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Supplement of

Chemical composition, sources and formation mechanism of urban $PM_{2.5}$ in Southwest China: a case study at the beginning of 2023

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S1 WRF-Chem model setup and validation

S1.1 Model setup

WRF-Chem v4.2 was employed to analyze the causes of $PM_{2.5}$ pollution in Chengdu. Multiple two-way nested simulations were conducted at 27-, 9-, and 3-km resolutions. The 27-km grid domain (D01, 128×108) almost entirely covered China, and the 9-km grid domain (D02, 115×91) mainly covered Sichuan Province. The 3-km grid domain (D03, 61×52) included all areas of Chengdu (**Fig. S1**). There were 35 layers along the vertical direction, and the atmospheric pressure at the top of the model layer was 50 hPa. The initial and boundary conditions of the meteorological field were provided by the National Centers for Environmental Prediction (NCEP) reanalysis data with a resolution of 1°×1°. The chemical initial and boundary conditions relied on the output results of the Community Atmosphere Model with Chemistry (CAM-Chem). The underlying surface data were derived from 2013 MODIS data (Liu et al., 2018). The physical parameterization schemes included the Purdue Lin microphysics scheme (Chen and Sun, 2002), YSU planetary boundary layer scheme (Hong et al., 2006), Grell 3D ensemble cumulus parameterization scheme (Grell, 1993; Grell and Devenyi, 2002), Dudhia shortwave scheme (Dudhia, 1989), RRTM longwave scheme (Mlawer et al., 1997), Unified Noah Land Surface Model (Tewari et al., 2004), revised MM5 surface layer scheme (Jimenez et al., 2012) and single-layer urban scheme (Chen et al., 2011). The chemical schemes adopted in the simulations included the MOZART gas-phase chemical mechanism (Emmons et al., 2010), which yields the advantage of photochemical pollution simulation, MOSAIC with a 4-bin aerosol process (Zaveri et al., 2008) and the TUV photolysis mechanism (Madronich, 1987). The simulation period ran from 23 January to 7 February, for a total of 16 days, with the first 3 days being used as the spin-up time for the model.

The Multi-resolution emission inventory for China (MEIC) in 2017 was employed for anthropogenic emissions. In addition, biogenic emissions were provided by MEGAN (Guenther et al., 2006), and dust emissions relied on the GOCART scheme (Ginoux et al., 2004).

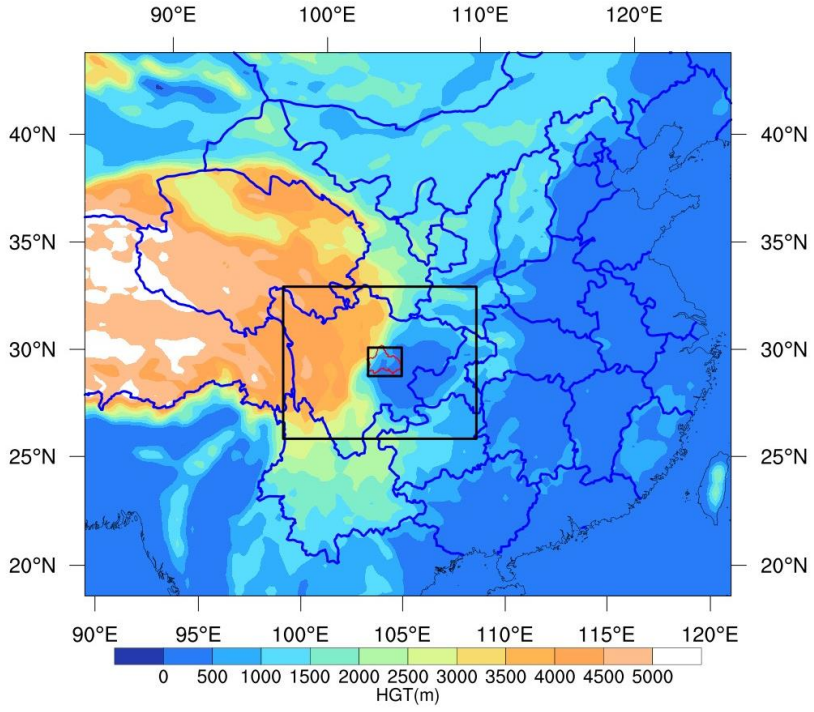

Figure S1: WRF-Chem simulation domain settings.

S1.2 Model validation

Model performance validation is a key step in all air quality model applications, i.e., to evaluate whether the model results can suitably reproduce the magnitude and spatiotemporal variation in the observed target pollutants (Huang et al., 2021). In this study, $PM_{2.5}$ concentration at the bottom of the vertical layer of the WRF-Chem model were compared to the observed values to verify the accuracy and reliability of the simulation results. The correlation coefficient (R), normalized mean bias (NMB) and normalized mean error (NME) were considered to evaluate the simulation results under the baseline scenario. R can reflect the model ability to capture temporal variations in observations, and NMB and NME can reflect the model ability in capturing the magnitude of observations (Huang et al., 2021). The equations to calculate these performance metrics are as follows:

$$R = \frac{\sum[(P_i-\bar{P})\times(O_i-\bar{O})]}{\sqrt{\sum(P_i-\bar{P})^2\times\sum(O_i-\bar{O})^2}} \quad (1)$$

$$\mathrm{NMB} = \frac{\sum(P_i-O_i)}{\sum O_i}\times 100 \quad (2)$$

$$\mathrm{NME} = \frac{\sum|P_i-O_i|}{\sum O_i}\times 100 \quad (3)$$

where P_i is the simulated value of hour i, and O_i is the observed value of hour i. The results of hourly $PM_{2.5}$ concentration evaluation are shown in **Fig. S2**.

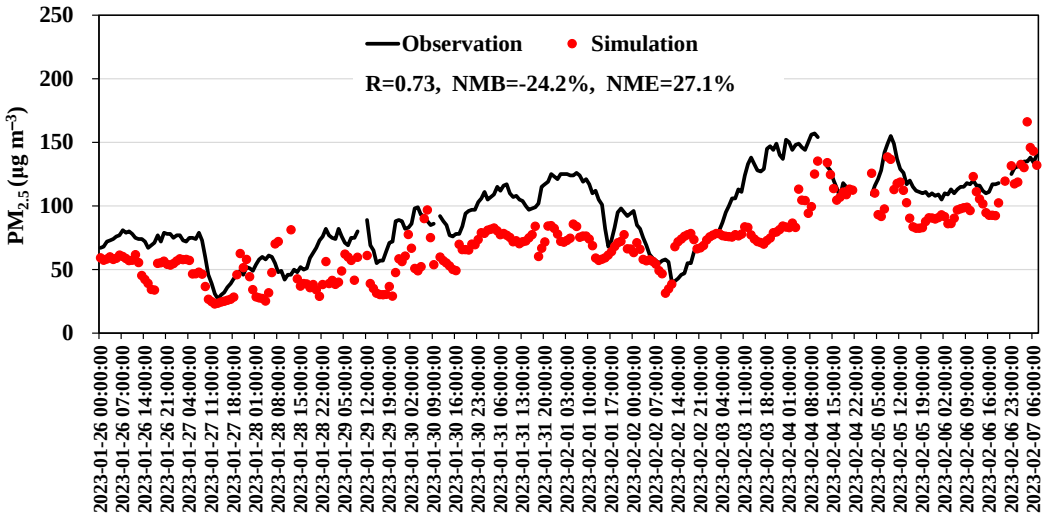

Figure S2: Temporal variation in the simulated and observed surface $PM_{2.5}$ concentration at the Chengdu station.

We found that the R value between the simulated and observed $PM_{2.5}$ concentrations was equal to 0.73 at the Chengdu station. this indicates a good correlation between the simulated and observed values. In addition, the rapid increase of $PM_{2.5}$ concentration during the two haze processes was also well reproduced. The simulated and observed NMB and NME values were –24.2% and 27.1%, respectively, indicating that the model underestimates the $PM_{2.5}$ concentrations. The R, NMB and NME of this study are consistent with the suggested parameter value intervals in Huang et al (2021), indicating that the simulation results can better respond to the current situation of $PM_{2.5}$ pollution in Chengdu, and the simulation results in this study can be used for the analysis of the causes of $PM_{2.5}$ pollution in Chengdu.

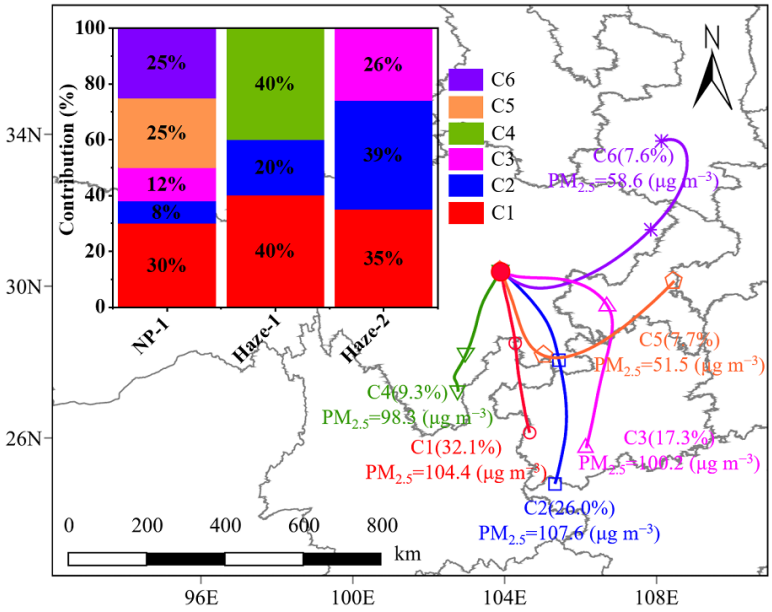

Figure S3: The clustering results of air masses during the study period and the corresponding $PM_{2.5}$ mass concentration for each cluster, and the bar chart shows the composition of air masses at different periods.

S2 Definition of different haze alarms

"Yellow" haze alarm: It is predicted that the daily average of air quality index (AQI) > 200 (or $PM_{2.5}$ mass concentration >115 μg m^{-3}) will last for 2 days (48 hours) or more, and did not meet higher level warning standards.

"Orange" haze alarm: It is predicted that the daily average of AQI > 200 will last for 3 days (72 hours) or more, or $PM_{2.5}$ mass concentration >115 μg m^{-3} will last for 3 days (72 hours) or more, and $PM_{2.5}$ concentration >150 μg m^{-3} will last for 1 day (24 hours) or more, and did not meet higher level warning standards.

"Red" haze alarm: It is predicted that the daily average of AQI > 200 will last for 4 days (96 hours) or more, and that the daily average of AQI > 300 will last for 2 days (48 hours) or more; Or it is predicted that the daily average of AQI will reach 500.

For detailed information on emergency response measures during different alarm periods, such as health protection guidance measures, initiative emission reduction measures and mandatory emission reduction measures, please refer to the Chengdu Ecological Environment Bureau (https://sthj.chengdu.gov.cn/cdhbj).

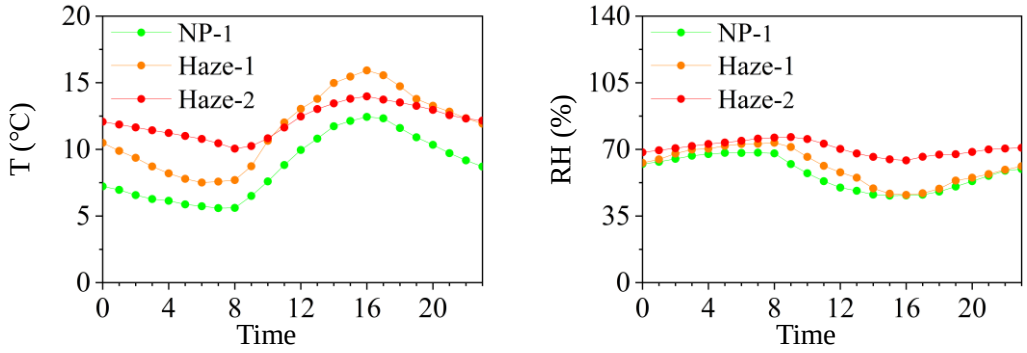

Figure S4: The diurnal variation of T and RH in different periods.

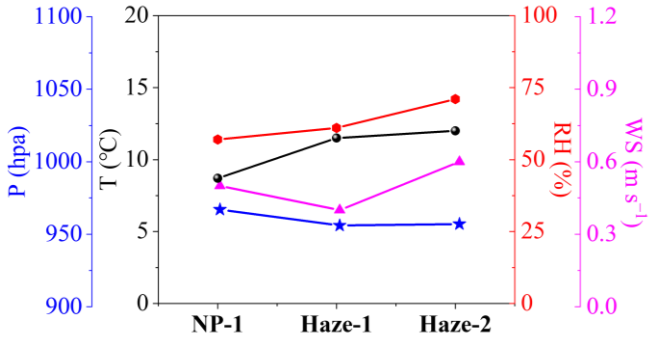

Figure S5: Average meteorological conditions in different periods.

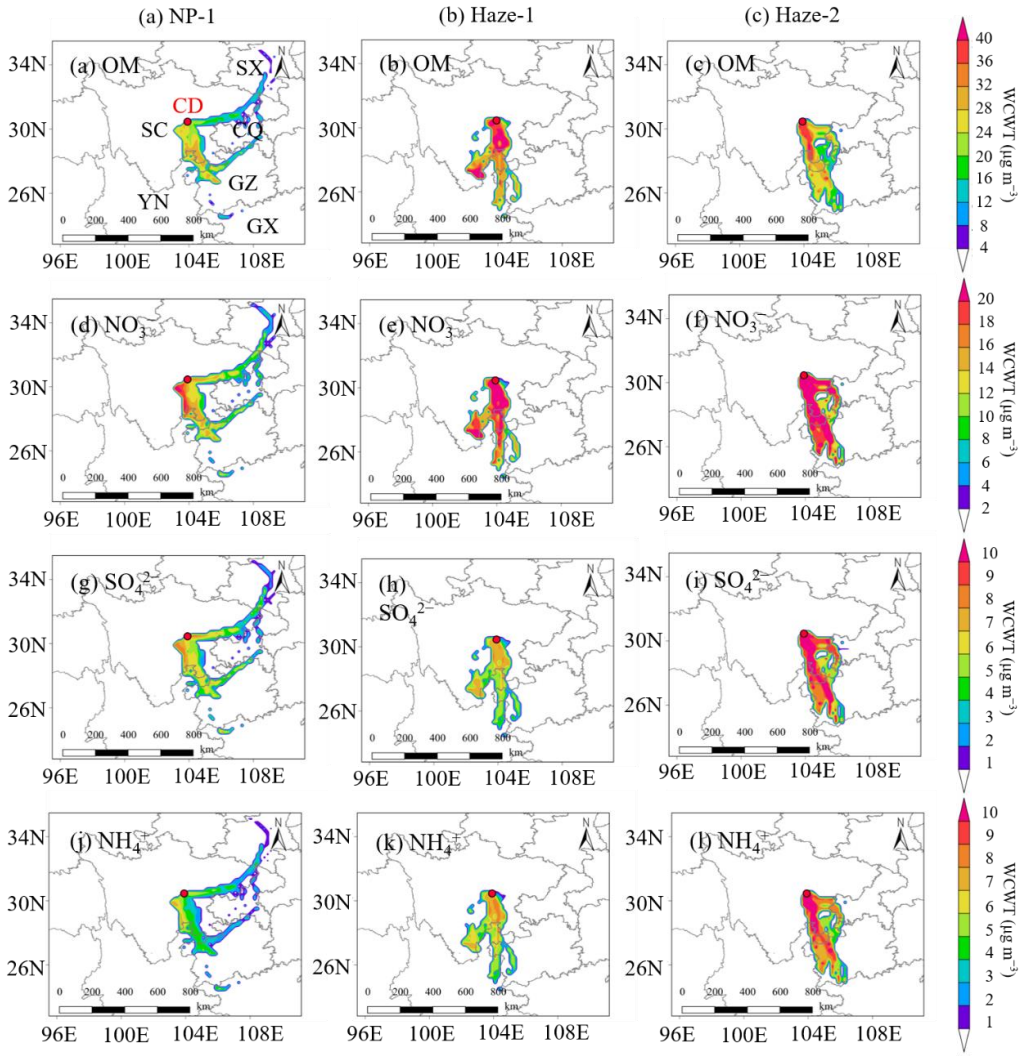

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Figure S6: WCWT maps of different $PM_{2.5}$ chemical components during (a) NP-1, (b) Haze-1 and (c) Haze-2 period (CD: Chengdu; SC: Sichuan Province; SX:Shaanxi Province; CQ:Chongqing; GZ:Guizhou Province; GX:Guangxi Province; YN: Yunnan Province).

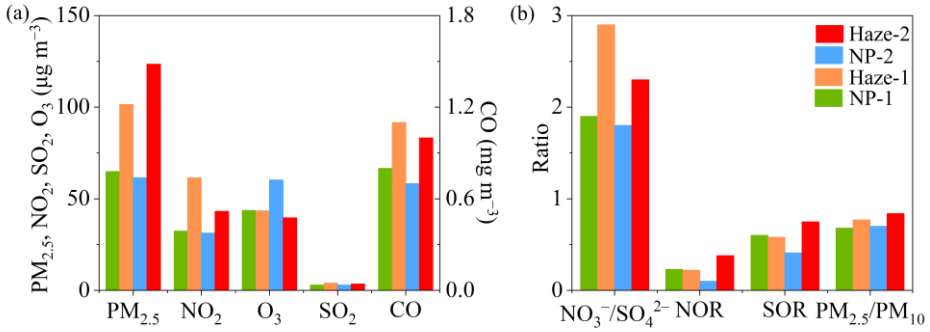

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Figure S7: Pollutant parameters in different periods.

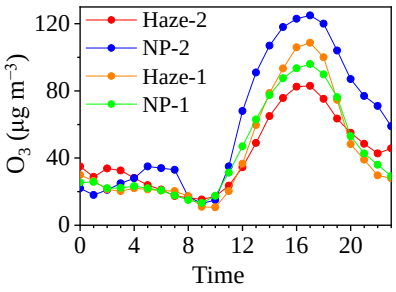

Figure S8: The diurnal variation of O_3 in different periods.

S3 Introduction to PMF factors

The source factors of $PM_{2.5}$ were apportioned by applying the PMF receptor model. The identification of the sources was based on certain chemical tracers that are generally presumed to be emitted by specific sources and are present in significant amounts in the collected samples. At the same time, it is necessary to consider the local pollution characteristics of Chengdu. Ultimately, six factors were identified in this study, and the source characteristics of all these factors are shown in Fig. S9.

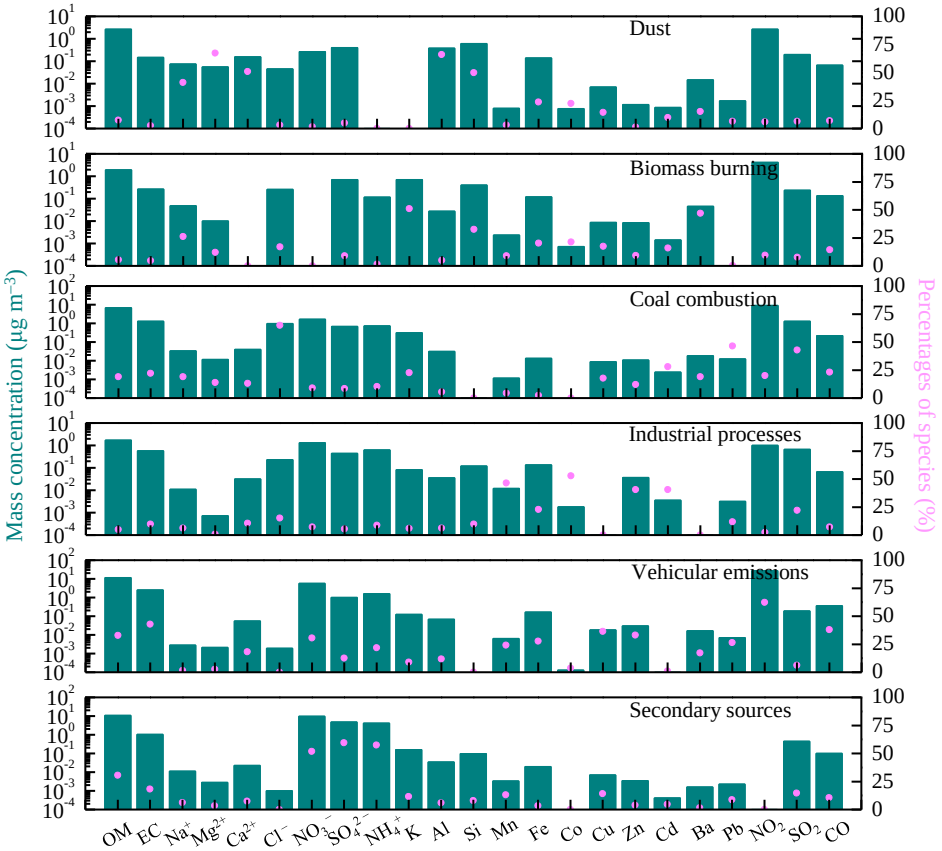

Figure S9: PMF source profiles for $PM_{2.5}$ samples in terms of concentrations and percentages.

Factor 1 was heavily weighted by Na^+, Mg^{2+}, Ca^{2+}, Al and Si, accounting for 41.1%, 67.2%, 50.9%, 65.8% and 49.8%, respectively, which was defined as the dust source (Sun et al., 2022; Zhang et al., 2023). Factor 2 was mainly weighted by K^+ (51.0%) and Na^+ (26.3%), which were identified as indicators of biomass burning (Zhang et al., 2016). Factor 3 was identified by a high loading of Cl^- (64.8%), Pb (46.5%) and SO_2 (42.9%), and a moderately loading of OM, EC, Cd, NO_2 and CO, which were regarded as signals of coal combustion (Zhang et al., 2012; Zhang et al., 2016; Cai et al., 2017). Factor 4 had a high abundance of Mn, Co, Zn and Pb, which are related to industrial processes (Chen et al., 2017; Dall'osto et al., 2008). Factor 5 can be considered as vehicular emissions, being mostly loaded with OM (32.9%), EC (42.9%), NO_2 (62.3%) and some metal elements released during the operation of motor vehicles, such as Mn, Fe, Cu and Zn (de Miranda et al., 2018; Chen et al., 2017). A high loading of NO_3^- (51.9%), SO_4^{2-} (59.6%) and NH_4^+ (57.6%), along with a moderate abundance of OM (30.6%), is apparent in Factor 6, which is typical of the secondary sources profile (Zhang et al., 2023; Huang et al., 2021).

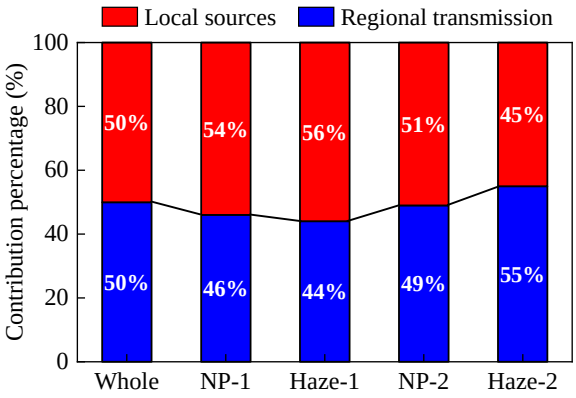

Figure S10: Relative contributions of local sources and regional transmission to $PM_{2.5}$ during different periods.

Table S1. Air quality and meteorological conditions of different cities in China in the same time period and the interannual changes in Chengdu.

	Beijing (2023)	Shanghai (2023)	Guangzhou (2023)	Xi'an (2023)	Chengdu (2015)	Chengdu (2018)	Chengdu (this study)
$PM_{2.5}$ ($\mu g\ m^{-3}$)	32.6±29.2	30.7±13.6	32.0±8.0	106.6±80.0	82.0±41.9	71.7±20.9	95.6±28.7
$PM_{2.5}/PM_{10}$	0.48	0.62	0.61	0.61	0.68	0.69	0.77

NO_2 (μg m^{-3})	27.1±18.7	32.0±18.4	26.2±14.4	54.4±27.0	49.6±20.0	49.7±13.0	43.5±19.2
SO_2 (μg m^{-3})	3.2±2.1	7.4±1.6	5.7±0.7	14.3±6.1	16.1±12.1	8.5±3.3	3.6±1.4
O_3 (μg m^{-3})	41.5±21.0	60.9±22.7	54.7±36.5	39.0±28.5	16.0±16.9	38.5±19.8	44.1±34.5
CO (mg m^{-3})	0.5±0.3	0.7±0.2	0.7±0.2	1.1±0.5	1.5±0.4	1.0±0.2	0.9±0.2
T (°C)	–0.5±5.1	4.8±4.6	14.8±5.3	3.3±4.0	5.9±2.6	3.0±2.5	11.0±3.0
RH (%)	31.4±15.8	64.1±21.7	69.7±24.4	32.7±14.0	80.5±15.0	75.4±20.8	62.4±11.6
WS (m s^{-1})	2.0±1.4	2.2±1.5	2.2±1.6	1.7±1.1	1.7±1.1	1.5±1.0	0.5±0.4

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