Modelling PM2.5 during severe atmospheric pollution episode in Lagos, Nigeria: Spatiotemporal variations, source apportionment, and meteorological influences

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Abstract

In 2021, the World Health Organization (WHO) ranked Nigeria among the most polluted nations in the world, an indication of a deteriorating air quality, especially in the major urban areas of the country, which might pose adverse human health impacts. In this study, the Integrated Source Apportionment Method (ISAM) tool in the Community Multiscale Air Quality (CMAQ) model (CMAQ-ISAM) was employed to quantify the contributions of eight emissions sectors to fine particulate matter (PM2.5) and its major components in Lagos during a prolonged severe atmospheric pollution episode (APE) in January 2021. The influence of meteorological conditions on the formation and dispersion of PM2.5 during the APE was also elucidated. Spatially, elevated PM2.5 concentrations were found in the northwestern region of Lagos, an urban area with larger anthropogenic emissions. Residential and industry were the two major sources of PM2.5. Residential contributed the most to total PM2.5 (~40 μ g/m3), followed by industry (~20 μ g/m3). High concentrations of secondary inorganic aerosols (SIA) at the northwest and upper northern areas of Lagos were majorly attributed to residential and industry sectors. In addition, sulfate accounted for the largest fraction of PM2.5, with residential, industry, and energy being its major sources. Residential, industry, and on-road sectors dominated the contributions to nitrate, while residential and industry were the major contributors to ammonium. Furthermore, the elevated PM2.5 concentrations during the APE were greatly enhanced by unfavorable meteorological conditions. This study provides insights for designing effective emissions control strategies to mitigate future severe PM2.5 pollution episode in Lagos.

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- 2 Spatiotemporal variations, source apportionment, and meteorological influences
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22	Key Points:
23	• Lagos experienced a prolonged severe PM _{2.5} pollution episode in January 2021.
24	• PM _{2.5} pollution was enhanced by unfavorable meteorological conditions.
25 26 27 28 29 30 31 32 33 34 35	• Effective emissions control strategies to reduce PM _{2.5} concentrations are urgently required in Lagos.

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Abstract

In 2021, the World Health Organization (WHO) ranked Nigeria among the most 38 polluted nations in the world, an indication of a deteriorating air quality, especially in the 39 major urban areas of the country, which might pose adverse human health impacts. In this 40 study, the Integrated Source Apportionment Method (ISAM) tool in the Community 41 42 Multiscale Air Quality (CMAQ) model (CMAQ-ISAM) was employed to quantify the contributions of eight emissions sectors to fine particulate matter (PM_{2.5}) and its major 43 components in Lagos during a prolonged severe atmospheric pollution episode (APE) in 44 45 January 2021. The influence of meteorological conditions on the formation and dispersion of PM_{2.5} during the APE was also elucidated. Spatially, elevated PM_{2.5} 46 47 concentrations were found in the northwestern region of Lagos, an urban area with larger anthropogenic emissions. Residential and industry were the two major sources of PM_{2.5}. 48 Residential contributed the most to total PM_{2.5} (~40 µg/m³), followed by industry (~20 49 $\mu g/m^3$). High concentrations of secondary inorganic aerosols (SIA) at the northwest and 50 upper northern areas of Lagos were majorly attributed to residential and industry sectors. 51 In addition, sulfate accounted for the largest fraction of PM_{2.5}, with residential, industry, 52 53 and energy being its major sources. Residential, industry, and on-road sectors dominated the contributions to nitrate, while residential and industry were the major contributors to 54 ammonium. Furthermore, the elevated PM_{2.5} concentrations during the APE were greatly 55 56 enhanced by unfavorable meteorological conditions. This study provides insights for designing effective emissions control strategies to mitigate future severe PM_{2.5} pollution 57 58 episode in Lagos.



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1. Introduction

Due to rapid population growth, accelerated urbanization, economic advancement, 62 and fast industrialization, Nigeria has been suffering from severe air pollution for more 63 than two decades (Abiye et al., 2013, 2014; Owoade et al., 2013; Sulaymon et al., 2020). 64 In 2021, the WHO ranked Nigeria among the most polluted nations in the world (WHO, 65 66 2021), making it a great concern for the Nigerian air quality experts. The anthropogenic and natural emissions are the major sources of particulate matter and gaseous primary air 67 pollutants (Owoade et al., 2013). Air pollution is largely linked to emissions from 68 69 anthropogenic activities (either local or regional) (Sulaymon et al., 2020, 2021a) and aggravated by unfavorable meteorological factors (Hua et al., 2021; Hu et al., 2016; 70 71 Sulaymon et al., 2021a, 2021b). Severe air pollution is strongly correlated with visibility reductions (Jiang et al., 2021; Li et al., 2019; Wang et al., 2018), changes in ecosystem 72 services, effects on climate (Jiang et al., 2021; Zhao et al., 2021), acid rain (Owoade et al., 73 2021), and very serious human health impacts (Croft et al., 2019; Hopke et al., 2019; 74 Shen et al., 2020; Yan et al., 2018), such as pneumonia, acute respiratory disorders, 75 breathing problems, and chronic asthma (Rizwan et al., 2013). The global burden of 76 77 disease project (GDB, 2020) has estimated that about 4.2 million premature deaths per 78 annum around the globe are the result of exposure to air pollution.

Meteorological factors play major roles in the formation, accumulation, and dispersion of air pollutants (Hu et al., 2016; Islam et al., 2015; Mao et al., 2022; Okimiji et al., 2021; Owoade et al., 2021; Sulaymon et al., 2021a, 2021b, 2021c). Previous studies have posited that the atmospheric pollution dispersion is greatly influenced by relative humidity (RH), wind speed (WS), and planetary boundary layer height (PBLH) (Li et al., 2019; Liu et al., 2017; Zhang et al., 2019; Zhang et al., 2018), and that low WS
and PBLH hinder atmospheric pollutant dispersion and consequently lead to severe air
pollution episodes (Dai et al., 2020; Li et al., 2019; Liu et al., 2017; Sulaymon et al.,
2021a; Wang et al., 2020; Zhang et al., 2019).

Prior to January 2021, there were no ambient air quality monitoring networks in 88 89 any of Nigerian cities. The lack of ambient data meant that the temporal and spatial variations of the major air pollutants (gaseous pollutants and particulate matter and its 90 components) were unknown. Thus, studies to investigate the air quality and support 91 92 development of effective pollution control strategies and reduce associated human health risks were very limited (Kitagawa et al., 2022; Hu et al., 2016). However, the problems 93 associated with the air quality assessment in countries/regions with no or limited 94 monitoring networks could be overcome through the use of source-tracking chemical 95 transport models (CTMs) (Kitagawa et al., 2022). CTMs can simulate air pollutants with 96 high resolution of temporal and spatial distributions (Kota et al., 2018), give hourly 97 averaged gridded concentrations (Kitagawa et al., 2022), be used for source 98 99 apportionment studies (Appel et al., 2020; Li et al., 2021; Ma et al., 2021; Shi et al., 100 2017), employed in human exposure assessment to air pollution (Guo et al., 2019; Reis et al., 2018), as well as utilized in epidemiological studies to unravel the adverse effects of 101 air pollution on population (Andreão et al., 2020). CTMs have been extensively used in 102 103 many parts of the world, such as North America (Guo et al., 2018; Hu et al., 2015a; Pan et al., 2017; Ying and Kleeman, 2006; Wang et al., 2021a; Ying et al., 2015; Zhang et al., 104 2013, 2014), China (Hu et al., 2015b, 2016, 2017; Gong et al., 2021; Ma et al., 2021; 105 106 Qiao et al., 2015, 2019; Sulaymon et al., 2021a, 2021b; Wang et al., 2020; Ying et al.,

2014), India (Guo et al., 2017, 2019; Guttikunda and Jawahar, 2014; Kota et al., 2014,
2018; Marrapu et al., 2014; Ye et al., 2022), South America (Albuquerque et al., 2018,
2019; Kitagawa et al., 2021, 2022; Nedbor-Gross et al., 2018; Pedruzzi et al., 2019, 2022),
and Africa (Kumar et al., 2022; Marais et al., 2014, 2019).

With a population of about 23.5 million as at 2018, Lagos is the largest 111 112 metropolitan city in Africa, and also the most economically and industrially-developed city in Nigeria. As a result, the city has been suffering from severe fine particulate matter 113 114 (PM_{2.5}) pollution for more than two decades. To design effective emissions control 115 strategies towards abating $PM_{2.5}$ pollution in any region, it is imperative to elucidate the 116 contributions of various emissions sectors to $PM_{2.5}$ and its major components, as well as the meteorological influences over the region. The present work represents the first study 117 that utilized the source-tracking Community Multiscale Air Quality (CMAQ) model to 118 quantify the contributions of different emissions sectors to PM2.5 and its major 119 120 compositions in Lagos during a prolonged severe atmospheric pollution episode (APE) in January 2021. The capability and fidelity of the CMAQ model in simulating PM_{2.5} in 121 Lagos was validated by comparing the predicted concentrations with observation data, 122 123 and the spatiotemporal distributions of PM_{2.5} during the APE period were analyzed. In 124 addition, the impacts of meteorological conditions on $PM_{2.5}$ during the APE were 125 elucidated. This study provides insights into the emissions sectors dominating $PM_{2.5}$ 126 pollution in Lagos and the meteorological influences, and the results could serve as scientific basis for formulating cost-effective emissions control strategies towards 127 128 mitigating PM_{2.5} pollution in Lagos and the surrounding regions.

130 2. Methodology

131 2.1. Model configurations

The Community Multiscale Air Quality version 5.3.2 (CMAOv5.3.2) model was 132 employed to simulate the air quality in Lagos, Nigeria. The required meteorological fields 133 for the CMAQ model were simulated using the Weather Research and Forecasting (WRF) 134 135 model version 4.0. The initial and boundary conditions (IC/BC) of WRF model were based on the dataset of NCEP FNL (Final) Operational Model Global Tropospheric 136 Analysis. The major physics schemes are listed in Table S1. Further detailed settings and 137 138 configurations of WRF model adopted in this study can be found in previous studies (Hu et al., 2015b, 2016; Wang et al., 2021b). In this study, the simulations were conducted in 139 two domains using one-way nesting approach (Fig. 1). Domain 1 covers the entire 140 Nigeria with a horizontal resolution of 36 km x 36 km (137 x 107 grids). Domain 2 141 mostly covers Lagos and surrounding areas at with a horizontal resolution of 12 km x 12 142 km (127 x 202 grids). The model was configured with the State-wide Air Pollution 143 Research Center version 07 (SAPRC07tic) photochemical mechanism and the AERO6i 144 aerosol module (Sulaymon et al., 202Ia, 2021b). Details about the vertical resolution of 145 146 the two CMAQ domains have been described in Sulaymon et al. (2021b). The simulation period was January 2021, during which two severe APEs occurred. The IC/BC of 147 Domain 1 simulation were generated using the default profiles provided by the CMAQ 148 149 model. The IC/BC used for the 12 km domain simulation were estimated based on the results of the nested simulation. To minimize the effects of initial conditions on the 150 model predictions, the simulation started five days ahead of the simulation period, hence, 151 152 regarded as the spin-up period (Tao et al., 2020).



Fig. 1. The WRF-CMAQ simulation domains used in this study (left) and the map of
Lagos and surrounding regions (right).

157 2.2. Emissions inventory

In this study, the Integrated Source Apportionment Model (ISAM), a source 158 159 tracking tool in the CMAQ model, was applied to quantify the source contributions to PM_{2.5} and its major components (Jiang et al., 2021; Kwok et al., 2013; Li et al., 2021) in 160 Lagos. The anthropogenic emissions used in this study were derived from the Emissions 161 Database for Global Atmospheric Research version 5.0 (EDGARv5.0) (Crippa et al., 162 2020). The EDGAR emissions inventory includes the annual emissions of carbon 163 monoxide (CO), nitrogen oxides (NO_x), sulfur dioxide (SO₂), ammonia (NH₃), non-164 methane volatile organic compounds (NMVOCs), PM2.5, particulate matter with an 165 aerodynamic diameter less than 10 μ m (PM₁₀), elemental carbon (EC), and organic 166 carbon (OC) from various emissions sectors and with a spatial resolution of $0.1^{\circ} \times 0.1^{\circ}$. 167 The emissions sectors were subsequently classified into six categories (agriculture, 168 residential, industry, energy, on-road transportation, and off-road transportation). It is 169 worthy to mention that the EDGAR emissions inventory had been used in numerous 170 previous studies in India (Guo et al., 2019, 2018, 2017; Kota et al., 2018), China (Qiao et 171

172 al., 2019), and Africa as a continent (Kumar et al., 2022; Mazzeo et al., 2022). In addition, biogenic emissions were estimated with the Model of Emissions of Gases and Aerosols 173 174 from Nature (MEGAN) version 2.1 (Guenther et al., 2012). Also, open burning emissions were generated based on the data obtained from the Fire Inventory from NCAR (FINN) 175 (Wiedinmyer et al., 2011). Furthermore, sea salt and windblown emissions were 176 generated inline (Sulaymon et al., 2021a, 2021b). Overall, a total of eight emission 177 sectors were tracked in the ISAM model. Further details regarding the emission 178 179 processing can be found in Qiao et al. (2019).

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181 **3.** Results and discussion

182 *3.1. WRF model performance*

Previous studies have elucidated the crucial roles of meteorological factors in the 183 formation, transportation, as well as dissipation of fine particulate matter and other air 184 185 pollutants (Hua et al., 2021; Hu et al., 2016; Sulaymon, et al., 2021a, 2021d). Therefore, in assuring the accuracy of air quality simulations, a robust and satisfactory WRF model 186 performance must be obtained. In evaluating the performance of WRF model, the 187 188 simulated temperature (T2) and relative humidity (RH) at 2 m above surface, and wind 189 speeds (WS) and wind directions (WD) at 10 m above ground level were compared with 190 observation data downloaded from the official website of the National Climate Data 191 Center (NCDC) (ftp://ftp.ncdc.noaa.gov/pub/data/noaa/, last accessed on October 30, 2022). The temporal variations of the meteorological variables and PM_{2.5} are illustrated 192 193 in Fig. 2. Table 1 provides the statistical indices, including the mean observation (OBS), 194 mean prediction (PRE), mean bias (MB), mean error (ME), and the root mean square



196 the suggested

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Fig. 2. Temporal variations of the predicted and observed meteorological variables (temperature, wind speed, wind direction, and relative humidity) and $PM_{2.5}$ concentrations in Lagos during January 2021.

benchmarks (MB≤±0.5; ME≤2.0) (Emery et al., 2001), T2 (Fig. 2a) was well-reproduced
by the WRF model. Similar statistical indices and model performance of T2 had been
reported in the Beijing-Tianjin-Hebei (BTH) region of China during winter (January
205 2014) (Lang et al., 2021). Wind speed was slightly over-estimated during the study
period, however, considering the fact that its MB, ME, and RMSE indices were far below
the suggested benchmarks (Table 1), the WRF model in this study showed a better
performance in capturing the observed WS as illustrated in Fig. 2(b).

Table 1. Model performance of meteorological factors in Lagos, Nigeria (OBS: observed

210 mean; PRE: predicted mean; MB: mean bias; ME: mean error; RMSE: root mean square211 error).

	Metrics	January	Benchmarks
T2 (K)	OBS	301.6	
	PRE	301.8	
	MB	0.24	<u>≤</u> ±0.5
	ME	0.6	≤2.0
	RMSE	0.78	
WS (m/s)	OBS	2.87	
	PRE	2.90	
	MB	0.03	<u>≤</u> ±0.5
	ME	0.14	≤2.0
	RMSE	0.17	≤2.0
WD (°)	OBS	226.3	
	PRE	228.5	
	MB	2.17	<u>≤</u> ±10
	ME	5.69	<u>≤</u> ±30
	RMSE	9.90	
RH (%)	OBS	72.6	
	PRE	68.6	
	MB	-4.04	
	ME	4.55	
	RMSE	6.21	

213 The over-estimation of WS might be due to unresolved topography in the WRF model (Li et al., 2014). The statistical indices (MB and ME) of WS in this study are consistent with 214 those reported by Lang et al. (2021) and Sulaymon et al. (2021b). As illustrated in Fig. 215 216 2(c), the predominant WD during the study period was southwesterly. Similar to T2 and WS, WD was also over-predicted during the simulation period. However, the predicted 217 218 WD showed better agreement with observations, having found both MB and ME indices falling below the suggested benchmarks (MB <= 10; ME <= 30). Similar model 219 performance had been reported by Ma et al. (2021) and Qiao et al. (2019). Relative 220 humidity (Fig. 2d) was underpredicted with MB and ME values of -4.04 and 4.55, 221 respectively. Although, there are presently no performance benchmarks for RH. Previous 222 studies in India (Bhati and Mohan, 2018), China (Hu et al., 2016; Li et al., 2021; 223 Sulaymon et al., 2021a, 2021b), and Brazil (Kitagawa et al., 2021; Pedruzzi et al., 2021) 224 had also reported under-estimation of RH. Bhati and Mohan (2018) had linked under-225 estimation of RH to the influence of boundary layer parameterization on meteorological 226 prediction. Generally, the WRF model performances in the present study were 227 satisfactory and better when compared to previous studies from other parts of the world 228 229 (Bhati and Mohan, 2018; Hu et al., 2016; Kitagawa et al., 2021; Kota et al., 2018; Pedruzzi et al., 2021; Sulaymon et al., 2021a; Yu et al., 2021). Since the WRF model has 230 shown a robust and better performance in this study, the meteorological fields were 231 232 utilized in driving the air quality simulation.

233 *3.2. CMAQ model performance*

To evaluate the performance of the CMAQ model in reproducing the observed PM_{2.5} in Lagos, the model was evaluated based on some statistical indices, including the

236	mean observations (OBS), mean predictions (PRE), mean fractional bias (MFB), mean
237	fractional error (MFE), mean normalized bias (MNB), and mean normalized error (MNE)
238	The hourly $PM_{2.5}$ observation data were downloaded from the official website of the
239	United States of America (USA) Embassy, Lagos
240	(https://www.airnow.gov/international/us-embassies-and-consulates/#Nigeria\$Lagos, last
241	accessed on October 30, 2022). Necessary quality checks were carried out on the data
242	before being used for evaluation (Sulaymon et al., 2021c). The CMAQ model
243	performances in predicting PM _{2.5} during January 2021 in Lagos are illustrated in Table 2.
244	The observed $PM_{2.5}$ concentration was well-simulated in Lagos, with the model
245	performance indices falling within the recommended benchmarks (MFB $\leq \pm 0.60$; MFE \leq
246	0.75) (Boylan and Russel, 2006). The temporal variations of the observed and simulated
247	$PM_{2.5}$ concentrations are illustrated in Fig. 2(e). With positive MFB (0.52), the CMAQ
248	model over-predicted $PM_{2.5}$ concentrations during the study period. Overall, the CMAQ
249	model in this study has exhibited a good performance when compared to previous studies
250	around the world (Jiang et al., 2021; Kitagawa et al., 2021; Kota et al., 2018; Mao et al.,
251	2022; Pedruzzi et al., 2021; Sulaymon et al., 2021a; Tao et al., 2020). Therefore, the
252	predicted PM _{2.5} concentrations were deemed acceptable for further analyses.

Table 2. Model performance of PM_{2.5} in Lagos, Nigeria (OBS: observed average; PRE:
predicted average; MFB: mean fractional bias; MFE: mean fractional error; MNB: mean
normalized bias; MNE: mean normalized error). The study period was January 2021. The
performance benchmarks for PM_{2.5} were suggested by Boylan and Russell (2006).

	Metrics	January	Benchmarks
$PM_{2.5} (\mu g/m^3)$	OBS	56.4	
	PRE	86.2	
	MFB	0.52	≤±0.60
	MFE	0.69	≤ 0.75
	MNB	1.09	
	MNE	1.21	

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260 *3.3. Statistical analysis of PM*_{2.5} *during different pollution levels*

As illustrated in Fig. 2 (e), it can be observed that throughout the simulation 261 period (except on January 2 and 3), the 24-hr total PM_{2.5} concentrations in Lagos greatly 262 exceeded the old daily air quality guideline (AQG) level (25 µg/m³) recommended by the 263 World Health Organization (WHO, 2005). Furthermore, considering the new WHO AQG 264 level (15 $\mu\text{g/m}^3)$ (WHO, 2021) for PM_{2.5}, only one day (January 2 with PM_{2.5} value of 265 11.92 μ g/m³) could be described as a clean day, while the remaining days witnessed 266 different levels of PM_{2.5} pollution, ranging from slight to severe pollution. Based on the 267 268 daily PM_{2.5} concentrations, air quality can be grouped into various pollution levels. In 269 this study, the $PM_{2.5}$ pollution levels were categorized into five groups (Table S2). Table 270 S3 shows the statistical results of $PM_{2.5}$ in January based on different pollution levels. Under clean level, the simulated PM_{2.5} concentrations were below the old WHO AQG 271 level. However, the simulated maximum PM2.5 concentration exceeded the new AQG 272 value of WHO, indicating that even during the clean days, exposure to PM2.5 at low 273 274 levels could still cause adverse effects to human health. Under the slightly-polluted level, there were 12 days with concentration range of $35 \le PM_{2.5} \le 75$. The simulated minimum, 275 maximum, and mean PM_{2.5} greatly breached the old and new AQG levels of WHO. For 276 277 instance, the simulated mean PM_{2.5} was 2.5 and 4.2 times the old and new AQG level, respectively. Considering the moderately-polluted level (10 days with concentration 278

279 range of $75 \le PM_{2.5} \le 115$), it could be noted that the simulated mean $PM_{2.5}$ was 3.7 and 6.2 times the old and new AQG level, respectively. Under the heavily-polluted level, the 280 simulated mean PM_{2.5} was 5.4 and 9.1 times the old and new AQG level, respectively. 281 The simulated PM_{2.5} ranged between 167.3-198.6 μ g/m³, with mean of 182.9 μ g/m³. 282 PM_{2.5} under the heavily-polluted level greatly exceeded the old and new AQG levels by 283 284 7.3 and 12.2 times, respectively. The results of this study showed that the residents of Lagos were greatly exposed to high PM_{2.5} pollution throughout January 2021, and this 285 could lead to adverse human health effects (Croft et al., 2019; Hopke et al., 2019; Shen et 286 al., 2020; Yan et al., 2018). 287

288 3.4. Spatial variations of $PM_{2.5}$ and its major components during the pollution 289 episode

The spatial distributions of the simulated PM2.5 concentrations and its major 290 components during the study period are illustrated in Fig. 3. An atmospheric pollution 291 episode (APE) is said to occur when daily PM_{2.5} concentration persistently exceeds 50 292 µg/m³ (interim target 2 of WHO) (WHO, 2021) for at least two consecutive days 293 (Sulaymon et al., 2021b; Zhang et al., 2019). An APE occurred in January 2021, and was 294 characterized with high PM_{2.5} concentrations. The APE (50 μ g/m³ \leq PM_{2.5}) lasted for 27 295 days (January 5-31). Spatially, highest PM_{2.5} concentrations (PM_{2.5} \ge 120 µg/m³) were 296 noted in the northwestern region of Lagos metropolis, while the rest of the city was 297 characterized with $60 \le PM_{2.5} \le 120 \ \mu g/m^3$. During the prolonged pollution episode, the 298 averaged city-wide concentration was 94.6 μ g/m³, while PM_{2.5} concentrations in the 299 northwest were as high as 150-220 μ g/m³. This indicates the severity of PM_{2.5} pollution 300 during the APE. 301

302 Among the three components that make the secondary inorganic aerosols (SIA), sulfate (SO_4^{2-}) had the highest contributions, followed by ammonium (NH_4^+) , and nitrate 303 (NO₃⁻). Sulfate was evenly distributed across the city, with higher concentrations (6-15 304 $\mu g/m^3$) located in the northwestern and upper northern areas. For ammonium, higher 305 concentrations (2-3 μ g/m³) were found in the upper north and northwest areas, while 306 other areas were concentrated with 0.8-2 μ g/m³ of NH₄⁺. Similar to sulfate and 307 ammonium, nitrate also peaked in the northwest area. It can be deduced that among the 308 three SIA species, sulfate contributed most to PM_{2.5}. 309



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Fig. 3. Spatial distributions of the predicted total $PM_{2.5}$ and its major components in Lagos during the atmospheric pollution episode. Units are $\mu g/m^3$.

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Overall, SIA (sum of SO_4^{2-} , NO_3^{-} , and NH_4^{+}) was evenly distributed over Lagos, with 314 higher concentrations (6-15 μ g/m³) located in the northwestern, northern, and central 315 areas. In addition, during the pollution episode, the concentrations of elemental carbon 316 (EC) and primary organic carbon (POC) were notably very high in the northwestern area 317 of Lagos. The northwestern area (which includes Ikeja, Agege, Ifako-Ijaye, Oshodi, 318 319 Mushin, Shomolu, Surulere, and Ajegunle areas) is densely populated and highly industrialized (Owoade et al., 2013), which results to high energy consumption. In 320 addition, the Muritala Muhammed International Airport (MMIA), which is the busiest 321 322 and largest international airport in Nigeria and West Africa, is located in the northwestern part of Lagos. Generally, Lagos is characterized with heavy daily traffic (Okimiji et al., 323 324 2021; Owoade et al., 2013), especially in the early morning and late afternoon (the local 325 rush hours). The northwestern region records more traffic than any other region. High traffic volumes can be attributed to the presence of the administrative offices of several 326 327 governmental agencies as well as those of many private enterprises. Therefore, emissions from local anthropogenic sources particularly in the northwestern region of the city are 328 likely the dominant contributors to the elevated PM_{2.5} concentrations. However, polluted 329 winds being transported from the neighboring cities in Ogun State, which borders Lagos 330 in the north could also aggravate the PM_{2.5} pollution, especially in the northwestern area 331 of Lagos. 332

333 3.5. Spatiotemporal variations of $PM_{2.5}$ during the pollution episode

Fig. 4 shows the daily spatial variations of the predicted $PM_{2.5}$ during the APE in Lagos and its neighboring cities. Except in the northwestern area of Lagos, $PM_{2.5}$

concentrations in most parts of the city were less than 100 μ g/m³ during the first 14 days 336 of the episode (January 5-18). This could be attributed to elevated PBLH during the 337 period (Fig. S1), since high PBLH values lead to reduced pollution level (Tao et al., 338 2020). The higher PBLH were generally noted within Lagos metropolis and its 339 surroundings during January 5-18. Within Lagos, the PBLH ranged between 200 and 850 340 m, with an average regional value of 560 m. In the northwestern region, however, a very 341 intense pollution episode was observed throughout the period (January 5-18), with 100 342 $\mu g/m^3$ being the 343



Fig. 4. Spatial distributions of the predicted total PM_{2.5} concentrations in Lagos during the atmospheric pollution episode. Units are $\mu g/m^3$.

347 minimum regional daily PM_{2.5} concentration. As stated in section 3.4, the northwestern region of Lagos is an urban area with several chemical industries and heavy traffic. Thus, 348 it is a major hub of anthropogenic emissions. During the first four days (January 5-8) of 349 the episode, high wind speeds (3-6 m/s) were observed in most of Lagos metropolis, 350 especially in the northwestern and central areas (Fig. S2). The APE began on January 5 351 (Fig. 4) with high haze ($50 \le PM_{2.5} \le 120 \ \mu g/m^3$) noted across the major parts of the 352 study area except at the northwestern area where a cluster of very high pollution (150 \leq 353 $PM_{2.5} < 200 \ \mu g/m^3$) was found. Prior to the inception of the city-wide heavy haze on 354 January 6, a high PM_{2.5} pollution cluster was initially noted in the northwestern area on 355 January 5 (Fig. 4). On January 6, a strong northwesterly wind (Fig. S2) enhanced the 356 357 transportation of pollutants to other parts of the Lagos metropolis. Due to the continuous flow of the northwesterly winds, the $PM_{2.5}$ pollution cluster that was initially formed in 358 the northwestern area on January 5, steadily got spread to and enveloped the entire study 359 area, with regional PM_{2.5} concentrations exceeding 100 μ g/m³ on January 6 (Fig. 4). This 360 trend persisted until January 8. On several days (e.g. January 9 and 11), the pollution was 361 low (PM_{2.5} \leq 50 µg/m³) in most areas of Lagos (except the northern and central areas), 362 especially in the western and eastern parts of the city. With different pollution intensities 363 across Lagos and its surrounding cities, the APE continued till January 31. As previously 364 discussed and illustrated in Fig. 4, there were tendencies of cross-regional transport of 365 366 PM_{25} pollution from the surrounding cities (bordering Lagos to the north) into the study area. The transported pollution could have aggravated the pollution level generally 367 caused by local anthropogenic emissions. Compared to January 16 (with moderate 368

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pollution), higher pollution was noted across the Lagos metropolis on January 17 and 18, with the highest being found in the northwestern area of the city.

371 Compared to the first 14 days, the PM_{2.5} concentrations across Lagos metropolis 372 and specifically in the northwestern area were relatively higher during the last 13 days of the episode (January 19-31) (Fig. 4). During this period (January 19-31), the wind speed 373 374 in most of Lagos ranged between 2-4 m/s (Fig. S2), with the lowest values being observed in the northwestern area. The lack of dispersion could have driven the severe 375 haze recorded in the northwestern region as lower wind speed reduces the dispersion of 376 pollutants (Sulaymon et al, 2021a). Generally, the PBLH within Lagos metropolis was 377 lower during January 19-30 compared to January 5-18. During January 19-30 of the APE, 378 the daily averaged PBLH was between 150-800 m, having an average regional value of 379 491 m. Generally, the formation of high $PM_{2.5}$ pollution is enhanced by low PBLH and 380 WS (Li et al., 2019; Sulaymon et al, 2021a; Tao et al., 2020; Wang et al., 2020). For 381 instance, on January 19, a high PM_{2.5} concentration was noted in northwestern Lagos (Fig. 382 4). Due to the stagnant and low wind speeds (especially during January 19-25) across the 383 Lagos metropolis and specifically in the northwestern region, the APE persisted until 384 385 January 31. The pollution episode became more significant on January 21, with average regional PM_{2.5} reaching 120 μ g/m³, while the concentrations in the northwestern region 386 exceeded 200 μ g/m³. The severe pollution continued until January 24 when it peaked 387 with the PM_{2.5} concentrations above 150 μ g/m³ widely spread across the Lagos 388 metropolis and its surrounding cities in the north. The highest concentrations ($PM_{2.5}>200$ 389 $\mu g/m^3$) were not only found in the northwestern but also in the southwestern and southern 390 parts of Lagos nearest to the ocean. On January 25, severe haze was still recorded in the 391



392 northwestern and southern parts of Lagos, although lower than January 24. The other 393 areas had $PM_{2.5}$

concentrations ranging between 60 and 120 μ g/m³, and the haze persisted until January 397 31 when the APE ended with a decline to moderate to high regional pollution. With 27 398 days characterized with high PM_{2.5} concentrations during the episode, Lagos was

subjected to severe $PM_{2.5}$ pollution during January 2021. Therefore, quantifying the contributions from each emission sector to total $PM_{2.5}$ and its major components during the APE became highly imperative, as the results would provide scientific basis for designing and implementing effective emission control strategies towards abating $PM_{2.5}$ pollution in the city.

404 3.6. Source apportionment of $PM_{2.5}$ and its major components

The temporal variations of the contributions of different sources to daily PM_{2.5} 405 and its major components in Lagos during the APE are illustrated in Fig. 5. During the 406 study period, the averaged contributions of residential, industry, open burning, and IC/BC 407 to total PM_{2.5} (Fig. 5a) in Lagos were 36.2, 16.5, 0.78, and 3.57 μ g/m³, respectively. The 408 three major sources of EC (Fig. 5b) were residential, industry, and ICBC, with averaged 409 contributions of 18.9, 4.46, and 1.00 μ g/m³, respectively. Following the same 410 arrangement of the sources for EC, the contributions of the sectors towards SO_4^{2-} (Fig. 5c) 411 were 3.71, 1.43, and 1.25 μ g/m³, respectively. In addition to the three major sources of 412 SO₄²⁻, there were also slight contributions from energy and open burning sectors on most 413 of the days of the episode. For NH_4^+ (Fig. 5d), residential and industry were the dominant 414 sectors with averaged contributions of 1.04 and 0.27 μ g/m³, respectively. The 415 contributions from on-road transport, off-road transport, energy, agriculture, and biogenic 416 sectors to $PM_{2.5}$, EC, and NH_4^+ were very low. Considering NO_3^- (Fig. 5e), all of the 417 emission sectors contributed towards its daily formation, with residential, industry, and 418 on-road serving as the three major contributors. It should be noted that the contributions 419 of residential and industry sectors were generally higher during the last 13 days of the 420 episode than the first 14 days of the episode. 421





Fig. 6 illustrates the spatial distributions of various source contributions to the averaged total $PM_{2.5}$ in Lagos during the episode. High contributions from residential (20-40 µg/m³) and industry (10-20 µg/m³) were found in the northwestern area of the city. In other areas of Lagos, the contributions of residential and industrial emissions were up to 20 µg/m³ and 10 µg/m³, respectively. This is consistent with the results of Guo et al. (2017).



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Fig. 6. Source apportionment of total $PM_{2.5}$ from (a) residential, (b) industry, (c) on-road, (d) off-road, (e) energy, (f) agriculture, (g) biogenic, and (h) open burning during the atmospheric pollution episode. Units are $\mu g/m^3$.

Guo et al. (2017) had previously used EDGAR emissions inventory in a source-oriented 437 CMAQ model for the source apportionment of PM2.5 in North India, and found 438 439 residential emissions as the highest contributor, followed by industrial sector. In addition, the elevated PM2.5 concentrations in major Chinese cities during winter period had been 440 linked to residential burning for heating purposes, industrial emissions, construction 441 442 activities, and road dust (Guo et al., 2020). PM_{2.5} contributed by open burning was up to $1 \,\mu\text{g/m}^3$, and uniformly distributed across the city. Energy also contributed up to $1 \,\mu\text{g/m}^3$, 443 especially in the upper northern areas The contributions from other emissions sectors to 444 $PM_{2.5}$ during the pollution episode were low (less than 1 µg/m³) across the city. 445

The percentage contributions of emission sectors to PM_{2.5} and its major 446 components during the APE are shown in Fig. 7, with residential and industrial sectors 447 dominating. The contributions of residential to $PM_{2.5}$, EC, SO_4^{2-} , and NH_4^+ were 66.7, 448 80.3, 65.3, and 79.1%, respectively, while the industry contributed 30.4, 19.0, 25.1, and 449 20.4% to PM_{2.5}, EC, SO₄²⁻, and NH₄⁺, respectively. Energy and open burning also 450 notably contributed to $SO_4^{2^-}$, with 4.0 and 3.1%, respectively. For NO_3^- , the contributions 451 of residential, industry, on-road, biogenic, open burning, agriculture, off-road, and energy 452 453 were 41.3, 16.8, 16.8, 10.0, 6.5, 3.4, 3.3, and 2.0%, respectively. These findings are consistent with the results of Zhao et al. (2021), where industrial and residential sectors 454 were the major contributors to PM_{2.5} in six cities of East-central China during the 455 COVID-19 lockdown. The dominance of residential sector could be attributed to the 456 cooking activities by the dense population of Lagos residents, while the dominance of 457 industrial sector could be associated with emissions from large-scale industries in the city. 458 Also, Li et al. (2020) and Ma et al. (2021) had reported residential and industrial sectors 459

460 as the dominant contributors to $PM_{2.5}$ in YRD region, China. Furthermore, residential and 461 industry were the major emission sectors contributing to $PM_{2.5}$ in the BTH region of 462 China, especially during winter season (Chang et al., 2019).



464 **Fig. 7.** Contributions of different emissions sectors to total $PM_{2.5}$ and its major 465 components during the atmospheric pollution episode.

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467 3.7. Source apportionment of secondary inorganic aerosols (SIA)

Previous studies have identified the secondary inorganic aerosols (sulfate, nitrate, and ammonium) as the major components of total $PM_{2.5}$ (Guo et al., 2017; Li et al., 2021; Shi et al., 2017). The contributions of different emissions sectors to SIA are shown in Fig. 8, while the source contributions to SO_4^{2-} , NO_3^{-} , and NH_4^{+} are illustrated in Figs S3, S4, and S5, respectively. Similar to $PM_{2.5}$, residential and industry served as the major emission sectors contributing to SIA. Considering the total contributions of all the emissions sectors to SIA during the APE (Fig. 8i), higher concentrations of SIA were majorly concentrated in the northwest, upper north, and central areas of Lagos, and were dominated by the contributions from residential (~9 μ g/m³) and industry (~3 μ g/m³) as illustrated in Figs 8(a) and 8(b), respectively, while other areas were filled with low but substantive SIA concentrations (3-6 μ g/m³) (Fig. 8i).





Fig. 8. Source apportionment of SIA from (a) residential, (b) industry, (c) on-road, (d) off-road, (e) energy, (f) agriculture, (g) biogenic, and (h) open burning during the atmospheric pollution episode. Units are $\mu g/m^3$.

484 High contributions from residential (2-9 μ g/m³) were located in the north and central 485 areas of Lagos, while up to 3 μ g/m³ contributions due to industry were located in the 486 northwest. The contributions from other emission sectors to SIA were very low. As

487 shown in Fig. S3, residential and industry were the dominant sources of sulfate. Sulfate concentrations from residential (up to $6 \mu g/m^3$) and industry (~2 $\mu g/m^3$) are concentrated 488 in the north of Lagos (particularly the northwest), as the region is densely populated and 489 highly industrialized. Residential, industry, and on-road transport sectors were the major 490 sources of nitrate (Fig. S4). Furthermore, only residential and industry served as the 491 major sectors contributing to NH_4^+ (Fig. S5). Just as the concentrations of $PM_{2.5}$ were 492 higher during the APE, higher SIA concentrations were also noted during the study 493 period. This reveals the severity of PM2.5 pollution in Lagos during the pollution episode 494 495 in January 2021. Overall, the substantive contributions of residential and industry sectors to PM_{2.5} and SIA during the APE suggests that controlling residential and industrial 496 emissions would pave way towards reducing PM_{2.5} and its major components in Lagos. 497

498 3.8. Influence of meteorological factors on $PM_{2.5}$ pollution

Previous studies have revealed that meteorological factors such as high RH, low 499 WS, and low PBLH hinder the dispersion of atmospheric pollutants, resulting into the 500 formation of severe air pollution, especially during the cold season (Li et al., 2019; Liu et 501 al., 2016; Sulaymon et al., 2021a; Wang et al., 2020; Zhang et al., 2019). As earlier 502 discussed, low PBLH (Fig. S1) and WS (Fig. S2) were noted across Lagos during the 503 study period. Therefore, the ventilation coefficient (VC) was used to evaluate the 504 effectiveness of atmospheric dispersion during the APE period. VC (in m^2s^{-1}) was 505 obtained as the product of WS and PBLH. Previous studies have widely employed VC in 506 evaluating the efficiency of atmospheric dispersion of air pollutants (Dai et al., 2020; 507 508 Sulaymon et al., 2021a; Tiwari et al., 2019), hence, further details about VC can be found in the referenced studies. The temporal variations of PM2.5 and VC in Lagos during 509

510 January are illustrated in Fig. S6. As a result of poor dispersion, low VC values (<2000 m^{2}/s) were generally noted during the APE period, an indication of very high PM_{2.5} 511 pollution. During January 2-3 and 6-9, however, highest VC values were observed, 512 indicating that the PM_{2.5} pollution during those days was low compared to the last 15 513 days of the month (especially January 17-30). Low VC enhanced the accumulation of 514 high PM_{2.5} concentrations (Dai et al., 2020; Tiwari et al., 2019), which subsequently led 515 to the prolonged high-pollution days during the study period, especially during January 516 17-30. For instance, the VC on January 2 (1844.1 m²s⁻¹) was the highest, and it was 517 approximately twice the VC on January 24 (1017.9 m²s⁻¹), when the APE attained its 518 peak. Correspondingly, the highest $PM_{2.5}$ concentration (208.7 $\mu g/m^3$) occurred on 519 January 24, the same day with the lowest VC. 520

Furthermore, the influence of meteorological factors on PM_{2.5} was assessed using 521 the Pearson correlation analysis between PM_{2.5} concentrations and four meteorological 522 parameters (WS, PBLH, RH, and T2) during the APE. PM2.5 was negatively correlated 523 with WS (r^2 =-0.535) and PBLH (r^2 =-0.539). Zhang et al. (2019) had reported negative 524 correlations between PM2.5 and WS and PBLH. Sulaymon et al. (2021c, 2021d) and 525 Zhang et al. (2015) also reported an inverse relationship between PM_{2.5} and WS. In 526 addition, the association between $PM_{2.5}$ and RH was negative (r^2 =-0.456). The negative 527 correlation found between RH and PM2.5 in this study is consistent with those previously 528 529 reported in Lagos (Okimiji et al., 2021), Wuhan (Sulaymon et al., 2021c), Dhaka (Islam et al., 2015), and Kathmandu (Giri et al., 2008). Negative correlations were also found 530 between PM_{2.5} and RH in Shanghai and Guangzhou (Zhang et al., 2015), especially 531 during the winter period. Considering the relationship between PM_{2.5} and temperature, 532

 $PM_{2.5}$ also exhibited negative correlation with T2 (r²=-0.347) during the APE, which is 533 contrary to the findings of Okimiji et al. (2021) and Zhang et al. (2015) during winter. 534 However, the inverse correlation found between T2 and PM_{2.5} in this study is consistent 535 with previous studies in Kathmandu (Giri et al., 2008) and Hefei and Suzhou (Sulaymon 536 et al., 2021d), and was attributed to the cooling effect of particulate matter as a result of 537 538 its negative radioactive forces (Islam et al., 2015). The results of this study also showed that the elevated PM2.5 concentrations in Lagos during the APE were aggravated by 539 unfavorable meteorological conditions. Therefore, when designing emission control 540 541 strategies towards reducing the level of PM_{2.5} in Lagos, it is very important to understand the crucial roles being played by both chemistry and meteorology in the formation, 542 transportation, and dispersion of air pollutants as revealed in this study. 543

544 4. Conclusions

This study investigated the source apportionment of PM_{2.5} and its major 545 components in Lagos during a prolonged severe atmospheric pollution episode in January 546 2021, using the CMAQ-ISAM model. The influence of meteorological factors on $PM_{2.5}$ 547 concentrations during the atmospheric pollution episode was also elucidated. The WRF 548 model reasonably and well-simulated the meteorological fields (temperature, wind speed, 549 wind direction, and relative humidity) for the CMAQ model, while the CMAQ model 550 exhibited good performances in reproducing the observed PM_{2.5} concentration during the 551 simulation period. Spatially, elevated PM_{2.5} concentrations were found in the 552 northwestern region of Lagos, an urban area with larger anthropogenic emissions. The 553 554 formation of elevated PM_{2.5} during the APE was greatly enhanced by low WS and PBLH. The results of the ISAM showed that residential and industry sectors were the major 555

contributors to PM_{2.5} and SIA concentrations in Lagos during the APE. Residential and 556 industry contributed about 40 μ g/m³ and 20 μ g/m³, respectively to total PM_{2.5}. SO₄²⁻ 557 accounted for the largest fraction of PM_{2.5}, attaining 6.0 μ g/m³, with residential, industry, 558 and energy being its major sources. Residential, industry, and on-road sectors dominated 559 the contributions to NO_3^{-} , while residential and industry served as the major sources of 560 NH4⁺. Also, this study improves the understanding of the mechanisms that resulted to the 561 prolonged atmospheric pollution episode in Lagos under diverse meteorological 562 influences. The results of this study reveal that the control of PM_{2.5} pollution in Lagos is 563 urgently required, and this could be achieved by substantially reducing the emissions 564 from residential and industry sectors in Lagos and its neighboring cities. 565

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569

570 **Conflict of Interest**

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

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573 **Open Research**

574 The code for the CMAQ model could be obtained from the website of the United States

575 Environmental Protection Agency (https://www.epa.gov/cmaq/access-cmaq-source-code,

last access: October, 2022) and the code of WRF model is available on the WRF users'

577 page (<u>https://www2.mmm.ucar.edu/wrf/users/download/get_sources.html</u>, last_access:

578 October, 2022). The Emissions Database for Global Atmospheric Research (EDGAR)

579 can be found online (https://edgar.jrc.ec.europa.eu/dataset ap50, last access: October, Fire from NCAR (FINN) download 2022). The INventory can be 580 (https://www.acom.ucar.edu/Data/fire/, last access: October 2022). The meteorological 581 observation is publicly available on the official website of the National Climate Data 582 Center (NCDC) (ftp://ftp.ncdc.noaa.gov/pub/data/noaa/, last access: October 2022). The 583 observation downloaded 584 hourly PM_{2.5} data can be online (https://www.airnow.gov/international/us-embassies-and-consulates/#Nigeria\$Lagos, last 585 access: October 2022). The simulated data (PM_{2.5}, its major components, and 586 587 meteorological factors) and the observation data (PM_{2.5} and meteorological factors) used for postprocessing (Figures and tables) in this study can be accessed online 588 (https://doi.org/10.5281/zenodo.7409425). All of the figures were created by the Python 589 Programming Language version 3.8 (https://www.python.org/downloads/, last access: 590 October 2022). 591

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Supplementary Material for

Modelling PM_{2.5} during severe atmospheric pollution episode in Lagos, Nigeria: Spatiotemporal variations, source apportionment, and meteorological influences

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Physics option	Scheme
Microphysics	Thompson scheme
Shortwave radiation	RRTMG scheme
Longwave radiation	RRTMG scheme
Surface layer	Revised MM5 Monin-Obukhov scheme
Land surface	Unified Noah land-surface scheme
Planetary boundary layer	YSU scheme
Cumulus parameterization	Grell-Freitas ensemble scheme

Table S1. The major physics options and the schemes used in the WRF model.

Table S2.	Concentration	range	under	different	PM _{2.5}	pollution	levels.

Levels	Clean	Slightly polluted	Moderately polluted	Heavily polluted	Severely polluted
Concentration ($\mu g/m^3$)	PM _{2.5} <35	$35 \le PM_{2.5} < 75$	$75 \le PM_{2.5} < 115$	$115 \le PM_{2.5} < 150$	$150 \le PM_{2.5}$

Table S3. Statistical results of PM_{2.5} under different pollution levels in Lagos during January 2021.

Clean			Slight		Moderate	2	Heavy		Severe		
(2 days)			(12 days)		(10 days)	(10 days)		(5 days)		(2 days)	
	Obs.	Sim.	Obs.	Sim.	Obs.	Sim.	Obs.	Sim.	Obs.	Sim.	
Minimum	15.7	11.9	44.3	48.8	72.5	79.1	121.7	117.4	161.8	167.3	
Maximum	23.7	17.3	76.7	72.4	109.5	107.0	148.2	143.9	214.6	198.6	
Mean	19.7	14.6	60.4	62.7	88.7	92.5	134.7	135.9	188.2	182.9	



Figure S1. Spatial distributions of the predicted planetary boundary layer height (PBLH) in Lagos during the atmospheric pollution episode. Units are m.



Figure S1. (continued).



Figure S2. Spatial distributions of the predicted wind speed at 10 m (WS10) in Lagos during the atmospheric pollution episode. Units are m/s.





Figure S3. Source apportionment of SO_4^{2-} from (a) residential, (b) industry, (c) on-road, (d) off-road, (e) energy, (f) agriculture, (g) biogenic, and (h) open burning during the atmospheric pollution episode. Units are $\mu g/m^3$.



Figure S4. Source apportionment of NO_3^- from (a) residential, (b) industry, (c) on-road, (d) off-road, (e) energy, (f) agriculture, (g) biogenic, and (h) open burning during the atmospheric pollution episode. Units are $\mu g/m^3$.



Figure S5. Source apportionment of NH_4^+ from (a) residential, (b) industry, (c) on-road, (d) off-road, (e) energy, (f) agriculture, (g) biogenic, and (h) open burning during the atmospheric pollution episode. Units are $\mu g/m^3$.



Figure S6. Temporal variations of $PM_{2.5}$ (µg/m³) and VC (m²/s) in Lagos during January 2021.