainty for major factors like HOA/OOA across Sites (Drosatou et al., 2019). It employs a bilinear unmixing  
 240 algorithm with non-negative constraints, utilizing distinctive factorization (Paatero and Tapper, 1994), which involves  
 241 iterative reweighting of the data to yield unique solutions, as shown in equation (1).

$$242 \quad x = \sum_{k=1}^p g_{ik} f_{kj} + e_{ij} \quad (1)$$

243 In this context,  $x_{ij}$  represents the concentration of the  $j^{\text{th}}$  species in the  $i^{\text{th}}$  sample. The source profile of the  $k^{\text{th}}$  source  
 244 to the  $j^{\text{th}}$  species is denoted by  $f_{kj}$ , while  $g_{kj}$  represents the time series of the  $j^{\text{th}}$  species in the  $k^{\text{th}}$  source. The residual  
 245 matrix is represented by  $e_{ij}$ , and the number of factors is denoted by  $p$ . To solve equation 1, the minimization of the  
 246 quantity  $Q$  (as defined in the equation) is performed. The measurement uncertainty determines this quantity  $Q$  ( $\sigma_{ij}$ )  
 247 and the elements of the residual matrix ( $e_{ij}$ ) defined in equation (2).

$$248 \quad Q = \sum_{i=1}^n \sum_{j=1}^m \left( \frac{e_{ij}}{\sigma_{ij}} \right) \quad (2)$$

249 Due to the rotational ambiguity inherent in PMF, multiple rotated solutions of the matrices  $F$  and  $G$  with similar  $Q$ -  
 250 values can emerge, potentially resulting in mixed-factor solutions. To address this, PMF (Paatero, 1999) was  
 251 implemented using the Source Finder interface (SoFi version 9.5.4.6, Datalystica Ltd., Villager, Switzerland) for  
 252 source apportionment analysis, as described in research articles (Canonaco et al., 2013; Crippa et al., 2014).  
 253 Additionally, the  $a$ -value in ME-2 was constrained to 0.5 for Sites 1 and 2 for the HOA factor. In this context, the  
 254  $a$ -value controls the allowed deviation of the optimized factor profiles from the selected reference profiles of HOA



255 (Mohr et al., 2012). The  $\alpha$ -value of 0.5 permits  $\pm 50\%$  relative variation around the anchor profile, providing flexibility  
256 to capture Site-specific variability while preventing unrealistic drift of the solution. The constraint was implemented  
257 by supplying the reference profiles to ME-2 and specifying values for the corresponding factors, following the standard  
258 constrained PMF implementation in the SoFi/ME-2 framework (Shukla et al., 2025).

259

### 260 **2.3.2 SAA of Combined Organic and Inorganic Aerosols**

261 Using half-hourly averaged HR-ToF-AMS data from all three Sites, the high-resolution ions ( $m/z$  12–120) were  
262 combined with unit mass resolution ions ( $m/z$  121–300) extracted from organic aerosol mass spectra, along with  
263 several major inorganic ions. The inorganic ions included SO ( $m/z$  47.97), SO<sub>2</sub> ( $m/z$  63.96), SO<sub>3</sub> ( $m/z$  79.96), HSO<sub>3</sub>  
264 ( $m/z$  80.96), H<sub>2</sub>SO<sub>4</sub> ( $m/z$  97.97), NH ( $m/z$  15.01), NH<sub>2</sub> ( $m/z$  16.02), NH<sub>3</sub> ( $m/z$  17.03), Cl ( $m/z$  35.98), HCl ( $m/z$  34.97),  
265 NO ( $m/z$  29.99), and NO<sub>2</sub> ( $m/z$  45.99). These ions were integrated to form the PMF input matrix, which consisted of  
266 64769 sample points and 513 variables for Site 1, 131060 sample points with 507 variables for Site 2, and 137,228  
267 sample points and 513 variables for Site 3, following procedures outlined in research articles (Sun et al., 2014; Ulbrich  
268 et al., 2009). Variables with a signal-to-noise ratio below two were down-weighted by a factor of 2, and those below  
269 0.2 by a factor of 10. CO<sub>2</sub>-related variables were excluded from the PMF analysis to prevent over-weighting the CO<sub>2</sub><sup>+</sup>  
270 variable, as these are calculated as constant fractions of the CO<sub>2</sub><sup>+</sup> signal in AMS data analysis (Lalchandani et al.,  
271 2022). This approach enabled the resolution of organic-rich factors, such as solid fuel combustion and biomass burning  
272 with diverse mass spectral characteristics, as well as traffic-related factors and oxygenated organic aerosol (OOA)  
273 factors, alongside inorganic-rich factors, at all three Sites. The combined organic and inorganic PMF resulted in clearly  
274 identified factors following the necessary selection of the optimal PMF solution (Lakra et al., 2024). The selection of  
275 factors was conducted according to the standard procedure followed in earlier studies (Akanksha et al., 2025;  
276 Bhowmik et al., 2022). For Site 1, PMF was explored for various factors, and a 7-factor solution was selected. For  
277 this configuration, repeated runs with different seeds and rotations yielded stable  $Q/Q_{\text{exp}}$  values of approximately 6–  
278 7, indicating a robust fit with minimal sensitivity to initialization. All seven factors (HOA, two BBOA factors,  
279 SV-OOA, LV-OOA, SO<sub>4</sub>-OA, and NO<sub>3</sub>-OA) showed physically interpretable profiles and time series, so this solution  
280 was adopted for further analysis. Similarly, the other factors are selected in a similar manner for other Sites.

### 281 **2.3.3 Uncertainty Evaluation in PMF Solutions via Bootstrapping Analysis**

282 To evaluate the uncertainty of the factor derived from the PMF model, a bootstrap resampling analysis (Paatero et al.,  
283 2014) was conducted in SoFi for each Site, with  $n = 1000$  iterations, employing criteria-based selection (Brown et al.,  
284 2015). A balance between computational and statistical efficiency was achieved by using the initial 20 seed runs,  
285 which demonstrated good reproducibility of results across all three Sites (Brown et al., 2015; Cash et al., 2023;  
286 Srivastava et al., 2018; Vlachou et al., 2018). The base case for Site 1 was established from a bootstrapped time series  
287 of seven factors, including one constrained HOA factor and six unconstrained factors. For Site 2, the base case  
288 comprised six factors (one HOA-constrained and five unconstrained), while Site 3 used seven fully unconstrained



289 factors. For each Site, Pearson's correlation coefficient (R) was calculated between the base-case factor time series  
290 and the randomly positioned bootstrap factors, and these R values were used to define the criteria scores. In SoFi, a  
291 criterion that is used to reposition and sort unconstrained factors is treated as an active criterion, whereas  
292 a passive criterion only reports the score for the current factor position and does not influence the sorting; this is  
293 typically applied to constrained factors, whose position is already known a priori, and for which criteria are used only  
294 for monitoring. Active or passive status is assigned by the user in the criteria panel, where selected (highlighted)  
295 criteria are active, and all others remain passive. In this study, correlation-based criteria were applied to unconstrained  
296 factors, and factors with active criterion R values greater than 0.8 and statistically non-significant differences from  
297 the base case (p-values above 0.05 from the t-test) were retained as the final robust factors for interpretation (Rai et  
298 al., 2020). Over 90% of the unmixed (unconstrained) factors achieved correlation scores greater than 0.8 at all three  
299 Sites, and solutions with 7, 6, and 7 factors were retained for Sites 1, 2, and 3, respectively. For Site 1, the constrained  
300 HOA factor also met the selection criterion, with approximately 50% of its bootstrap realizations exceeding the  $R >$   
301 0.8 threshold, consistent with its weaker, more mixed traffic signature at this background Site (Shukla et al., 2021).

#### 302 **2.4 Analysis of Back Trajectories and Concentrated Weighted Trajectories**

303 Trajstat model v4.1 was used to conduct back trajectory analysis, tracing the origins and pathways of sources  
304 influencing NR-PM<sub>2.5</sub> species and combining inorganic and organic aerosols. It uses the Global Data Assimilation  
305 System (GDAS) weekly files, which provide data at 1-degree resolution in latitude and longitude. The backward  
306 trajectories were computed every 3 hours at 500 m above ground level, thereby incorporating the average Planetary  
307 Boundary Layer Height (PBLH). These trajectories were then weighted using time-series data that combined organic  
308 and inorganic factors. Since similar concentration patterns were observed at given altitudes, the 500m altitude was  
309 chosen to ensure that the backward trajectories remained within the mixing layer (Ghosh et al., 2015; Jain et al., 2023;  
310 Rai et al., 2020). Further, the Concentrated Weighted Trajectory (CWT) model was applied to each source factor using  
311 3-hour averaged time-series data. This averaging ensured compatibility with the GDAS output interval required for  
312 CWT analysis, which was performed using Zefir, an Igor-based interface, to identify air parcels contributing to  
313 elevated concentrations at the sampling Site (Petit et al., 2017), using equation (3).

$$314 \quad CWT_{ij} = \frac{1}{\tau_{ij}} \sum_{k=1}^N C_k \tau_{jk} \quad (3)$$

315 In this context, j and i represent longitude and latitude, respectively. The average concentration of the weighted  
316 trajectory in the ij<sup>th</sup> cell is denoted by CWT<sub>ij</sub> (in  $\mu\text{g m}^{-3}$ ). Here, k is the trajectory index, N is the total number of  
317 trajectories,  $\tau_{ijk}$  is the residence time of trajectory k in the ij<sup>th</sup> cell, and  $C_k$  (in  $\mu\text{g m}^{-3}$ ) is the measured concentration of  
318 the factor associated with trajectory k.

#### 319 **2.5 Estimation of the particle growth rates and condensation sink**

320 We estimated the particle growth rates for sizes ranging from 12 nm to 672 nm using SMPS data collected across the  
321 three Sites. The growth rate is calculated using equation (4) is provided below



322  $GR = \frac{\Delta d}{\Delta h}$  (4)

323 Here,  $d$  represents the geometric mean diameter, and  $h$  denotes the measurement time in hours (Pushpawela et al.,  
 324 2018). The particle growth rate is measured in nanometers per hour(nm/hr). The condensation sink (CS) was  
 325 determined from the particle number size distribution using equation (5).

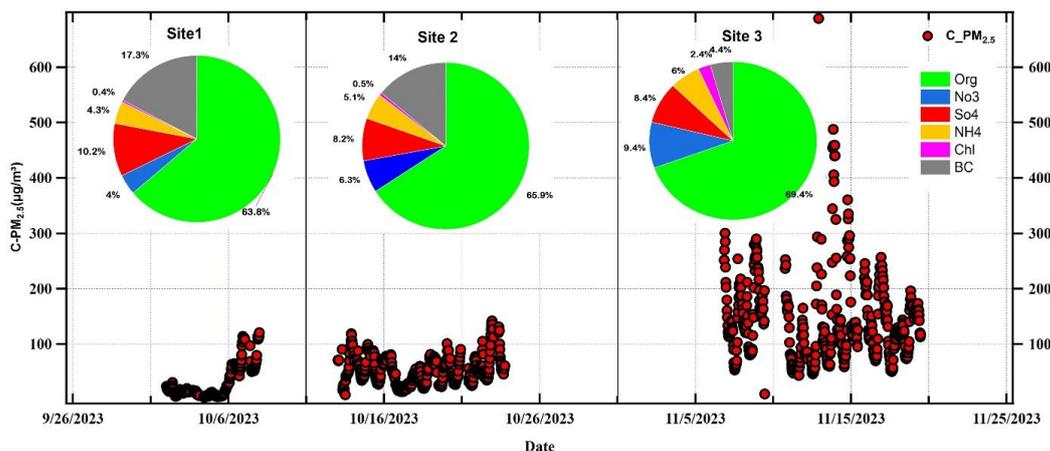
326  $CS = 2\pi D \sum \beta_i N_i d_i$  (5)

327 where  $D$  is the diffusion coefficient of the vapor,  $\beta_i$  is the Fuchs–Sutugin correction factor in  $\text{cm}^2\text{s}^{-1}$   $d_i$  is the diameter  
 328 of particles in the size bin  $i$  in nm, and  $N_i$  represents their number concentration(Tuovinen et al., 2020). The  
 329 calculation assumes that vapor condensation onto aerosol particles can be approximated by condensation onto  
 330 stationary particles. This assumption holds when particle sizes are significantly larger than those of vapor molecules,  
 331 and their diffusion is correspondingly slower. Here we have considered the approximate value of  $0.07 \text{ cm}^2\text{s}^{-1}$   
 332 , and 1, for the  $D$  and  $\beta_i$  respectively (Wang et al., 2020).

333

334 **3. Results and Discussion:**

335 **3.1 PM<sub>2.5</sub> Composition during campaign using MAQMS.**



336

**Figure.2: Time series of the C-PM<sub>2.5</sub> (µg/m<sup>3</sup>) (NR-PM<sub>2.5</sub> + BC) with metals over Site1 (CSIR -CIMAP), Site2 (BBAU) and Site3 (Talkatora) during the measurement period.**

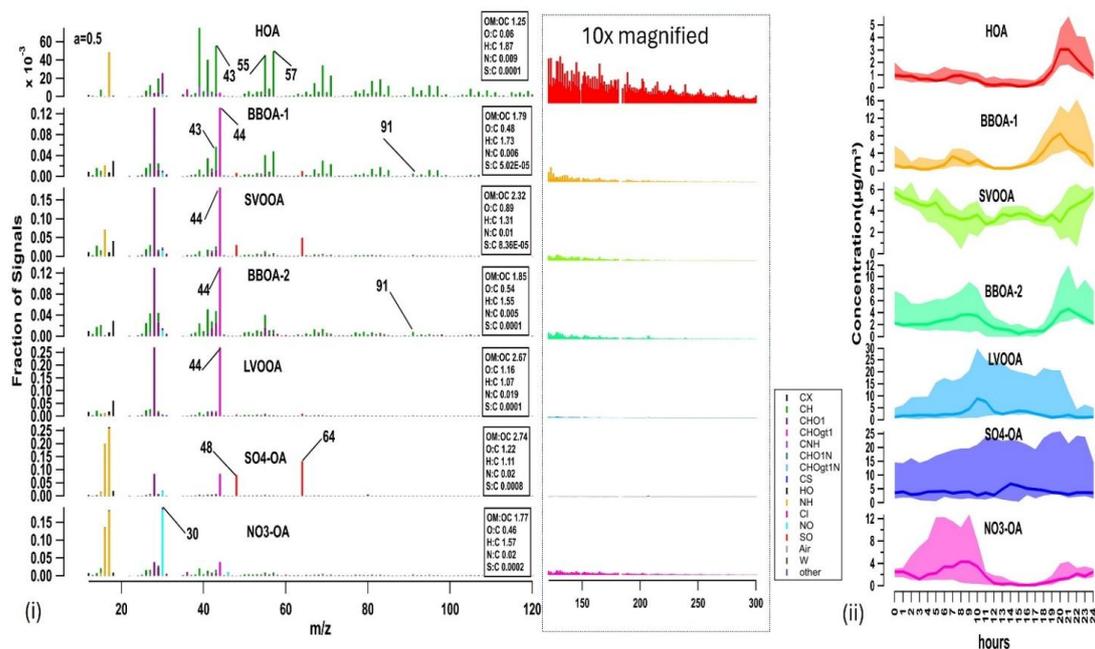
337 Figure.2 shows the temporal variation of C-PM<sub>2.5</sub> recorded during on-site air quality measurements by a VSAAQMP  
 338 from October 2 to November 19, 2023, across three locations in Lucknow. The average PM<sub>2.5</sub> concentrations recorded  
 339 during the campaign at Sites 1, 2, and 3 were  $32.6 \pm 33.8 \text{ µg/m}^3$ ,  $58.31 \pm 26.9 \text{ µg/m}^3$ , and  $227.20 \pm 138.96 \text{ µg/m}^3$ ,  
 340 respectively. The composition-based PM<sub>2.5</sub> estimate (C-PM<sub>2.5</sub>), calculated as the sum of NR-PM<sub>2.5</sub>, black carbon (BC),



341 and metal components, was validated by correlating with reference-grade PM<sub>2.5</sub> measurements obtained from an  
342 EBAM (PM<sub>2.5</sub>-EBAM) (Lakra et al., 2024).

343 Figure S1a-c shows the linear curve fitting between the C-PM<sub>2.5</sub> (NR-PM<sub>2.5</sub> + metals fraction of PM<sub>2.5</sub>) and PM<sub>2.5</sub> -  
344 EBAM. We observed strong correlations between C-PM<sub>2.5</sub> and PM<sub>2.5</sub>-EBAM ( $R^2 = 0.97$  for Site 1, 0.86 for Site 2, and  
345 0.90 for Site 3), indicating good agreement in the temporal variation of C-PM<sub>2.5</sub> with PM<sub>2.5</sub>. The C-PM<sub>2.5</sub> mass was  
346 lower than the PM<sub>2.5</sub>-EBAM values, likely due to the slight underestimation of NR-PM<sub>2.5</sub> by the AMS and the  
347 exclusion of metal oxides in Xact measurements, which report only elemental metals (Lalchandani et al., 2022).  
348 Further, the study indicates that Sites 1 and 2—representing background and traffic-residential environments—  
349 experience comparatively lower anthropogenic influence than Site 3(i.e., C-PM<sub>2.5</sub> about 7 times that of Site 1). The  
350 industrial activities at Site 3, combined with stagnant meteorological conditions ( $T = 22.1$  °C,  $RH = 70.67\%$ , and  
351  $PBLH = 594.94$  m), contribute to the enhanced concentrations of both organic and inorganic aerosols observed at this  
352 Site (Fig. S4b). In contrast, the relatively higher BC levels (~14% ) at Site 1 are attributed to periodic wood and  
353 biomass burning associated with a lemongrass filtration process carried out for institutional research and commercial  
354 purposes (CSIR-CIMAP, 2025) (Fig. S10a). This activity, which occurred about 1.5km from the van, is also captured  
355 in our measurements during the last days of monitoring at Site 1 (6-7 October 2023), which are treated as the Polluted  
356 Day (PD) (Fig. S1ii). We have noted a significant concentration of OA (64%, 66%, and 70%) at all three Sites(Figure  
357 2). This trend has also been observed at numerous locations throughout India and worldwide (Akanksha et al., 2025;  
358 Lalchandani et al., 2021; Shukla et al., 2025; Tsimpidi et al., 2024). Additionally, we observed concentrations of NO<sub>3</sub>(  
359 4%,6%, and 9%), SO<sub>4</sub> (10%,8%, and 9%), NH<sub>4</sub> ( 4%,5%, and 6%), Chl( 1%,1%, and 2%) and BC ( 17%,14%, and  
360 4%) at Site 1, Site 2, and Site 3, respectively as shown in Figure 2. In the background Site 1 (post-monsoon), sulfate  
361 constitutes (~10%) a relatively larger share of the inorganic aerosol, and nitrate remains modest, consistent with a  
362 regional sulfate dominance, and a substantial fraction of nitric acid stays in the gas phase, limiting ammonium nitrate  
363 formation. At Site 2 (also post-monsoon), locally high NO<sub>x</sub> and NH<sub>3</sub> emissions enhance ammonium nitrate formation  
364 compared with Site 1, but sulfate still represents an important sink for ammonia, so nitrate fractions are higher than at  
365 the background site yet remain below typical winter values. At Site 3 (winter), lower temperatures favor partitioning  
366 of nitric acid to the particle phase and, together with strong sulfuric-acid production from industrial/coal emissions,  
367 drive concurrent formation of ammonium sulfate and nitrate; this both increases total inorganic mass and elevates  
368 nitrate and sulfate fractions relative to the post-monsoon background and traffic Sites (Bhandari et al., 2020; Tsimpidi  
369 et al., 2024).

### 370 **3.2 Source Apportionment of combined organic and inorganic aerosols**



371

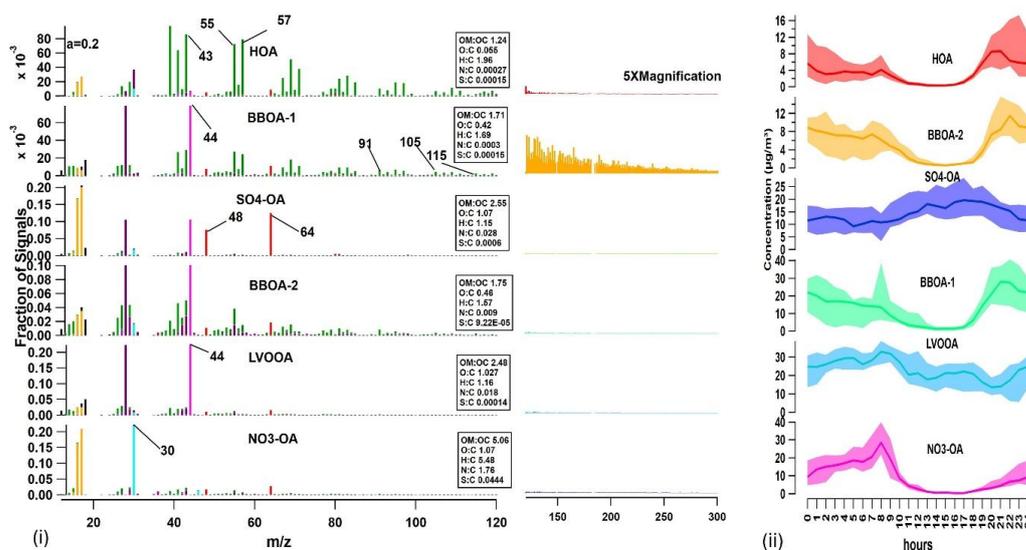
372 **Figure 3. (i) Mass spectral profiles of sources/factors obtained from the combined Organics and Inorganics sources**  
 373 **apportionment for m/z 12 to m/z 115, along with UMR spectra highlighted inside a grey border for m/z 120 to m/z 300.**  
 374 **Elemental Ratios (O/C, OM/OC, H/C, and N/C) have been calculated using only the organic signals, and (ii) Diurnal**  
 375 **variation of obtained sources during the measurement period over Site 1.**

376 The source factors were derived at three characteristically distinct sites in Lucknow. By applying a combined organic–  
 377 inorganic Positive Matrix Factorization (PMF) approach, aerosol sources were resolved into primary and secondary  
 378 organic components as well as inorganic species. Primary organic aerosols (POA) included hydrocarbon-like organic  
 379 aerosol (HOA), representing traffic emissions, and biomass-burning organic aerosol (BBOA), indicating direct  
 380 combustion sources. Solid fuel combustion (SFC) was identified as an additional primary organic marker, particularly  
 381 evident at Site 3, and serves as an indicator of wood-burning activities. Secondary organic aerosols (SOA) were  
 382 characterized by two OOA factors - low-volatile OOA (LVOOA) and semi-volatile OOA (SVOOA) representing  
 383 oxidized organic-rich components derived from regional and urban photochemical processes. Inorganic-rich factors  
 384 included sulfate-related organic aerosol (SO4-OA) and nitrate-related OA (NO3-OA), both formed via secondary  
 385 oxidation and acid-base equilibria (Tsimpidi et al., 2024). The factor composition varied substantially across the three  
 386 sampling Sites: Site 1 resolved two BBOA and one HOA factor alongside secondary OOA factors, reflecting mixed  
 387 primary and secondary pollution; Site 2 was characterized predominantly by OOA factors with strong inorganic  
 388 associations, indicating aged and processed aerosols; and Site 3 was dominated by the SFC factor, indicating solid  
 389 fuel combustion as the primary pollution source during the sampling period. To gain insights into atmospheric  
 390 composition from inter-site variation, we discuss these factors in the following sections.

391 **3.2.1 Factor abundant in organic and inorganic compounds:**



392 **3.2.1.1 HOA and SFC-OA:** The mass spectra of HOA were predominantly composed of the hydrocarbon ion series  
 393 of  $C_nH^{+2n-1}$  and  $C_nH^{+2n+1}$ , which characterizes the HOA spectrum. The  $C_nH^{+2n-1}$  includes  $m/z$  such as  $m/z$   
 394 41( $C_3H_5$ ), 55( $C_4H_9$ ), 69( $C_5H_9$ ) as these are typical tracers of unsaturated hydrocarbon. The identification of alkane  
 395 fragments ( $C_nH_{2n+1}$ ) at  $m/z$  43 ( $C_3H_7^+$ ), 71 ( $C_5H_{11}^+$ ), and 98 ( $C_7H_{12}^+$ ), along with polycyclic aromatic hydrocarbon  
 396 (PAH) fragments such as  $m/z$  131, as traffic-related organic markers in aerosol mass spectrometry, is documented in  
 397 several key references (Aiken et al., 2008; Lalchandani et al., 2022; Sun et al., 2012). The O: C ratio is observed to be  
 398 nearly 0.05 at Site 1 and Site 2, which aligns with the values reported in a recent study over Lucknow (Lakra et al.,  
 399 2024) (Figs. 3 and 4i–ii). Not only that, but worldwide studies have reported a lower O: C ratio of  $\sim 0.04$  (Aiken et al.,  
 400 2008; Sun et al., 2012). Additionally, the O: C ratio at Site 3 is 0.15, which may be attributed to a greater contribution  
 401 from CHO groups (Fig. 5i–ii). This indicates the secondary contributions, as indicated by the  $m/z$  signal, all  $C_xH_yO$  ions  
 402 with  $z > 1$  Figure. 3 (i). Moreover, the HOA factor is further confirmed by its strong correlation with  $NO_x$ , with R  
 403 values of 0.73, 0.83, and 0.48 (Fig. S8a-c). HOA levels from primary sources should be higher in the early morning  
 404 or at night, as reported in earlier studies (Kumar et al., 2022; Lalchandani et al., 2021). Here, we have also observed  
 405 bimodal peaks in the HOA variation during morning and nighttime hours. At Site 1, we have observed a very small  
 406 variation in HOA, attributed to the lesser traffic influence. However, Site 2 experiences heavy traffic influence,  
 407 resulting in significant observed variations. On the other hand, Site 3 exhibited greater variability, likely because its  
 408 measurements were conducted during winter, when the PBLH is particularly shallow in the early morning and  
 409 nighttime hours. We observed the highest HOA contribution in Site 3 ( $\sim 6.9\%$ ), followed by Site 2 ( $\sim 5.7\%$ ) and Site 1  
 410 ( $\sim 3.5\%$ ) (Fig. 6-8(ii)). Lower



411

412 **Figure 4. i) Mass spectral profiles of sources/factors obtained from the combined Organics and Inorganics sources**  
 413 **apportionment for  $m/z$  12 to  $m/z$  115, along with UMR spectra highlighted inside a grey border for  $m/z$  120 to  $m/z$  300.**  
 414 **Elemental ratios (O/C, OM/OC, H/C, and N/C) have been calculated using only the organic signals, and ii) Diurnal variation**  
 415 **of the obtained sources during the measurement period over Site2.**



416 HOA at traffic-influenced Site 2 may result from higher PBLH and post-monsoon conditions, while elevated HOA at  
417 Site 3 likely stems from winter stagnation and lower PBLH(average ~594m) (Kumar et al., 2022).

418 The time-series variation of the HOA at site 1 shows smoother variation, indicating a lesser influence of traffic activity  
419 and a more urban background nature at site 1 (Fig. 6(i)). At Site 2, the HOA shows periodic variation, capturing daily  
420 traffic activity at the site; however, we observed a 30% decrease in HOA concentration within the university campus  
421 on every Wednesday, which, on 18th October 2023, was a no-diesel/petrol vehicle use day(Fig.7(i)). The time-series  
422 variation of the daily HOA values at site 3 shows higher variability, with a maximum value of  $50 \mu\text{g m}^{-3}$  (Fig. 8(i)).

423 As far as the regional and local transport of these source factors is concerned, using the CWT, Figure. S9(i) panel (a)  
424 shows relatively low to moderate HOA source influence, with a narrow plume extending along the Indo-Gangetic  
425 Plain but weaker intensities near the receptor. This suggests that Site 1 is affected by more regional/transported traffic  
426 and combustion emissions rather than strong local HOA sources. Moreover, for Site 2, the CWT plot in Figure.  
427 S9(ii)(a) displays a more compact and intense HOA plume over north-central India, centered closer to the receptor  
428 point. The higher CWT values indicate stronger contributions from regional traffic/combustion corridors upwind of  
429 Site 2, consistent with its greater local traffic influence. On the other hand, Figure. S9(iii)(d) shows the strongest and  
430 most localized HOA concentrations, with high CWT values tightly concentrated around the receptor region. This  
431 implies dominant contributions from nearby traffic and combustion sources (e.g., roadside emissions at the downwind  
432 Talkatora location), making Site 3 the most impacted by HOA among the three Sites.

433 The SFC-OA factor exclusively resolved at Site 3 exhibits diagnostic m/z peaks: oxygenated fragments at m/z 29  
434 ( $\text{CHO}^+$ ), 39 ( $\text{C}_3\text{H}_3^+$ ), 41 ( $\text{C}_2\text{HO}^+$ ); unsaturated hydrocarbons at m/z 50 ( $\text{C}_4\text{H}_2^+$ ), 51 ( $\text{C}_4\text{H}_3^+$ ), 53 ( $\text{C}_4\text{H}_5^+$ ); aromatic  
435 tracers at m/z 77 ( $\text{C}_6\text{H}_5^+$ ), 91 ( $\text{C}_7\text{H}_7^+$ ); high m/z PAHs at m/z 105, 107, 115, 128, 152, 165, 178, 189, 202; and  
436 nitrogen-containing organics at m/z 42 ( $\text{C}_2\text{H}_4\text{N}^+$ ), m/z 45 ( $\text{CH}_3\text{NO}^+$ ), m/z 71 ( $\text{C}_3\text{H}_7\text{NO}^+$ ), m/z 73 ( $\text{C}_3\text{H}_9\text{NO}^+$ ),  
437 reflecting incomplete combustion and NOx incorporation during solid fuel burning. We observed elevated SFC  
438 concentrations at night (20-23h), attributed to a shallow PBL with a mean height of ~266m and thus lesser vertical  
439 mixing during the night. The heat map confirms the SFC-OA shows a good Pearson coefficient,  $r = 0.77, 0.78,$  and  
440  $0.98$  with m/z 77,91, and 115, respectively (Fig. S8c), which are the key tracers for incomplete combustion at Site 3  
441 (Lakra et al., 2024; Mohr et al., 2012) (Fig. S8c). The strong relationship between key tracers and SFC-OA confirms  
442 that the source factor is primarily linked to solid-fuel combustion. Moreover, the diurnal correlation of SFC-OA with  
443 BC ( $r = 0.39$ ) further indicates the contribution of SFC-OA from solid fuel combustion activities near Site 3 (Fig.  
444 5(i)). We have observed that the SFC-OA contribution accounts for about 10% of all OA-resolved factors at Site 3  
445 (Fig.8(ii)). The diurnal profiles of CO and SFC-OA show a strong correlation ( $r = 0.91$ ), supporting their shared origin  
446 from incomplete combustion processes (Tong et al., 2021). We also observed a quite high Pearson correlation of 0.51  
447 between RH and SFC. High RH enhances SFC-OA by driving semi-volatile combustion products into the particle  
448 phase and promoting aqueous/heterogeneous processing on wet aerosols (Tobler et al., 2021). Under shallow, humid  
449 boundary-layer conditions (e.g., winter nights and early mornings), when domestic heating and biomass burning are  
450 more prevalent, and dispersion is limited, this results in elevated SFC-OA (Tong et al., 2021). The time-series variation  
451 of the SFC-OA shows a peak whenever there is solid-fuel combustion activity near the site during the measurement



452 period(Fig.8(ii)). The CWT plot for the SVOOA (Fig.S9(iii)) shows the major contribution with concentrations 10-  
453  $16\mu\text{gm}^{-3}$  from local activities near Site 3.

#### 454 3.2.1.2 LVOOA:

455 In AMS source apportionment, Low-volatile-OOA (LVOOA) is an aged, highly oxidized SOA factor, typically  
456 interpreted as regionally transported or long-aged OA rather than fresh local emissions. LVOOA is characterized by  
457 a dominant peak at  $m/z$  44 ( $\text{CO}_2^+$ ), often accompanied by prominent peaks at  $m/z$  28 ( $\text{CO}^+$ ) and 18 ( $\text{H}_2\text{O}^+$ ), reflecting  
458 high oxidation. Higher O: C and oxidation state than SV-OOA and primary factors (HOA, SFC-OA, BBOA), which  
459 vary between 1.02 and 1.16 across the three Sites, indicating its highly oxidized or aged nature (Fig.3-d). We observed  
460 that LV-OOA accounted for 24.0%, 31.6%, and 17.9% of the total organic and inorganic aerosol at Sites 1, 2, and 3,  
461 respectively (Fig.6-8(ii)). Our findings at the urban Sites are in line with concentrations reported in urban European  
462 cities, which have shown that LVOOA commonly contributes ~30-40% of OA (Canonaco et al., 2015; Reyes-villegas  
463 et al., 2016).

464 At Site 1, we have observed a flat diurnal profile compared to Sites 2 and 3, with only a modest afternoon increase,  
465 reflecting photochemical production and mixing, and thus an aged LVOOA contribution(Kumar et al., 2022). The  
466 diurnal profile at Site 1 shows that LVOOA concentrations remain elevated from 10:00 to 19:00, indicating SOA  
467 formation through photochemical oxidation during daytime hours (Fig. 3 (i)). The lesser variation we observed for  
468 the larger daytime span (10:00-19:00 hours) at Site 1, with nearly 50% lower concentration, suggests that it is of a  
469 background nature. During the night hours, a fall after a small rise in the LVOOA over Site 1 was observed; on the  
470 other hand, a rise in the LVOOA occurred after a dip in concentration at 20:00 hours at Sites 2 and 3, respectively. At  
471 night, it is common to see LVOOA decrease at cleaner/background Sites and increase at traffic/industry Sites, because  
472 nighttime chemistry and boundary-layer dynamics favor local primary and semi-volatile emissions in polluted areas,  
473 while cleaner Sites mainly see regional background (Lalchandani et al., 2021; Liu et al., 2024b).

474 The time series variation of the LVOOA shows the smooth variation with almost 50% lesser concentration as  
475 compared to the Sites 2 and 3, respectively (Fig.6-8(ii)).

476 At Site 1, LVOOA's  $r = 0.41$  with  $\text{NO}_3$  (vs. 0.22 with  $\text{SO}_4$ ) arises from shared nocturnal partitioning, nitrate radical  
477 oxidation, and regional ammonium nitrate transport dominating over stable sulfate in post-monsoon conditions  
478 (Bhandari et al., 2020) (Fig. S8a).

479 The  $\text{NO}_3$  drives LVOOA via nighttime  $\text{NO}_3$  radical reactions on VOCs (common in urban backgrounds), yielding a  
480 higher  $r=0.41$  despite regional origins.  
481 The  $\text{SO}_4$ 's weaker  $r = 0.22$  reflects the decoupling of its aqueous-phase formation from LVOOA's gas-phase oxidation  
482 steps. This pattern reflects nitrate's semi-volatility, aligning with LVOOA formation, while sulfate's regional aqueous  
483 processing decouples temporally. A similar nitrate preference in oxygenated OA is observed in Delhi PMF studies  
484 (Bhandari et al., 2020; Cash et al., 2021). This was further confirmed by the CWT of LVOOA for Site 1, as a good  
485 relation with nitrate might be due to IGP transport of nitrate-rich plumes. In line with the above, we observed a weaker



486 correlation with sulfate at Site 2 ( $R = 0.30$ ). However, at Site 2, a higher LVOOA–nitrate ( $R=0.54$ ) than LVOOA–  
487 sulfate ( $R=0.30$ ) correlation implies that LVOOA variability is more closely linked to local/near-regional nitrate  
488 formation than at the background Site 1 (Fig. S8a). At Site 3, weaker correlations with both sulfate and nitrate ( $R=0.37$   
489 and  $0.25$ ) suggest a more heterogeneous mixture of industrial plumes and regional background, which reduces the  
490 simple LVOOA–sulfate coupling seen at the background Site (Fig. S8(b-c)) (Hao et al., 2014). Furthermore, the above  
491 pattern aligns with the CWT spatial pattern, in which Site 1 receives contributions from long-range transport from  
492 Punjab and Haryana during the biomass burning period (Fig. S9(i) and (e)). The Site 2, LVOOA concentration gradient  
493 is more local to the Site 2 (Fig. S9(ii),c). Moreover, at Site 3, LVOOA concentration contributed more from both local  
494 and distant sources (Fig. S9(ii) (g)).

### 495 3.2.2.3 SVOOA:

496 The less oxidized SVOOA (semi-volatile oxygenated organic aerosol), typically characterized by higher  $f_{43}$  ( $m/z$  43,  
497  $C_2H_3O^+$ ) and lower  $f_{44}$  ( $m/z$  44,  $CO_2^+$ ) signals relative to LVOOA represent fresher SOA in standard classifications.  
498 However, our hyperlocal Sites 1 and 3 show the reverse— $f_{44} > f_{43}$ —indicating relatively aged SVOOA despite its  
499 semi-volatile nature (Fig. 3i, iii)(Sun et al., 2012), with Site 3 exhibiting a particularly prominent  $(CHO)_1^+$  group at  
500  $m/z$  44 as a classic aging marker. PMF analysis resolved SVOOA exclusively at Sites 1 (10.1% of OA) and 3 (18.6%  
501 of OA)(Fig.6 and 8 (ii)), aligning with background values (15–35% of OA) and urban environments (~20–40% of  
502 OA)(Budisulistiorini et al., 2016; Minguillón et al., 2015; Ripoll et al., 2015; Xu et al., 2019) Site 1 showed a higher  
503 O: C ratio (0.89 vs. 0.58 at Site 3). This pattern mirrors Barcelona's SV-OOA, featuring high  $f_{44}$  from carboxylic acids  
504 alongside hydrocarbon fragments ( $m/z$  29, 43, 55, 57)(Mohr et al., 2012; Zhang et al., 2014). Moreover, in some  
505 ambient and chamber cases, spectra of fresher SOA or SV-OOA already show a strong  $m/z$  44 peak due to carboxylic  
506 acids. They can show  $f_{44}$  comparable to or larger than  $f_{43}$ , while still being clearly less oxidized than LV-OOA in O:  
507 C and overall pattern(Canonaco et al., 2015; Zhang et al., 2014).

508 SVOOA at Site 1 exhibits multimodal variation dominated by nighttime peaks. The strong midnight maximum reflects  
509 efficient  $NO_3/O_3$ -driven and aqueous-phase SOA formation under humid (~80%) conditions and shallow boundary  
510 layer (~324 m)(Gu et al., 2023). A weaker afternoon peak indicates diluted daytime photochemistry, while a fairly flat  
511 overall variation suggests the presence of aged SVOOA. At Site 1, SVOOA shows much lower daily temporal  
512 variability throughout the measurement period (Fig. 6 (i)). Moreover, at Site 3, the elevated values range from 10 to  
513  $80 \mu g m^{-3}$ . As time progresses, the variation becomes smoother (Fig. 8(i)).In contrast, Site 3 shows bimodal variation  
514 with a primary peak between 9:00 and 10:00 h. This daytime maximum is linked to active photochemical oxidation,  
515 boundary-layer rise, and nearby combustion (Gu et al., 2023; Kuang et al., 2020). A secondary late-afternoon peak  
516 arises from limited winter photochemistry.

517 Site 1 demonstrates nighttime dominance of nitrate over aqueous species. Site 3 reflects daytime photochemistry  
518 control, consistent with central IGP winter studies(Lakra et al., 2024). Supporting correlations include strong  $NO_3$  ( $r$   
519 = 0.83 vs. 0.52), moderate  $NH_4^+$  ( $r = 0.20-0.31$ ), and weak  $SO_4$  linkage. SVOOA typically couples to fresh  
520 traffic/anthropogenic SOA via  $NO_x$  chemistry. At a clean Site 1, aging processes transform POA into SVOOA, leading



521 to an increase in organic nitrate fractions, while inorganic nitrate and sulfate levels may decline due to volatilization  
522 or limited precursor availability (Wu et al., 2024). Another reason could be differences in volatility in the clean  
523 environment; inorganic nitrate is more volatile and tends to evaporate under warmer conditions. Site 1 has the highest  
524 temperature among all Sites, at  $\sim 29.6$  °C during the sampling period. Therefore, it reduces its particle-phase  
525 concentration (Matsumoto and Tanaka, 1996; Zhang and McMurry, 1992). The above-mentioned SVOOA reason for  
526 Sites 1 and 3 was also confirmed by the CWT plots, which explained their different origins.

527 The CWT plots (Fig. S9i (f–g) and c) reveal long-range transport of SVOOA from northern India to Site 1, enhanced  
528 by high temperatures ( $\sim 29.6$ °C) volatilizing inorganic nitrate/sulfate, while aging promotes organic nitrates, versus  
529 steady traffic/industry formation at Site 3 under stagnant winter conditions (Fig. S9iii (a)). In line with our observation  
530 at Site 1, the studies reported that overclean sites often originate from biogenic VOCs oxidized by nitrate radicals,  
531 producing organic nitrates and semi-volatile organics that do not necessarily co-occur with inorganic nitrates and  
532 sulfates, which are more anthropogenic or long-range transported (Fry et al., 2014; Song et al., 2024).

#### 533 3.2.2.4 BBOA-1 ,BBOA-2 and O-BBOA

534 BBOA was resolved into three related factors, each with characteristics that vary across the three Sites (Lakra et al.,  
535 2024). At Site 1, BBOA-1 represents fresh, less oxidized biomass-burning emissions, with relatively low signal at  $m/z$   
536 43 and 44 but enhanced high- $m/z$  ( $>120$ ) fragments characteristic of PAHs and incomplete combustion (Fig. 3), and  
537 strong anhydrous fragments at  $m/z$  60 and 73 indicating levoglucosan-like tracers (Daellenbach et al., 2017). Its O: C  
538 ratio ( $\approx 0.48$ ) is lower than that of the more aged BBOA-2, (Florou et al., 2017) and it contributes about 10.1% of  
539 OA (Fig. 6(ii)), showing a clear early-morning peak (07:00–08:00) (Fig. S3 g) linked to residential wood/coal use,  
540 agricultural burning, and trapping in a shallow boundary layer (Zhang et al., 2015). Its correlation with nitrate is weak  
541 ( $r = 0.19$ ), consistent with decoupled primary biomass-burning emissions (Fig. S8a). BBOA-2 at Site 1 is more  
542 oxidized (O: C  $\approx 0.54$ ), with enhanced  $m/z$  43 and 44 and much lower PAH intensities, contributes about 12.9% of  
543 OA (Fig. 6(ii)), peaks 1–2 hours after BBOA-1 (around 09:00–10:00), and correlates strongly with nitrate ( $r \approx 0.69$ ),  
544 implying rapid oxidation of fresh BBOA-1 (Fig. S8a) and stronger association with nitrate-related chemistry and  
545 organonitrates (Kaltsonoudis et al., 2025). The time-series variation of BBOA-1 shows greater daily variability than  
546 that of BBOA-2, which has much less (Fig. 6(i)). These variations depict the aged nature of BBOA-2. CWT analysis  
547 indicates that both BBOA-1 and BBOA-2 at Site 1 are influenced by regional biomass burning across the IGP,  
548 particularly post-monsoon agricultural burning in Punjab and Haryana (Fig. S9.(i)) (Lalchandani et al., 2022), with  
549 additional contributions from local open burning and lemongrass filtration activity.

550 At Site 2, a traffic-influenced urban location, BBOA-1 again represents fresh biomass-burning OA with spectral  
551 features similar to Site 1 but a slightly lower O: C ( $\approx 0.42$ ), indicating even less oxidation; it contributes a larger  
552 fraction ( $\approx 20.1\%$  of OA) (Fig. 7(ii)), and its morning peak (07:00–08:00) reflects proximity to fresh emissions from  
553 waste burning, residential cooking, and open biomass burning near the roadway (Fig. S3.g). Its correlation with nitrate  
554 remains weak ( $r \approx 0.26$ ), reinforcing its characterization as a primarily primary, non-nitrate-coupled biomass-burning  
555 factor (Fig. S8b). BBOA-2 at Site 2 is the aged counterpart, with higher  $m/z$  43 and 44 and low PAH signals, an O: C

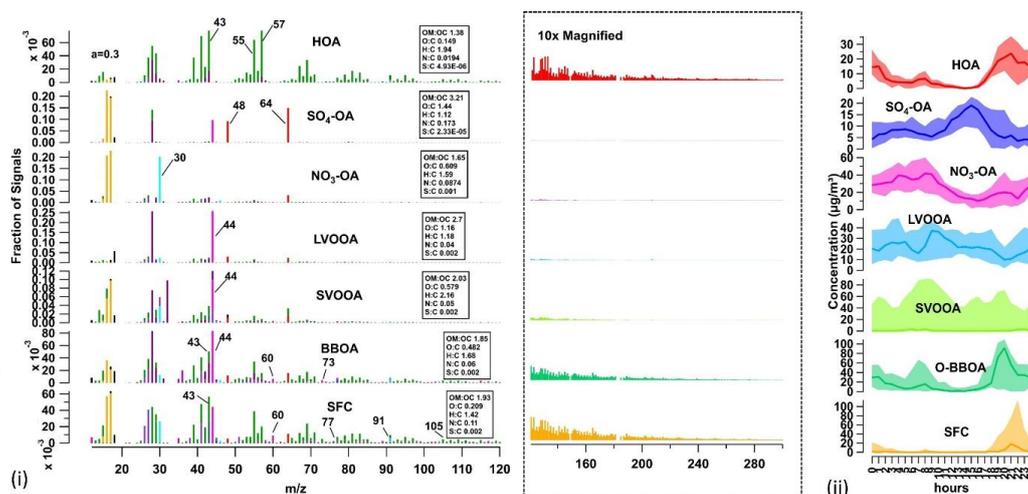


556 of  $\approx 0.46$ , and a more minor OA contribution ( $\approx 7.7\%$ ) than BBOA-1; it peaks about 1–2 hours after BBOA-1, consistent  
557 with rapid oxidation, and shows a stronger correlation with nitrate ( $r \approx 0.41$ ), pointing to enhanced connections with  
558 nitrate and organonitrate chemistry (Zheng et al., 2020). In contrast to Site 1, BBOA-1 shows more variability with  
559 concentration at Site 2 (Fig. 7(i)). CWT results for BBOA-1 at Site 2 show an elongated plume along the IGP with a  
560 hotspot near the receptor (Fig. S9(ii)), indicating a combination of regional biomass-burning transport and strong  
561 local sources.

562 At Site 3, a highly polluted industrial environment, biomass-burning OA is dominated by an oxidized BBOA factor  
563 (O-BBOA). O-BBOA shares biomass-burning tracers such as  $m/z$  60 and 73 but has very low high- $m/z$  ( $>120$ ) PAH  
564 intensities, indicating substantially aged biomass-burning aerosol (Fig. 5i). Its O:C ratio ( $\approx 0.48$ ) is comparable to  
565 BBOA-1 at Site 1 but lower than values reported at other CIGP Sites, such as Kanpur and Lucknow (Lakra et al.,  
566 2024), and it contributes about 20% of OA (Fig. 8(ii)). O-BBOA shows a low correlation with nitrate ( $r \approx 0.03$ ) and  
567 only a weak association with the biomass-burning marker  $m/z$  60 ( $r \approx 0.24$ ), both consistent with a more aged  
568 biomass-burning plume whose variability is decoupled from contemporaneous nitrate formation. Diurnally, O-BBOA  
569 exhibits relatively high morning concentrations, exceeding those at the other two Sites (Fig. 5ii), and a very strong  
570 nighttime enhancement in winter, with nighttime levels about three times those during the day (Fig. S3h). This  
571 behavior likely reflects  $\text{NO}_3$  radical-driven oxidation of fresh biomass burning emissions, combined with a shallow,  
572 stable nocturnal boundary layer that traps combustion products and high relative humidity ( $\sim 71\%$ ) that promotes the  
573 partitioning of semi-volatile biomass burning products and aqueous/heterogeneous oxidation (Lhotka et al., 2025).  
574 The strong correlations of O-BBOA with CO,  $\text{NO}_x$ ,  $\text{SO}_2$ , and BC ( $r > 0.6$ ) further indicate its close linkage to  
575 combustion sources under humid, stagnant conditions (Awasthi et al., 2024). The time-series variation of the O-  
576 BBOA shows greater variation between 8-17 October 2023; thereafter, we observed lesser variation approaching the  
577 extreme winter conditions (Fig. 8(i)).

578 The CWT results indicate a broad source region extending from north-central to eastern IGP, with a dominant air mass  
579 influence from Punjab and Haryana, likely linked to post-monsoon agricultural stubble burning in these areas for Site  
580 1 (Fig. S9(i)). The map for Site 2 shows the potential source regions that influence the BBOA-1 factor (Fig. S9(ii)).  
581 The elongated plume from northwest to southeast, with a hotspot near the receptor, indicates that BBOA-1 is mainly  
582 associated with regional biomass burning upwind of the IGP, with moderate–strong contributions reaching the Site  
583 2. CWT analysis of the O-BBOA-dominated factor at Site 3 reveals a compact, intense hotspot over northern India  
584 (Fig. S9. (iii)), highlighting a strong local and near-regional biomass-burning influence around the receptor, consistent  
585 with elevated nighttime O-BBOA and nearby residential and solid-fuel burning.

586 Across the three Sites, BBOA-1 captures fresh biomass-burning emissions, BBOA-2 represents more aged  
587 biomass-burning OA with stronger nitrate coupling at Sites 1–2, and O-BBOA at Site 3 characterizes aged, highly  
588 polluted biomass-burning aerosol that accumulates under humid, stagnant winter conditions. Site-wise differences in  
589 O:C ratios, contributions, correlations with  $\text{NO}_3^-$  and tracers, and diurnal cycles together indicate a transition from  
590 regionally influenced biomass burning at the background Site, through locally amplified emissions at the Site 3, to  
591 strongly aged and trapped biomass-burning aerosol at the industrial, highly polluted Site.



592

593 Figure 5. i) Mass spectral profiles of sources/factors obtained from the combined Organics and Inorganics sources  
 594 apportionment for m/z 12 to m/z 115, along with UMR spectra highlighted inside a grey border for m/z 120 to m/z 300.  
 595 Elemental Ratio(O/C, OM/OC, H/C, and N/C) has been calculated using only the organic signals, and ii) Diurnal variation  
 596 of obtained sources during the measurement period over Site3.

597 **3.2.2.5 NO<sub>3</sub>-OA and SO<sub>4</sub>-OA:** The NO<sub>3</sub>-OA and SO<sub>4</sub>-OA factors together represent the secondary inorganic aerosol  
 598 (SIA) component. The SO<sub>4</sub>-OA, a major inorganic-rich factor, exhibited higher intensity of sulfate ions at m/z 64  
 599 (SO<sub>2</sub>), m/z 48 (SO), and ammonium ions at m/z (NH<sub>3</sub>) and m/z 16 (NH<sub>2</sub>) in its mass spectra (Fig. 3-5) at all Sites. It  
 600 primarily consisted of ammonium sulfate, with ~68%, 59%, and 68% of the total mass of this factor. Followed by the  
 601 organics contributed, 29%, 39%, and 32%, with O/C ratios among all factors (1.22, 1.07, 1.07, and 1.44) in Site 1,  
 602 Site 2, and Site 3, respectively (Fig. 6-8(i)). The highest O/C ratio indicates a highly oxidized nature of the organics  
 603 within the factor. The SO<sub>4</sub>-OA shows correlations ( $r = 0.98, 0.68, \text{ and } 0.61$ ) with sulfate from AMS and sulfur from  
 604 the metal monitor ( $r = 0.72$ ) across three Sites (Fig. S9 and S2). The very high correlations of SO<sub>4</sub>-OA with AMS  
 605 sulfate at all three Sites ( $r = 0.98, 0.68, 0.61$ ) and with sulfur from the metal monitor ( $r = 0.72$ ) indicate that this factor  
 606 is tightly linked to sulfate-rich aerosol and sulfur-containing emissions (Saarikoski et al., 2019). This implies that  
 607 SO<sub>4</sub>-OA predominantly represents secondary sulfate-associated organic aerosol formed in or on sulfate particles (e.g.,  
 608 via multiphase oxidation of SO<sub>2</sub> and organics), and that the same sulfur sources and formation pathways  
 609 (industrial/coal SO<sub>2</sub>, regional sulfate production) control both the inorganic sulfate and the associated SO<sub>4</sub>-OA factor  
 610 (Xu et al., 2016). The time-series variation of SO<sub>4</sub>-OA shows very low variability (~4-5 times less) at site 1 compared  
 611 to sites 2 and 3 (Fig. 6-8(i)).

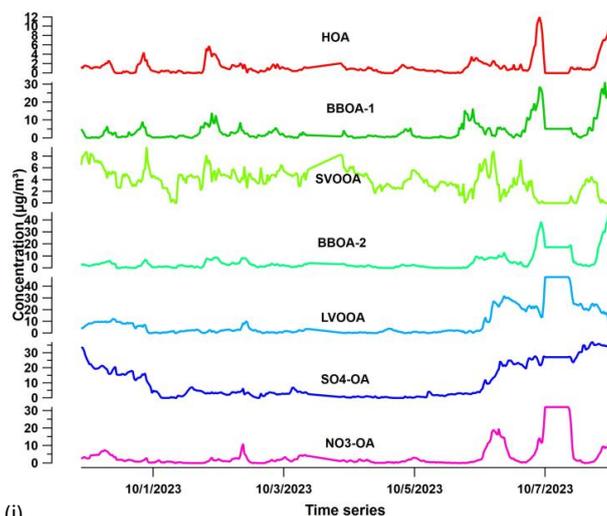
612 All the secondary inorganic components show a prominent peak at ~9-10 am, coinciding with NO<sub>x</sub> ( $r=0.85$ ) at  
 613 Site1(Fig. S4a and S2). NO<sub>x</sub> peaks align with NO<sub>3</sub>-OA and SO<sub>4</sub>-OA because NO<sub>x</sub> is a key precursor for both nitrate  
 614 and sulfate formation. Elevated NO<sub>x</sub> promotes production of nitric acid (driving particulate nitrate) and enhances  
 615 multiphase oxidation of SO<sub>2</sub> to sulfate, so secondary inorganic aerosol and NO<sub>x</sub> rise together in the late morning  
 616 (Squizzato et al., 2013). The NO<sub>x</sub> morning peak during the traffic rush hours and onset of mixing (PBLH) increases



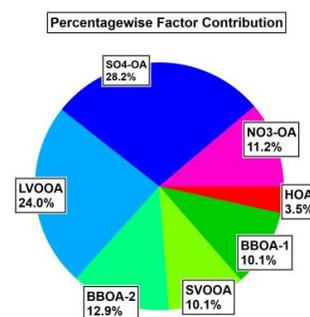
617 from ~200m at 9:00 to 11:00 over 1 km (Fig. S4a and g). The SO<sub>4</sub>-OA shows a strong correlation with ozone (O<sub>3</sub>) at  
 618 Site 1, with  $r = 0.62$  (Fig. S9 and S4d). The strong correlation between SO<sub>4</sub>-OA and O<sub>3</sub> suggests that sulfate-related  
 619 OA forms under photochemically active, oxidizing conditions where ozone is abundant. Elevated O<sub>3</sub> reflects intense  
 620 daytime photochemistry and high oxidant levels, which enhance multiphase oxidation of SO<sub>2</sub> to sulfate (one of the  
 621 main pathways involves O<sub>3</sub> as an aqueous oxidant), so periods with more O<sub>3</sub> naturally coincide with greater sulfate  
 622 and associated SO<sub>4</sub>-OA production. The SO<sub>4</sub>-OA formation insights are further obtained by estimating the sulfur  
 623 oxidation ratio (SOR) for all three Sites (Fig. S5) (Lakra et al., 2024). SOR was estimated to be higher at the highly  
 624 polluted Site 3 than at the moderately polluted Site 2 and the less polluted Site 1 (Fig. S5). The range of SOR values  
 625 at Sites 1 and 2 was approximately half that observed at the highly polluted Site 3 (Fig. S5). Both SOR and SO<sub>4</sub>-OA  
 626 (see Fig.S5 and 6d) increase between 9:00 and 19:00 hours, indicating photochemical production of SO<sub>4</sub>-OA under  
 627 conditions of high temperature and low relative humidity. Additionally, the SOR values at Sites 3 and 2 showed  
 628 minimal variation, suggesting that both aqueous-phase and photochemical processes contribute significantly to  
 629 sulphate formation at Site 3 (Lalchandani et al., 2022; Sun et al., 2012).

630 The CWT analysis for site 1 indicates that the SO<sub>4</sub>-OA factor predominantly contributes at low concentrations (2–3  
 631  $\mu\text{g m}^{-3}$ ), with primary transport from the northwest region of the Uttar Pradesh district (Fig. S9i-c). At sites 2 and 3,  
 632 regional advection from Punjab and Haryana governs contributions in the 10–15  $\mu\text{g m}^{-3}$  range (Fig. S9ii-c) and 15–25  
 633  $\mu\text{g m}^{-3}$  range (Fig. S9iii-b), respectively. Regional transport concentrations exhibit a marked seasonal escalation from  
 634 post-monsoon to winter, consistent with enhanced secondary sulfate formation under stagnant meteorological  
 635 conditions.

636 At all three Sites, the NO<sub>3</sub>-OA factor was predominantly ammonium nitrate, accounting for approximately 52%, 62%,  
 637 and 66% at each Site, respectively (Fig. 3–5). The mass spectra for this factor, displayed in Figures 3–5, show high-  
 638 intensity peaks at  $m/z$  46 (NO<sub>2</sub>) and  $m/z$  30 (NO).



639 (i)



(ii)



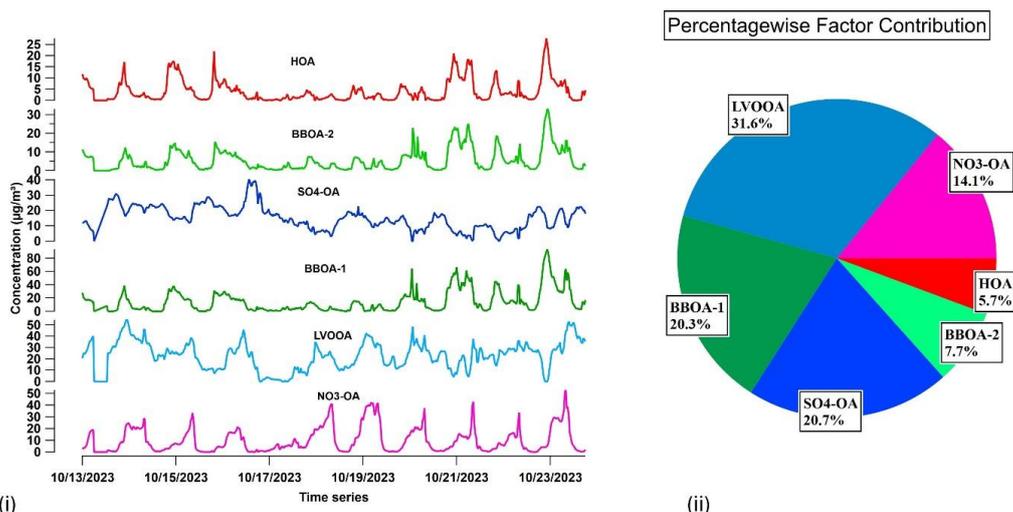
640 **Figure 6. i) Time series variation of the NR-PM<sub>2.5</sub> sources obtained from combined OA and Inorganics sources**  
641 **apportionment ii) Average chemical composition of NR-PM<sub>2.5</sub> composition during the measurement period over Site 1.**

642 Furthermore, the Nitrogen Oxidation Ratio (NOR), an indicator of secondary NO<sub>3</sub>-OA formation, was high during the  
643 nighttime and early morning, peaking around 05:00. NOR values were consistently higher at the more polluted sites  
644 (Fig. S5). In addition, both SOR and NOR in this study were substantially higher than those reported in previous  
645 winter campaigns (2021–2023) at CIGP Sites, such as Lucknow and Kanpur, indicating a higher degree of SOA  
646 formation under more polluted conditions (Lakra et al., 2024). During the post-monsoon to winter period in Lucknow  
647 and Kanpur, levels are higher than those reported in previous winter campaigns, likely due to enhanced atmospheric  
648 oxidation capacity and changing emission or meteorological conditions (Chang et al., 2022; Wang et al., 2024).

649 The time-series variation of NO<sub>3</sub>-OA is too smooth, except at the last observation period on the polluted event day,  
650 compared with Sites 2 and 3 (Fig. 6-8(i)). This might be due to the regional build-up contribution only at Site 1.

651 The diurnal profile reveals that the peak of NO<sub>3</sub>-OA occurred approximately two hours after the morning peak of  
652 HOA at Site 1, and about one hour later at Site 3 (Fig. 6c and h). Morning rush-hour traffic emits primary HOA and  
653 NO<sub>x</sub> first, peaking under a shallow boundary layer. The rising sun then drives NO<sub>x</sub> oxidation to particulate nitrate (1–  
654 2 h timescale) and its mixing into NO<sub>3</sub>-OA, causing the delayed peak (Chen et al., 2021; Poulain et al., 2011). In  
655 contrast, at Site 2, which is characterized by both traffic and residential influences, HOA and NO<sub>3</sub>-OA peaked  
656 simultaneously during the morning hours (Fig. S3e and c). This pattern suggests that ammonium nitrate formation  
657 results from the reaction between NO<sub>x</sub> (emitted from traffic) and ammonia, facilitated by the gas-phase oxidation of  
658 NO<sub>x</sub> by OH radicals under conditions of low ambient temperature (~20.5°C) and high RH (90%) (see Fig. S4e and  
659 S4f). Following these peaks, concentrations decrease rapidly throughout the day as rising temperatures promote the  
660 conversion of nitrate particles to nitric acid and ammonia (NH<sub>4</sub>NO<sub>3</sub> → HNO<sub>3</sub> + NH<sub>3</sub>), consistent with findings from  
661 previous studies (Faisal et al., 2022; Lakra et al., 2024; Sun et al., 2012). The concentration further increases at night,  
662 primarily driven by nighttime chemistry in which NO<sub>x</sub> is oxidized by ozone at higher altitudes through heterogeneous  
663 reactions (Wang et al., 2018; Yun et al., 2018). This process occurs under conditions of high relative humidity (RH >  
664 92.16% at Site 1, RH > 86.23% at Site 2, and RH > 82.66% at Site 3) and low temperatures (AT < 25.8°C at Site 1, <  
665 21.3°C at Site 2, and < 19.0°C at Site 3). Such conditions facilitate the formation of these compounds in the upper  
666 tropospheric layers across all three Sites (Chang et al., 2011). This process was further validated by strong nighttime  
667 correlations between NOR and NO<sub>3</sub>-OA, with coefficients of 0.92, 0.96, and 0.97 at the three Sites (Fig. S3.6c and  
668 S5b). These high correlation values provide additional evidence for the concentration increase at both Sites, supporting  
669 the role of heterogeneous reactions. In contrast, rising ambient temperature (AT) during the afternoon is associated  
670 with a decrease in concentration.

671 The CWT analysis shows that NO<sub>3</sub>-OA mainly contributes at 2–3.5 μgm<sup>-3</sup> at site 1, located northwest of Site 1 (Fig.  
672 S9i-c). Sites 2 and 3 show major local contribution from northwesterly wind transport of 2–8 μgm<sup>-3</sup> (Fig. S9ii-d) and  
673 15–30 μgm<sup>-3</sup> (Fig. S9iii-f), respectively. The local transport concentrations increase progressively from post-monsoon  
674 to winter, reflecting enhanced secondary nitrate formation under stagnant conditions.

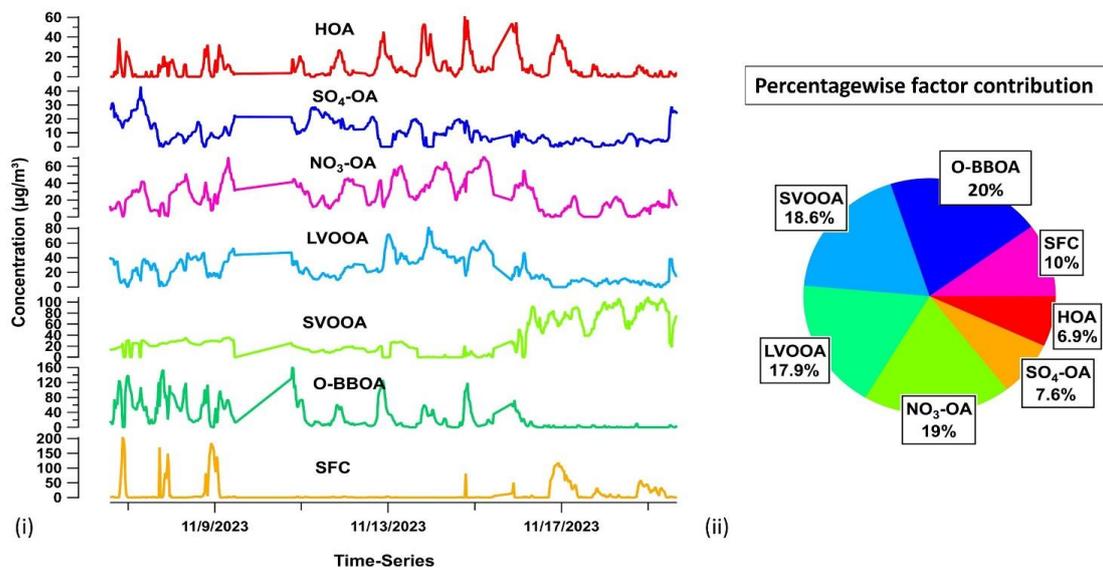


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Figure 7. i) Time series variation of the NR-PM<sub>2.5</sub> sources obtained from the combined OA and IOA sources apportionment. ii) Average chemical composition of NR-PM<sub>2.5</sub> during the measurement period over Site 2.



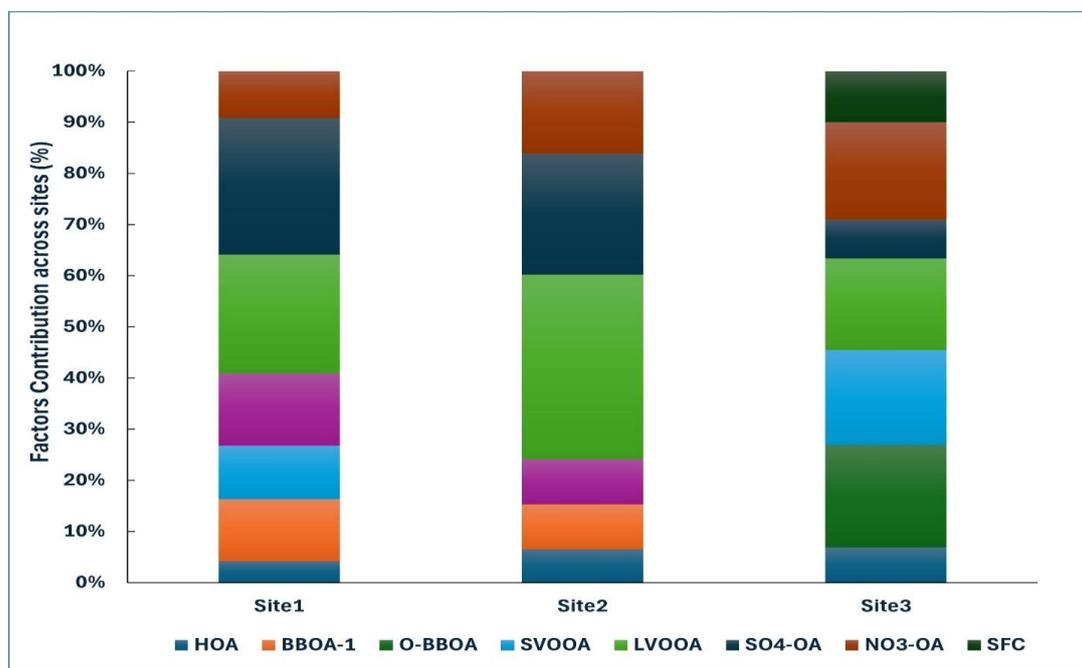
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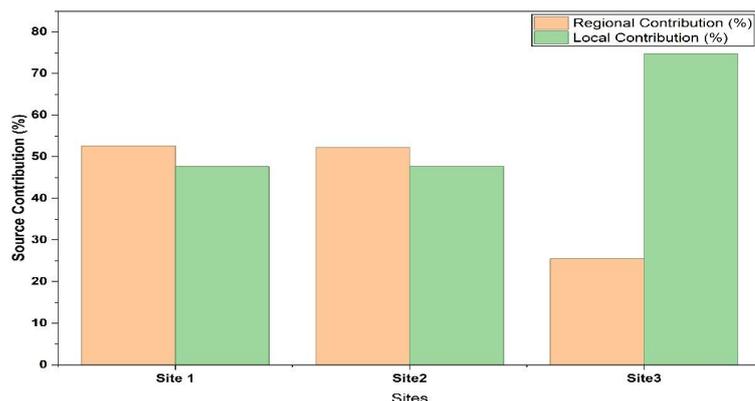
Figure 8. i) Time series variation of the NR-PM<sub>2.5</sub> sources obtained from combined OA and IOA sources apportionment ii) Average chemical composition of NR-PM<sub>2.5</sub> during the measurement period over Site 3.



682  
683 **Figure 9. Factor abundant contribution across all three measurement Sites.**

### 684 **3.3 Local and Regional contribution in three Sites**

685 Previous research has demonstrated that source apportionment factors are valuable for distinguishing and quantifying  
686 the contributions of local and regional aerosol transport within a given area (Jimenez et al., 2009; Sun et al., 2011;  
687 Zhang et al., 2007). Specifically, the combined presence of LVOOA and SO<sub>4</sub>-OA typically indicates regional  
688 transport. In contrast, SVOOA and NO<sub>3</sub>-OA are primarily linked to the oxidation of local emissions at a Site. Figures  
689 9 and 10 show the factors contributing to each Site, as well as long-range and local transport at all three Sites during  
690 the measurement period. Regional transport dominated at Sites 1 and 2, contributing 52.6% and 52.3%, respectively,  
691 but only 25.3% at Site 3 (Fig.10). Conversely, local sources accounted for 74.7% at Site 3, compared to 47.4% at Site  
692 1 and 47.7% at Site 2 (Fig.10). The reason for this contribution might be that the transboundary emissions from  
693 neighboring states account for 70-80% of PM<sub>2.5</sub> in Lucknow, carried by prevailing winds, stubble burning, industrial  
694 sources, and vehicular traffic, which overwhelms local contributions even at Sites 1 and 2, respectively(The Energy  
695 and Resources Institute, 2022).



696

697 **Figure 10. Source contribution across different Sites during the measurement period.**

698 This regional influence is clearly reflected in the concentration-weighted trajectory plots, which show that  
699 transboundary emissions transported by prevailing winds from neighboring states constitute a major share of PM<sub>2.5</sub>  
700 sources in Lucknow (Fig. S9i–iii). Notably, there was a significant contribution from the northwestern direction,  
701 particularly from the northern IGP regions of Punjab and Haryana. Figures S9(i)e and S9(i)g clearly demonstrate the  
702 long-range transport of aged LVOOA and NO<sub>3</sub>-OA particles from neighboring states and countries over Site 1.  
703 Similarly, at Site 2, we observed a notable influence of regional transport, likely due to comparable meteorological  
704 conditions and prevailing wind patterns, as all measurements were conducted during the same season. Additionally,  
705 concentrations of LVOOA and SO<sub>4</sub>-OA were considerably higher at Site 2 (Fig. S9(ii)). Given Site 2's proximity to a  
706 highway or high-traffic area, Figure S9(ii, b) shows a notable contribution of HOA from nearby sources. In contrast,  
707 at the highly polluted Site 3, we detected a significant local contribution, with elevated levels of BBOA, HOA, SFC,  
708 NO<sub>3</sub>-OA, and SVOOA observed in close proximity to the Site (Fig. S9(iii)).

### 709 **3.4 Case study of the PGE**

710 PGEs are defined as periods during which particle number concentration increases, accompanied by a shift in the  
711 mode toward larger geometric diameters, typically following new particle formation (Zimmerman et al., 2020). This  
712 growth is driven by condensation of low-volatility vapors (such as sulfuric acid and organic compounds) onto freshly  
713 nucleated clusters, leading to a relative decrease in the organic mass fraction compared to inorganic components in  
714 the growing mode (Tröstl et al., 2016). In the present study, PGEs are selected under conditions where the CS exceeds  
715 0.1 s<sup>-1</sup>, indicating a relatively high background aerosol loading that efficiently scavenges condensable vapors. Here,  
716 we will discuss the PGEs that we observed at Site 1 during the campaign (Fig. S7(i)). We observed the two particle  
717 growth events, PGE 1 and 2, respectively. The estimated particle growth rates are 2.68nm/hr. and 4.82nm/hr.,  
718 respectively. During the measurement period at Site 2, five PGE events were recorded (Fig. S7 (ii) and Table 1). Four  
719 of these events exhibited growth rates between 1.12 and 1.59 nm/hr, except for October 20, 2023, when an elevated  
720 growth rate of approximately 6.15 nm/hr was observed. The van, stationed within the university campus area



721 surrounded by vegetation and shrubs, showed comparatively lower growth rates. At Site 3, we detected 10 such events,  
722 with growth rates ranging from 4.97 to 16.51 nm/hr (Fig. S7(iii) and Table 1). Despite this high sink, sustained growth  
723 is observed, suggesting that local vapor sources (e.g., from traffic, cooking, or secondary formation) are sufficient to  
724 support continued particle growth even in a polluted urban environment. We observed higher particle growth rates in  
725 polluted environments and lower growth rates in cleaner environments, consistent with previously reported data for  
726 similar conditions over the northern IGP (Mishra et al., 2023; Rathore et al., 2025). The observed PGEs with elevated  
727 condensation sinks ( $>0.1 \text{ s}^{-1}$ ) in Lucknow's highly polluted environment align with findings from Chinese megacities,  
728 where high CS values signify increased scavenging of gaseous precursors, such as sulfuric acid and organics, by  
729 preexisting aerosols, yet still permit growth dynamics (Du et al., 2022; Tuovinen et al., 2020).

730 BBOA-1 (fresh BB, high m/z 60) correlates strongly with nitrate ( $r=0.66$ ) at Site 1 via regional stagnant chemistry,  
731 but weakly ( $r=0.26$ ) at Site 2 due to local NO<sub>x</sub> overriding BB signals—directly tying secondary nitrate formation to  
732 apportioned BBOA transport.

733 O-BBOA (high f<sub>44</sub>,  $r=0.24$  with m/z 60) at Site3 reflects processed BB organics growing via industrial vapors,  
734 explaining winter nighttime PGE maxima (5–15 nm/h) present at industrial Sites(Sarangi et al., 2018).

735 PMF source apportionment reveals distinct organic aerosol compositions across Lucknow Sites during the highly  
736 polluted post-monsoon period, directly linking to Site-specific dynamics observed in this study. At the Site1 (CSIR-  
737 CIMAP), SO<sub>4</sub>-OA (28%), LV-OOA (24%), and BBOA-2 (13%) dominate, driven by regional nitrate-BBOA co-  
738 variation ( $r=0.66$ ) that fosters secondary OA buildup under stagnant conditions. The Site 2 (BBAU) features LV-OOA  
739 (32%) and BBOA-1 (20%), with weak nitrate decoupling ( $r=0.26$ ) from local NO<sub>x</sub> emissions overriding BB signals  
740 amid traffic's 50–60% contribution to ultrafine particle number concentration (PNC). At the Site 3 (Talkatora), O-  
741 BBOA (20%) and NO<sub>3</sub>-OA (19%) prevail, enhanced by nocturnal particle growth events (5–15 nm/h) where stack  
742 vapors process BB organics (f<sub>60</sub>  $r=0.24$ ) under winter inversions. These hyperlocal patterns explain the dominance of  
743 organics (64–70% of NR-PM<sub>2.5</sub>), underscoring the value of PMF for targeted controls on traffic PNC, regional  
744 precursors, and industrial emissions in the Indo-Gangetic Plain.

745 Traffic drives daytime particle number concentration via direct ultrafine emissions, but experience higher dispersion  
746 and scavenging at night, thereby suppressing PGEs during the post-monsoon period. Industrial Sites show less dilution  
747 and more persistent chemistry.  
748 In the Indo-Gangetic Plain, during winters (e.g., Lucknow), nighttime PGEs are linked to industrial plumes under  
749 inversion layers, with growth rates enhanced by SO<sub>2</sub>/NO<sub>x</sub> emissions from stacks (Sarangi et al., 2018). These studies  
750 emphasize that polluted urban Sites with substantial aerosol loading exhibit enhanced vapor removal, which influences  
751 nucleation and growth processes, consistent with the current results.

752

753



754 **4. Conclusion**

755 Overall, this study presents around 31 days of hyperlocal sampling across the CIGP city during the most polluted  
756 period (post-monsoon to winter) at three Sites representing distinct environmental conditions. For the first time in an  
757 Indian urban setting, a VSAAQMP platform was deployed to obtain high-resolution measurements of PM<sub>2.5</sub>  
758 composition and to perform source apportionment using a robust PMF approach coupled with the ME-2 engine,  
759 supported by collocated reference-grade instruments that strengthened the reliability of the observations. Using these  
760 near-source hyperlocal measurements collected in the post-monsoon to winter season of 2023 at multiple intra-city  
761 locations, the work investigates C-PM<sub>2.5</sub> levels in detail and yields the following key findings:

762 a) We found that during the highly polluted post-monsoon period, organics accounted for the largest contribution at  
763 all Sites, ranging from 64% to 70%. This was followed by nitrate (4–9%), sulfate (8–10%), ammonium (4–6%), and  
764 chloride (1–2%), while the refractory component, black carbon (BC), ranged from 4% to 17%. BC and sulfate (SO<sub>4</sub>)  
765 dominance at the clean/urban background site (Site 1) suggests regional/long-range transport and minimal local  
766 influence, whereas chloride (Chl), nitrate, and ammonia at the highly polluted Site 3 indicate intense local primary  
767 emissions and secondary aerosol formation under high-pollution conditions.

768 b) At Site 1, SO<sub>4</sub>-OA (28.2 %) and LV-OOA (24.0 %) were the largest contributors, with additional contributions from  
769 BBOA-2 (12.9 %), SV-OOA (10.1 %), BBOA-1 (10.1 %), NO<sub>3</sub>-OA (11.2 %), and a smaller share from HOA (3.5 %).  
770 At Site 2, LV-OOA (31.6 %) and SO<sub>4</sub>-OA (20.7 %) again dominated, followed by BBOA-1 (20.3 %), NO<sub>3</sub>-OA  
771 (14.1 %), BBOA-2 (7.7 %), and HOA (5.7 %). At Site 3, O-BBOA (20 %), SV-OOA (18.6 %), NO<sub>3</sub>-OA (19 %), and  
772 LV-OOA (17.9 %) accounted for most of the organic mass, with additional contributions from solid fuel combustion  
773 (SFC) (10 %), SO<sub>4</sub>-OA (7.6 %), and HOA (6.9 %).

774 c) The strong correlation observed between SO<sub>4</sub>-OA and RH indicates that aqueous-phase processing significantly  
775 contributes to SO<sub>4</sub>-OA formation during nighttime. This process was most pronounced at the highly polluted Site 3,  
776 followed by moderate activity at Site 2, and was least evident at the cleaner Site 1. Additionally, photochemical  
777 production was more prominent during the daytime at Site 3 than at Sites 1 and 2. In summary, at the highly polluted  
778 Site, diurnal variation in SO<sub>4</sub>-OA indicates that photochemical processes dominate during the day, while aqueous-  
779 phase chemistry is more influential at night. In contrast, at less- and moderately polluted Sites, both mechanisms  
780 contribute to SO<sub>4</sub>-OA formation throughout the day and night.

781 d) At Site 3, the CS varies from 0.1 to 0.7 s<sup>-1</sup>, indicating a strong scavenging rate for gaseous precursors such as sulfuric  
782 acid and organic vapors—conditions typical of Lucknow's highly polluted urban environment. Hyperlocal PMF source  
783 apportionment across Lucknow's background (Site1), traffic (Site2), and industrial (Site3) Sites during the post-  
784 monsoon period elucidates organics' dominance (64–70% NR-PM<sub>1</sub>) through distinct mechanisms: regional secondary  
785 buildup of SO<sub>4</sub>-OA (28%) and LV-OOA (24%) via nitrate-BBOA co-variation (r=0.66) at Site 1; traffic-fueled  
786 nucleation into LV-OOA (32%) and BBOA-1 (20%) with NO<sub>x</sub> decoupling (r=0.26) at Site 2; and industrial vapor-  
787 driven nocturnal PGEs (5–15 nm/h) processing O-BBOA (20%, f60 r=0.24) at Site 3.



788 e)Our analysis indicates that local and regional sources contributed 43.4% and 56.6%, respectively, across three  
789 measurement Sites, showing the strong background/transboundary influence.

790 f)The results indicate that particle growth events in the study domain are strongly controlled by pollution level and  
791 nocturnal secondary aerosol formation. A majority of PGEs occur at the most polluted Site, and about 53% take place  
792 at night, coinciding with high contributions from biomass-burning OA (BBOA), low-volatility oxygenated OA  
793 (LV-OOA), and nitrate-associated OA (NO<sub>3</sub>-OA) at Sites 2 and 3. This implies that abundant organic precursors from  
794 combustion sources, together with nocturnal oxidants and shallow boundary layers(~250m-700m), efficiently drive  
795 nighttime SOA formation(as O: C ratio was high ) and condensation-driven particle growth( as CS was higher),  
796 whereas the cleaner background Site, with weaker BBOA/LV-OOA influence, exhibits far fewer growth events. Thus,  
797 polluted C-IGP environments strongly enhance secondary aerosol formation and particle growth, whereas the cleaner  
798 background Site lacks sufficient precursors to sustain frequent growth events. These findings align with observations  
799 from the northern IGP studies.

800 These findings underscore the value of advanced PMF in resolving urban OA heterogeneity beyond bulk  
801 compositions, highlighting the need for traffic PNC controls, industrial vapor mitigation, and sustained high-resolution  
802 AMS/SMPS networks to address the impacts on air quality/climate in the Indo-Gangetic Plain. These regional  
803 influences in the background Site highlight the need for an integrated approach that combines regional airshed  
804 management with targeted hyperlocal interventions. Temporal analysis of PM<sub>2.5</sub> composition and sources, in  
805 conjunction with CWT analysis and meteorological conditions, suggests that transboundary emissions carried by  
806 prevailing winds from neighboring states significantly contribute to PM<sub>2.5</sub> in Lucknow. In contrast, hyperlocal  
807 monitoring at the most polluted Sites reveals substantial local contributions. Together, these findings demonstrate that  
808 effective air quality management must prioritize both coordinated regional controls and Site-specific, neighborhood-  
809 level measures, using hyperlocal data to design strategic, source-focused actions that protect human health and the  
810 environment. The novelty of the present work lies in its exploration of air quality at the hyperlocal level within the  
811 city scale. This type of hyperlocal measurement helps strategically target air pollution sources rather than  
812 implementing bulk measures. Additionally, more coordinated and systematic actions are essential to address air  
813 pollution in highly impacted areas.

#### 814 **Data availability**

815 Data will be made available on request to the corresponding author.

816

#### 817 **Author contributions**

818 DS designed the study, performed data analysis and conducted field investigations, developed methodology and  
819 software, conducted validation and formal analysis, and wrote the original draft with review and editing. AL performed  
820 data analysis and software, and conducted field investigations, with review and editing. AK, with review and editing,



821 SC, HB, VJ, and SNT commented on and discussed the manuscript drafts. SNT provided supervision, resources,  
822 project administration, and funding acquisition. All authors reviewed and approved the final manuscript.

### 823 **Competing interest**

824 The contact author has declared that none of the authors has any competing interests.

### 825 **Disclaimer**

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