Supplement of

Chemical composition, source and formation mechanism of urban $PM_{2.5}$ in Southwest China

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S1 Model configuration

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 January 23 to February 3, for a total of 18 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).

S2 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 Base 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:

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

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

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

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**.

We find that the R-value between the simulated and observed $PM_{2.5}$ concentrations is equal to 0.67 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 is also well reproduced. The simulated and observed NMB and NME values are -18.5% and 28.9%, 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.

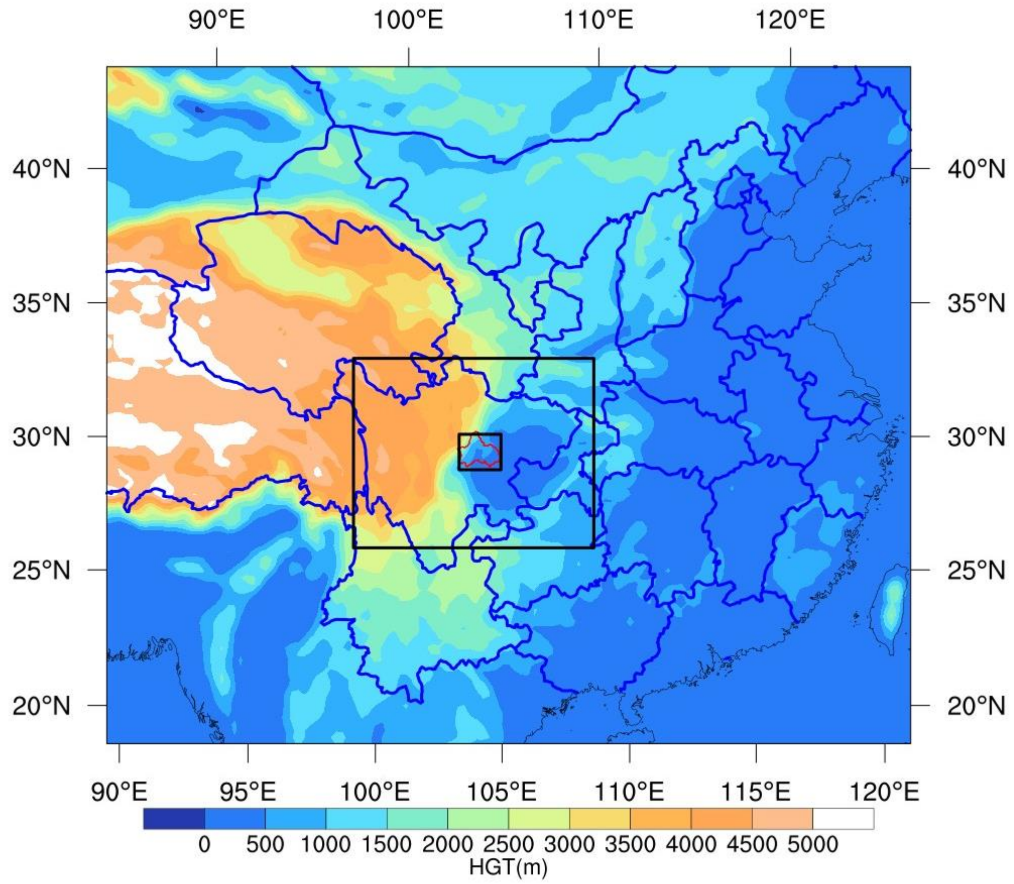

Figure S1. WRF-Chem simulation domain settings.

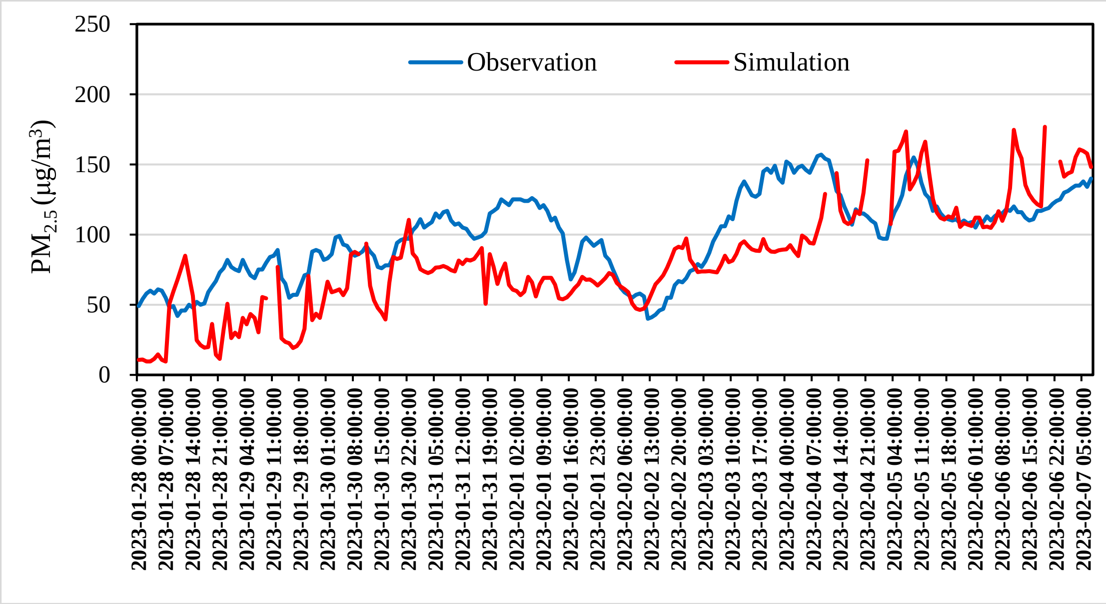

Figure S2. Temporal variation in the simulated and observed surface $PM_{2.5}$ concentration at the Chengdu station.

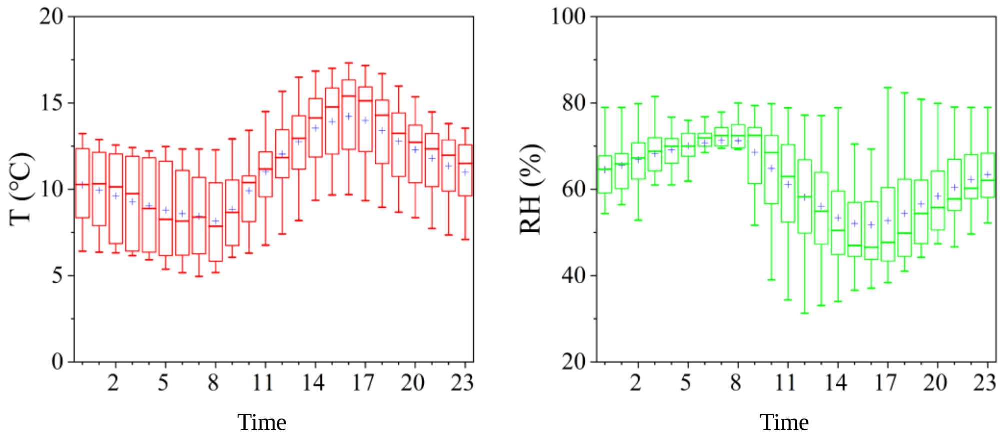

Figure S3. The average diurnal variation of temperature (T) and relative humidity (RH) throughout the whole study period.

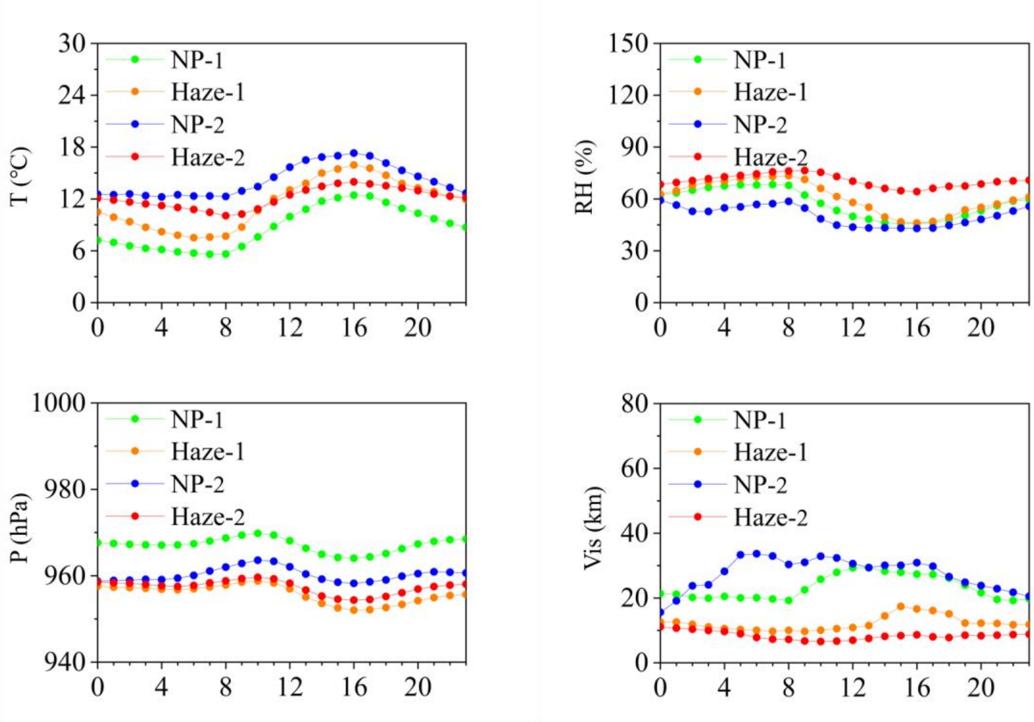

Figure S4. The diurnal variation of four meteorological parameters (T, RH, P and Vis) in four periods.

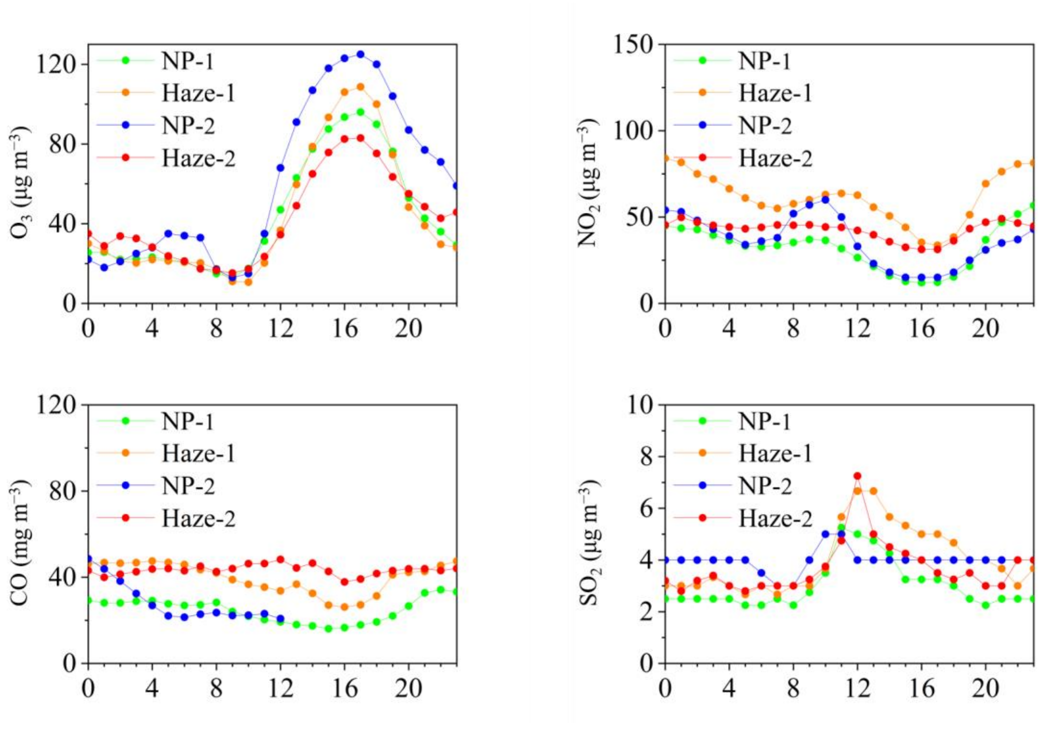

Figure S5. The diurnal variation of four gaseous pollutant (O_3, NO_2, CO and SO_2) in four periods.

References

Chen, F., Kusaka, H., Bornstein, R., Ching, J., Grimmond, C. S. B., Grossman-Clarke, S., Loridan,T., Manning, K. W., Martilli, A., Miao, S., Sailor, D., Salamanca, F. P., Taha, H., Tewari, M., Wang, X., Wyszogrdzki, A. A., and Zhang, C.: The integrated WRF/urban modelling system: development, evaluation, and applications to urban environmental problems, Int. J. Climatol., 31, 273-288, http://pdxscholar.library.pdx.edu/mengin_fac/45, 2011.

Chen, S. H., and Sun, W. Y.: A one-dimensional time dependent cloud model, J. Meteorol. Soc. Jpn., 80, 99-118, https://doi.org/10.1175/JAS-3304.1, 2002.

Dudhia, J.: Numerical study of convection observed during the winter monsoon experiment using a mesoscale two-dimensional model, J. Atmos. Sci., 46, 3077-3107, https://doi.org/10.1175/1520-0469(1989)046<3077:NSOCOD>2.0.CO;2, 1989.

Emmons, L. K., Walters, S., Hess, P. G., Lamarque, J. F., Pfister, G. G., Fillmore, D., Granier, C., Guenther, A., Kinnison, D., Laepple, T., Orlando, J., Tie, X., Tyndall, G., Wieinmyer, C., Baughcum, S. L., and Kloster, S.: Description and evaluation of the model for ozone and related chemical tracers, version 4 (MOZART-4), Geosci. Model Dev., 3, 43-67, https://doi.org/10.5194/gmd-3-43-2010, 2010.

Ginoux, P., Prospero, J. M., Torres, O., and Chin, M.: Long-term simulation of global dust distribution with the GOCART model: correlation with North Atlantic Oscillation, Environ. Modell. Softw., 19, 113-128, https://doi.org/10.1016/S1364-8152(03)00114-2, 2004.

Grell, G. A.: Prognostic evaluation of assumptions used by cumulus parameterizations, Mon. Weather Rev., 121, 764-787, https://doi.org/10.1175/1520-0493(1993)121<0764:PEOAUB>2.0.CO;2, 1993.

Grell, G. A., and Devenyi, D.: A generalized approach to parameterizing convection combining ensemble and data assimilation techniques, Geophys. Res. Lett., 29, 38-1-38-4, https://doi.org/10.1029/2002GL015311, 2002.

Guenther, A., Karl, T., Harley, P., Wiedinmyer, C., Palmer, P. I., and Geron, C.: Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature), Atmos. Chem. Phys., 6,3181-3210, https://doi.org/10.5194/acp-6-3181-2006, 2006.

Hong, S.-Y., Noh, Y., and Dudhia, J.: A new vertical diffusion package with an explicit treatment of entrainment processes, Mon. Weather Rev., 134, 2318-2341, https://doi.org/10.1175/MWR3199.1, 2006.

Huang, L., Zhu, Y., Zhai, H., Xue, S., Zhu, T., Shao, Y., Liu, Z., Emery, C., Yarwood, G., Wang, Y., Fu, J., Zhang, K., and Li, L.: Recommendations on benchmarks for numerical air quality model applications in China - Part 1: $PM_{2.5}$ and chemical species, Atmos. Chem. Phys., 21, 2725-2743, https://doi.org/10.5194/acp-21-2725-2021, 2021.

Jimenez, P. A., Dudhia, J., Gonzalez-Rouco, J. F., Navarro, J., Montavez, J. P., and Garcia-Bustamante, E.: A revised scheme for the WRF surface layer formulation, Mon. Weather Rev., 140, 898-918, https://doi.org/10.1175/MWR-D-11-00056.1, 2012.

Liu, P., Qiu, X., Yang, Y., Ma, Y., and Jin, S.: Assessment of the performance of three dynamical climate downscaling methods using different land surface information over China, Atmosphere, 9, 1-22, https://doi.org/10.3390/atmos9030101, 2018.

Madronich, S.: Photodissociation in the atmosphere.1. Actinic flux and the effects of ground reflections and clouds, J. Geophys. Res.-Atmos., 92, 9740-9752, https://doi.org/9740-975210.1029/JD092iD08p09740, 1987.

Mlawer, E. J., Taubman, S. J., Brown, P. D., Iacono, M. J., and Clough, S. A.: Radiative transfer for inhomogeneous atmospheres: RRTM, a validated correlated-k model for the longwave, J. Geophys. Res.-Atmos., 102, 16663-16682, https://doi.org/10.1029/97JD00237, 1997.

Tewari, M., Chen, F., Wang, W., Dudhia, J., LeMone, M. A., and Mitchell, K., Implementation and verification of the unified NOAH land surface model in the WRF model, 20th conference on weather analysis and forecasting/16th conference on numerical weather prediction. pp. 11–15,2004.

Zaveri, R. A., Easter, R. C., Fast, J. D., and Peters, L. K.: Model for simulating aerosol interactions and chemistry (MOSAIC), J. Geophys. Res.-Atmos., 113, https://doi.org/10.1029/2007JD008782, 2008.