



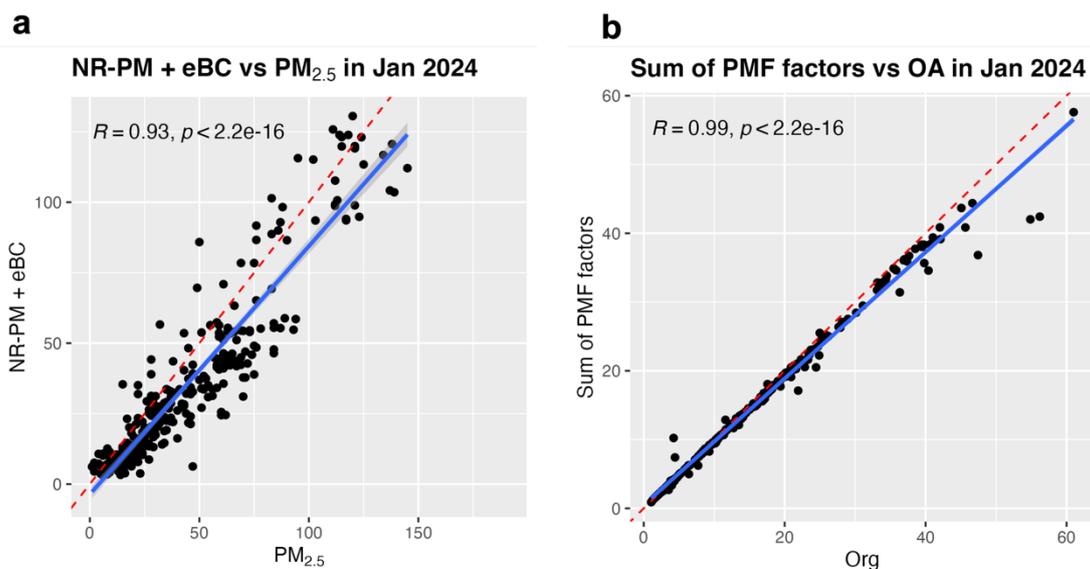
*Supplement of*

## **Growing role of secondary organic aerosol in the North China Plain from 2014 to 2024**

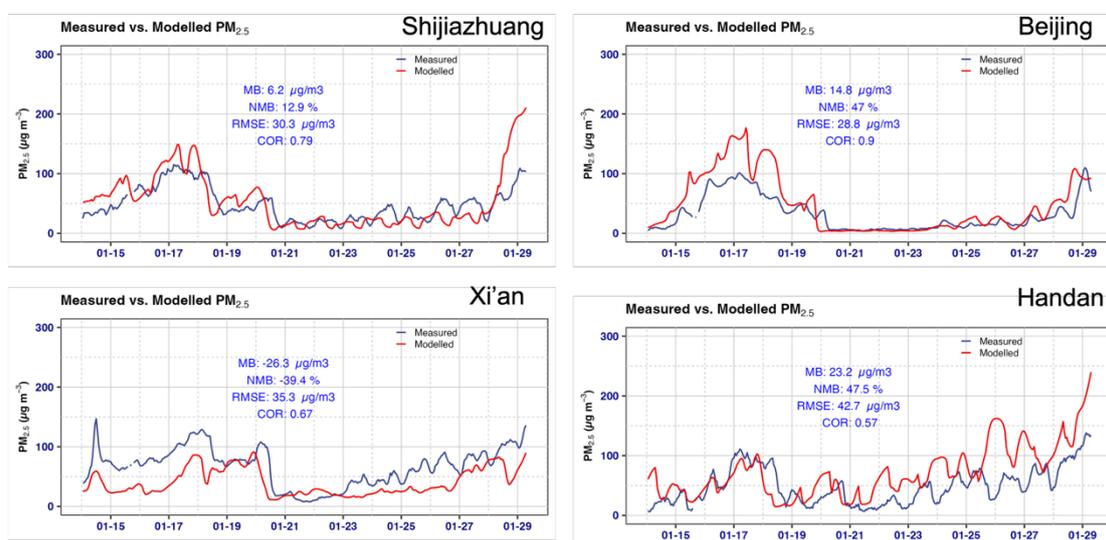
**Chunshui Lin et al.**

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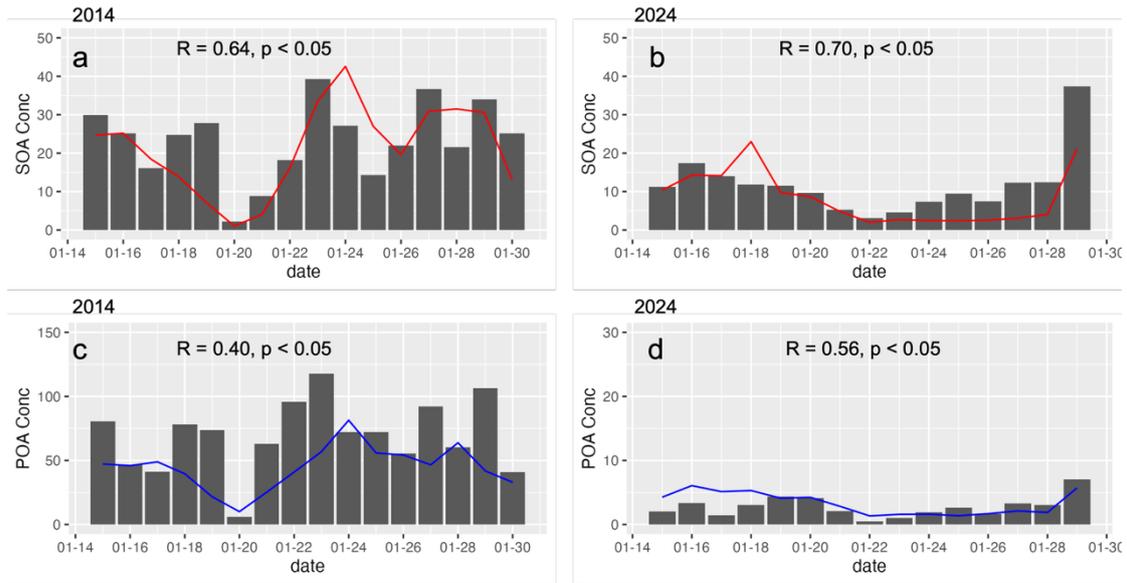
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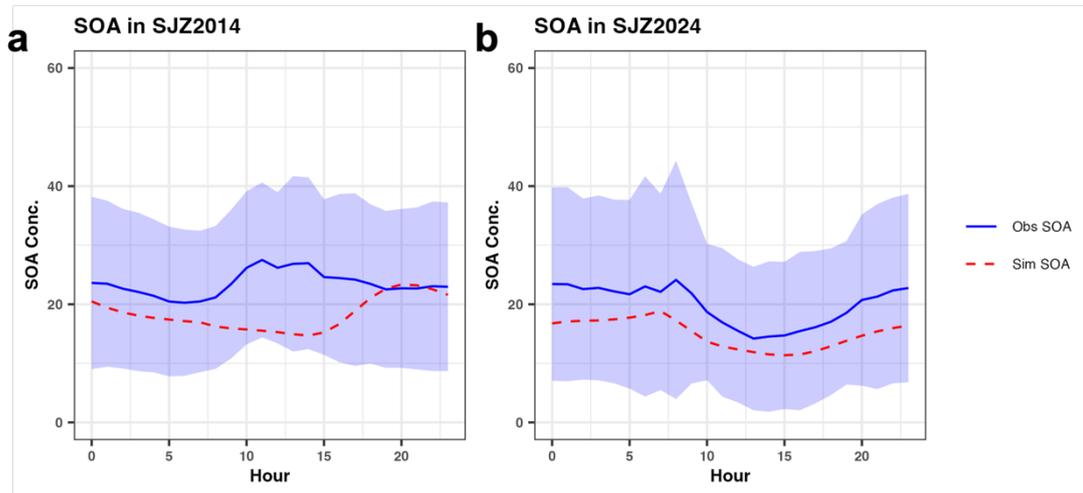
**Figure S1.** (a) scatter plot between the sum of non-refractory particulate matter (NR-PM<sub>2.5</sub>; measured by SP-LToF-AMS) and equivalent black carbon (eBC; measured by AE-33) versus bulk PM<sub>2.5</sub> measured at a nearby air quality monitoring site. (b) Relationship between the sum of organic aerosol (OA) factors and the measured organic concentrations. All values are in  $\mu\text{g m}^{-3}$ . The red dashed line represents the 1:1 ratio, while the blue line indicates the linear fit, with the correlation coefficient and p-value shown in the top left corner.



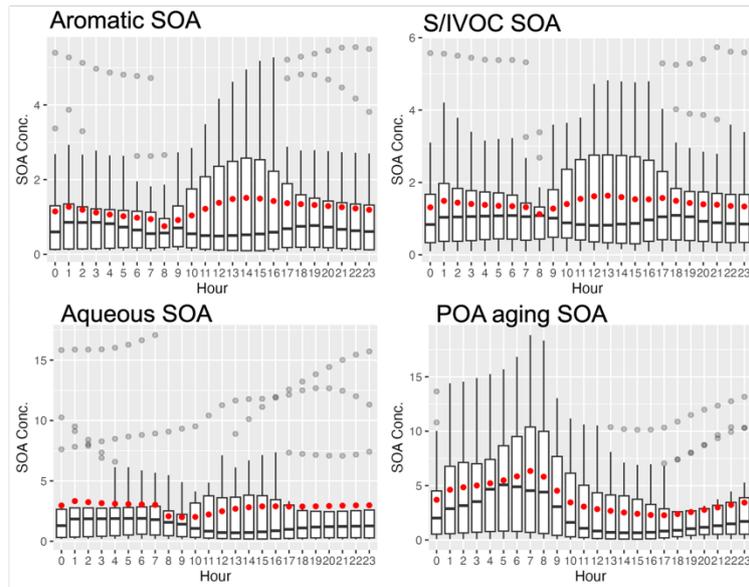
**Figure S2.** Time series of CMAQ-modeled PM<sub>2.5</sub> and measured PM<sub>2.5</sub> in Shijiazhuang, Beijing, Xi'an, and Handan. The Mean Bias (MB), Normalized Mean Bias (NMB), Root Mean Square Error (RMSE), and correlation coefficient (COR) are displayed in blue text.



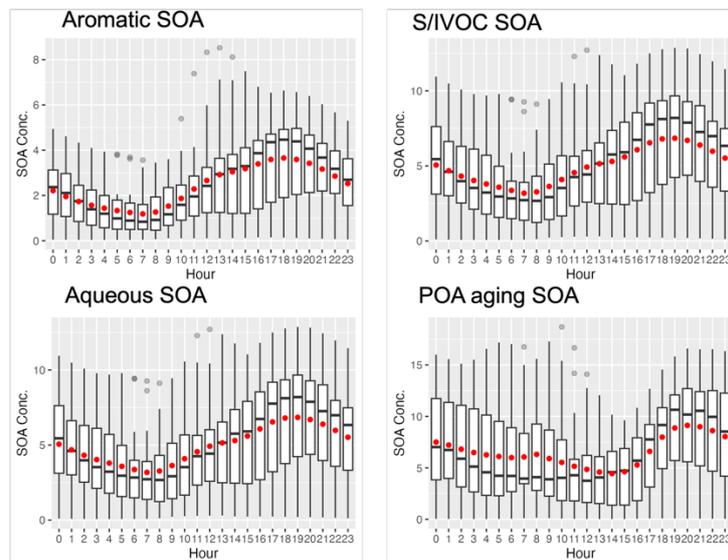
**Figure S3.** Comparison of simulated SOA and POA with observed values for 2014 and 2024. Bars represent measured daily values, while lines represent the corresponding modeled values.



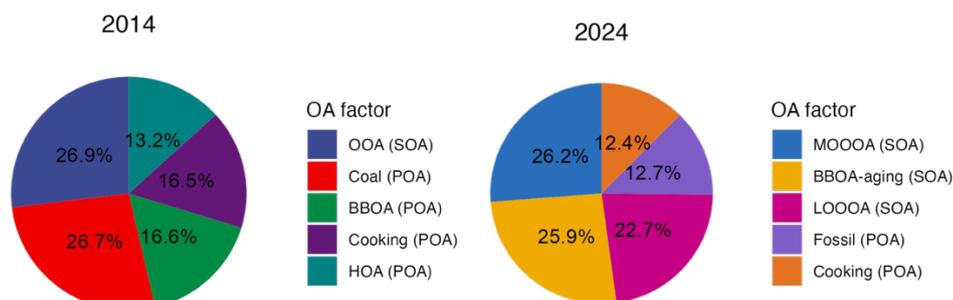
**Figure S4.** Diurnals of observed and simulated SOA using the adjusted model.



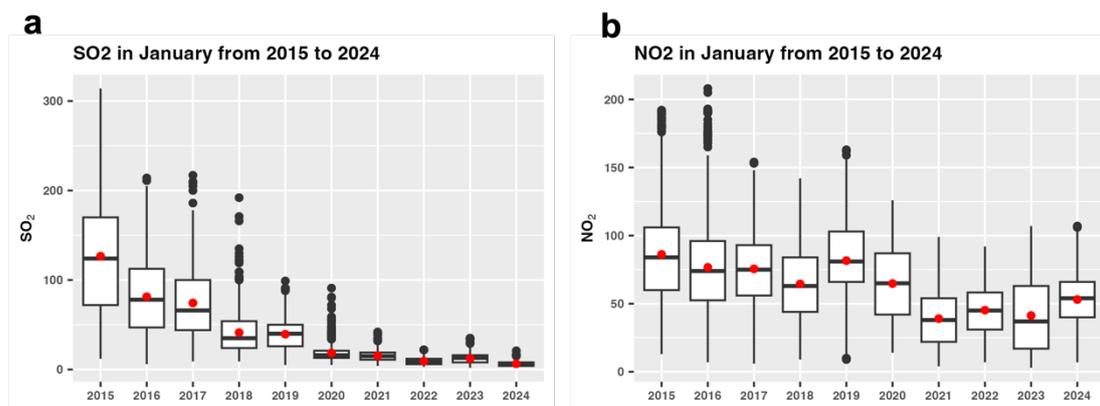
**Figure S5.** Diurnal variations in distinct SOA formation pathways in 2024, including SOA derived from aromatic oxidation, SVOCs/IVOCs oxidation, the aqueous pathway, and POA aging. Box plots display the median, 25th, and 75th percentiles; whiskers extend to 1.5 times the interquartile range (IQR). Grey dots represent outliers, and red dots indicate mean values.



**Figure S6.** Same as Figure S5, but for 2014.

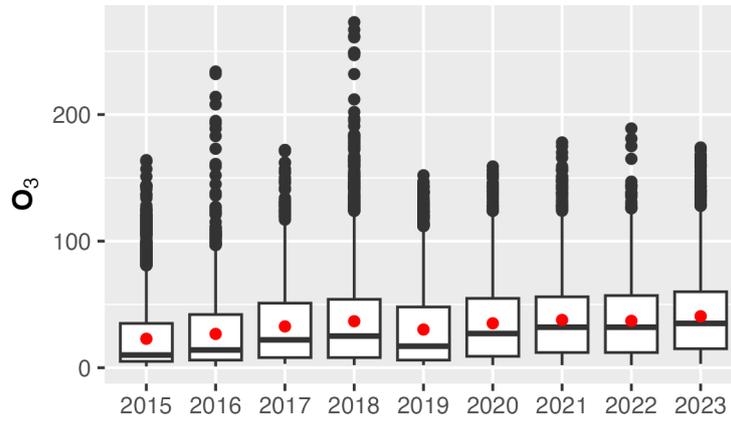


**Figure S7.** OA factor contributions in Shijiazhuang (2014 vs 2024). Non-cooking POA (ncPOA) represents the sum of all other POA fractions. SOA surrogates were resolved only as bulk OOA for 2014, whereas three distinct SOA types were identified for 2024, attributed to the use of high-mass-resolution AMS and PMF analysis.



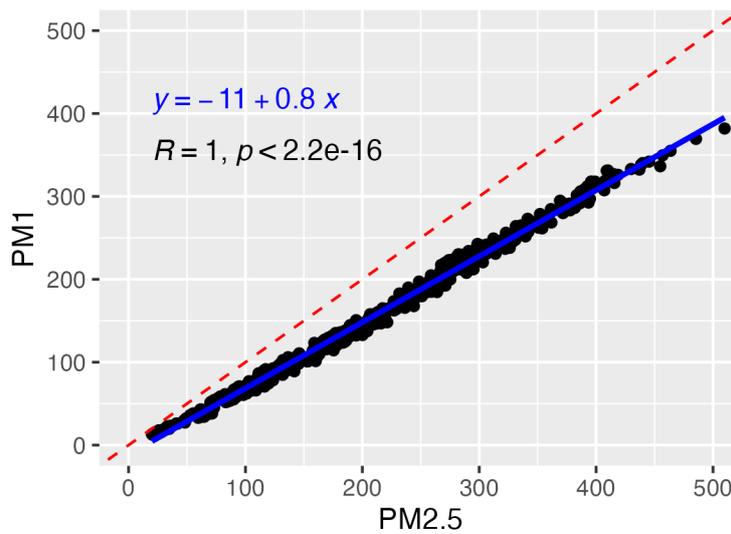
**Figure S8.** Box plot of SO<sub>2</sub> (a) and NO<sub>2</sub> (b) concentrations in January from 2015 to 2024. The box plot displays the 50th percentile (median), 25th percentile, 75th percentile, and the values extending to 1.5 times the interquartile range. The red dot represents the mean concentration, while the black dots outside the box indicate the outliers.

### Ozone in winter Shijiazhuang

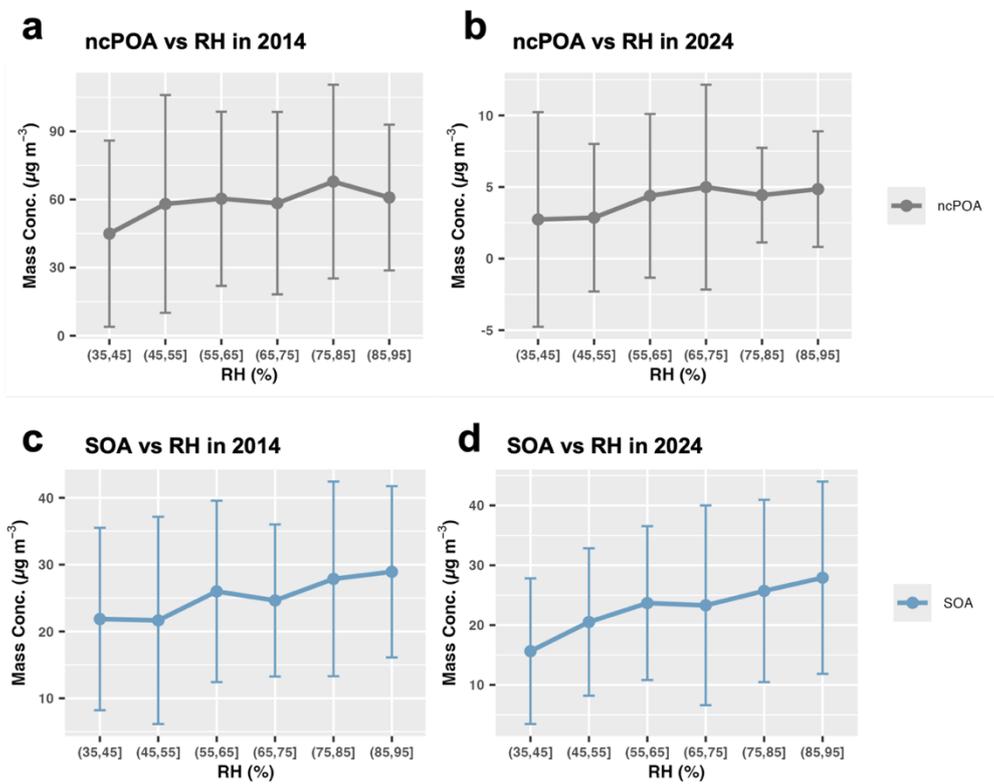


**Figure S9.** Box plot of O<sub>3</sub> concentrations in winter from 2015 to 2024. The box plot displays the 50th percentile (median), 25th percentile, 75th percentile, and the values extending to 1.5 times the interquartile range. The red dot represents the mean concentration, while the black dots outside the box indicate the outliers.

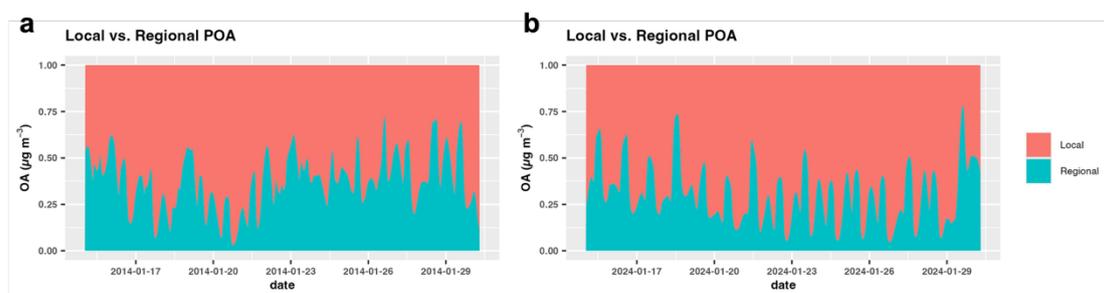
### PM<sub>1</sub> vs PM<sub>2.5</sub>



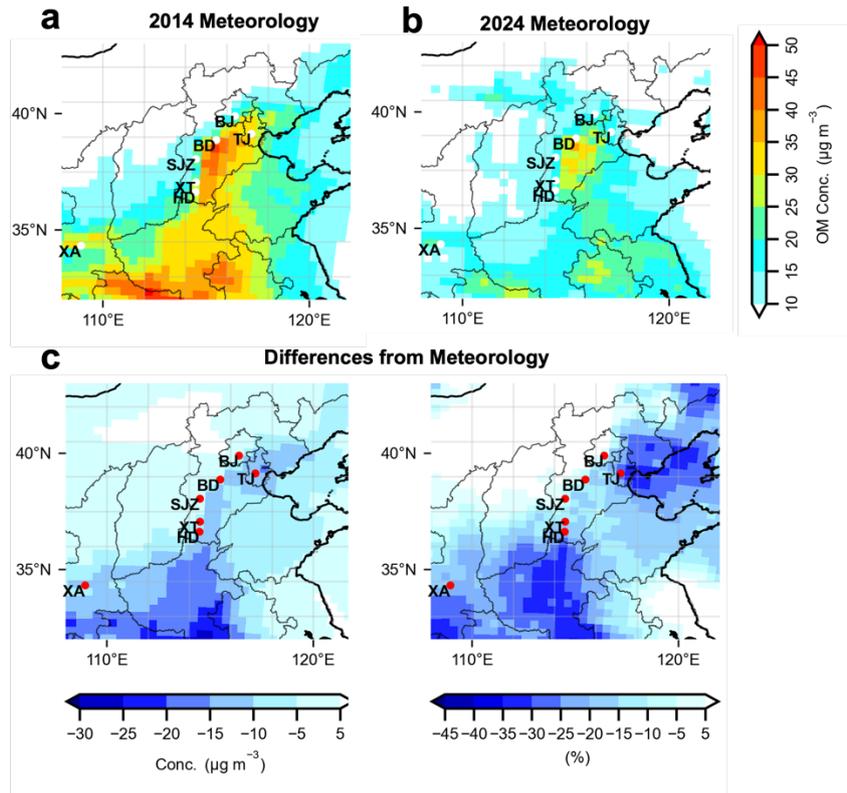
**Figure S10.** Scatter plot of PM<sub>1</sub> (AMS size cut) versus PM<sub>2.5</sub> from CMAQ. The red dashed line represents the 1:1 ratio line, while the blue line denotes the linear fit. Text annotations indicate the corresponding linear fit equation and correlation coefficient.



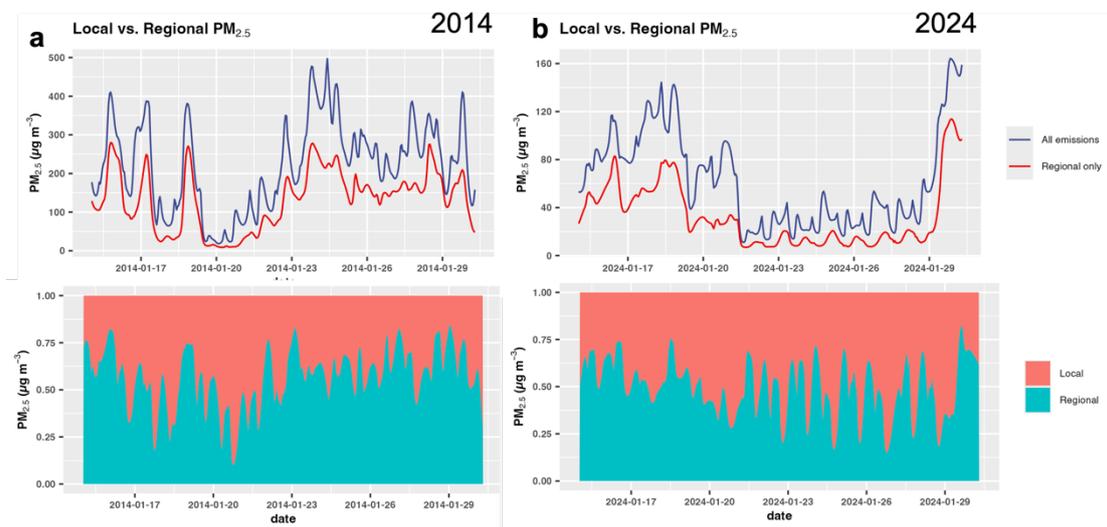
**Figure S11.** Organic Aerosol Subtypes vs RH. Variation of non-cooking primary organic aerosol (ncPOA) and secondary organic aerosol (SOA) across different relative humidity (RH) brackets in 2014 (a) and (b) and in 2024 (c) and (d). The error bars represent one standard deviation.



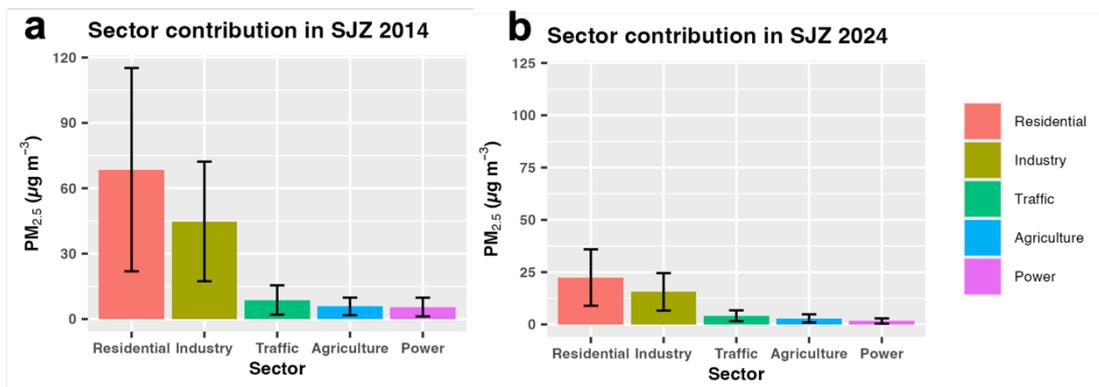
**Figure S12.** Time series of the fraction between local and regional contributions for primary organic aerosol (POA).



**Figure S13.** Differences in CMAQ simulations attributed to meteorological variations, comparing 2024 with 2014 while using the same emission inventory.



**Figure S14.** Time series of  $PM_{2.5}$  in January 2014, comparing scenarios with all emissions considered (i.e., All Emissions) and with emissions from Shijiazhuang zeroed out (i.e., Regional Only). The difference between "All Emissions" and "Regional Only" represents local emissions. The lower panel displays the time series of the fraction between local and regional contributions.



**Figure S15.** The contribution of five major sectors in MEIC, i.e. residential combustion, industry, traffic power generation, agriculture to the total PM<sub>2.5</sub> in Shijiazhuang (SJZ) in (a) 2014 and (b) 2024.

**Table S1.** Model set-up in different model runs.

Scenario	Run # (Figures)
Without Considering RH-mediated POA conversion	Base (Fig. 3)
Considering RH-mediated POA conversion	Adjust (Fig. 3)
MEIC 2024 + Meteorology 2024	Emission diff (Fig. 5)
MEIC 2014 + Meteorology 2024	Emission diff (Fig. 5)
MEIC 2024 + Meteorology 2014	Meteorological diff (Fig. S13)
MEIC 2024 (zero SJZ) + Meteorology 2024	Regional vs local (Fig. S12, S14)
MEIC 2014 (zero SJZ) + Meteorology 2014	Regional vs local (Fig. S12, S14)

**Table S2.** Mean simulated SOA concentration from the pathways of aqueous-phase (aq), semi/intermediate volatility organic compounds (S/I), aromatics (Arom), and POA aging (in  $\mu\text{g m}^{-3}$ ) across different cities. The sum of these pathways equals total simulated SOA.

	2014 (aq + S/I + Arom + Aging = SOA)	2024 (aq + S/I + Arom + Aging = SOA)
Shijiazhuang (SJZ)	$7.1 + 4.9 + 2.4 + 6.5 = 20.9$	$2.8 + 2.3 + 1.2 + 5.8 = 12.1$
Beijing (BJ)	$6.5 + 3.7 + 2.6 + 4.7 = 17.5$	$3.5 + 2.2 + 1.8 + 3.2 = 10.7$
Tianjin (TJ)	$11.1 + 5.9 + 4.1 + 9.3 = 30.4$	$2.3 + 1.9 + 1.3 + 4.8 = 10.3$
Baoding (BD)	$6.2 + 4.7 + 2.1 + 7.1 = 20.1$	$2.9 + 2.4 + 1.3 + 5.0 = 11.6$
Xingtai (XT)	$7.5 + 4.5 + 2.3 + 5.4 = 19.7$	$2.6 + 2.3 + 1.1 + 5.8 = 11.8$
Handan (HD)	$11.7 + 6.5 + 3.4 + 10.6 = 32.2$	$3.4 + 3.1 + 1.6 + 8.6 = 16.7$
Xi'an (XA)	$5.2 + 3.9 + 2.0 + 4.9 = 16.0$	$2.1 + 1.9 + 1.0 + 4.4 = 9.4$