Supplementary for

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

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This document includes 10 supplementary figures (Fig. S1–S10) and 1 table (Table S1).
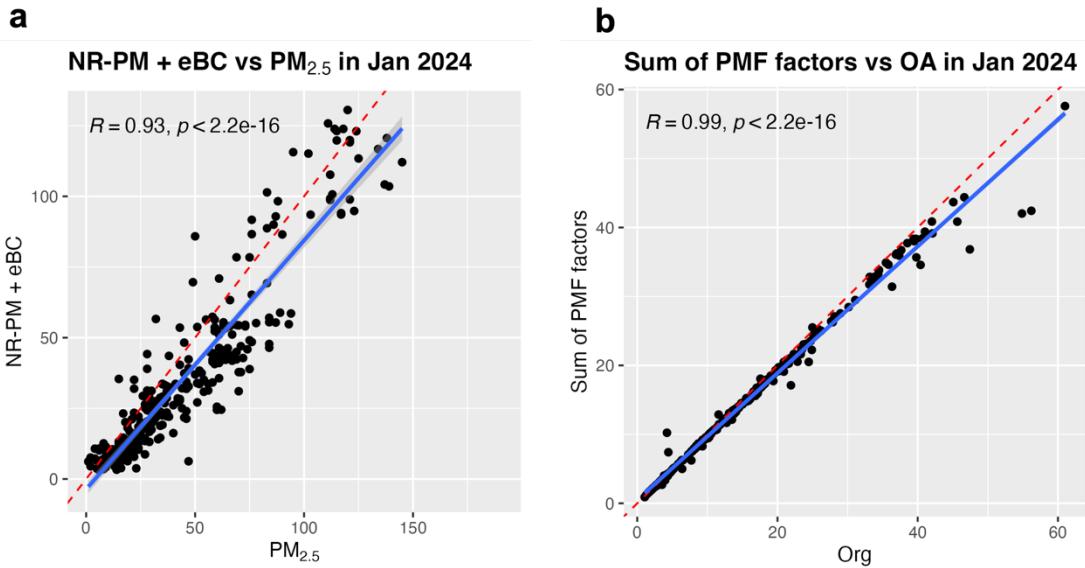
Figure S1. (a) scatter plot between the sum of non-refractory particulate matter (NR-PM$_{2.5}$; measured by SP-LToF-AMS) and equivalent black carbon (eBC; measured by AE-33) versus bulk PM$_{2.5}$ measured at a nearby air quality monitoring site. (b) Relationship between the sum of organic aerosol (OA) factors and the measured organic concentrations. All values are in μg 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.
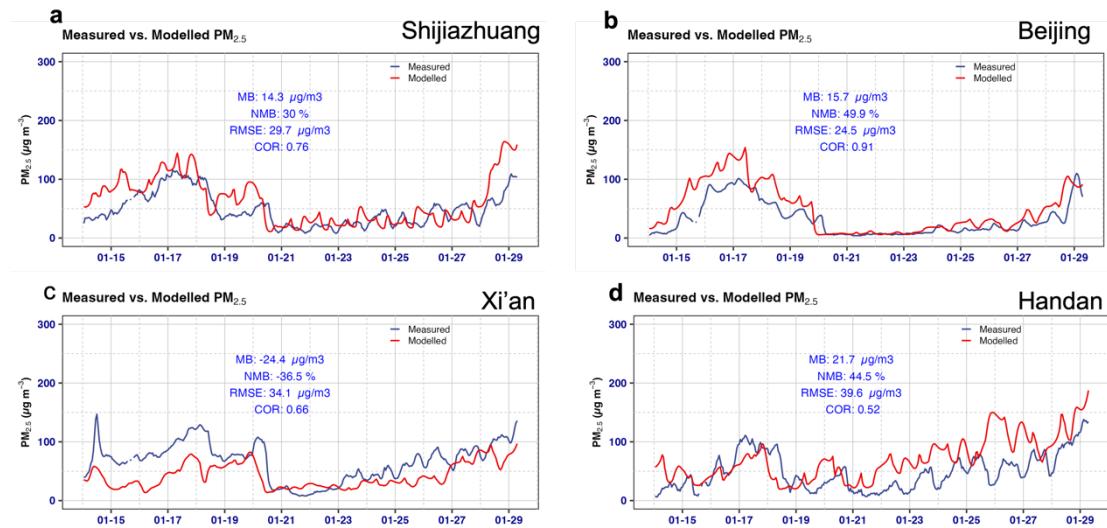
Figure S2. Time series of CMAQ-modeled PM$_{2.5}$ and measured PM$_{2.5}$ in Shijiazhuang (a), Beijing (b), Xi'an (c), and Handan (d). The Mean Bias (MB), Normalized Mean Bias (NMB), Root Mean Square Error (RMSE), and correlation coefficient (r) are displayed in blue text.
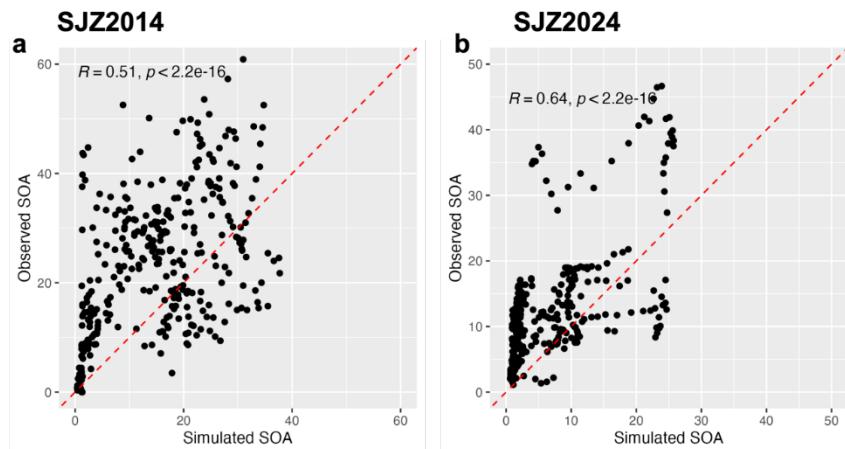
Figure S3. comparison of the simulated SOA and observed SOA.
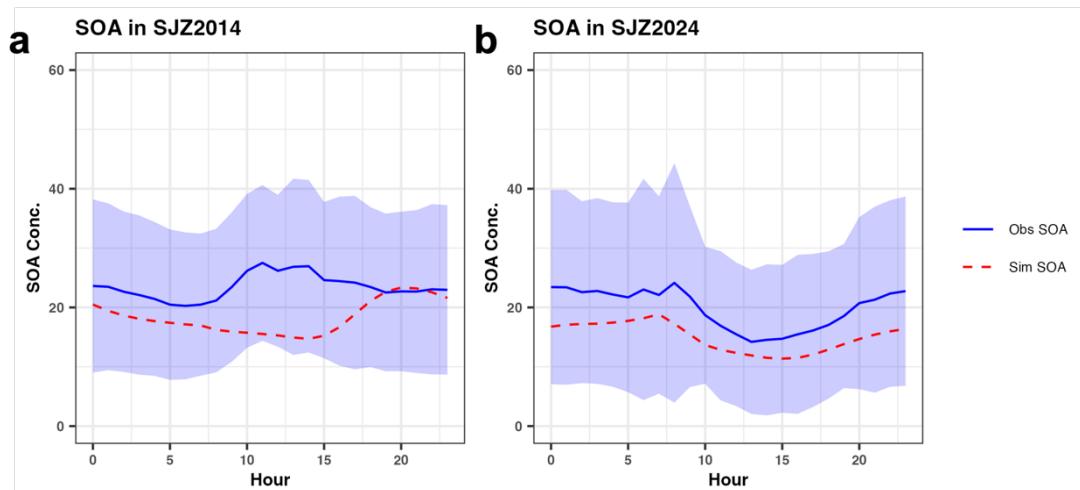
Figure S4. Diurnals of observed and simulated SOA using the adjusted model.
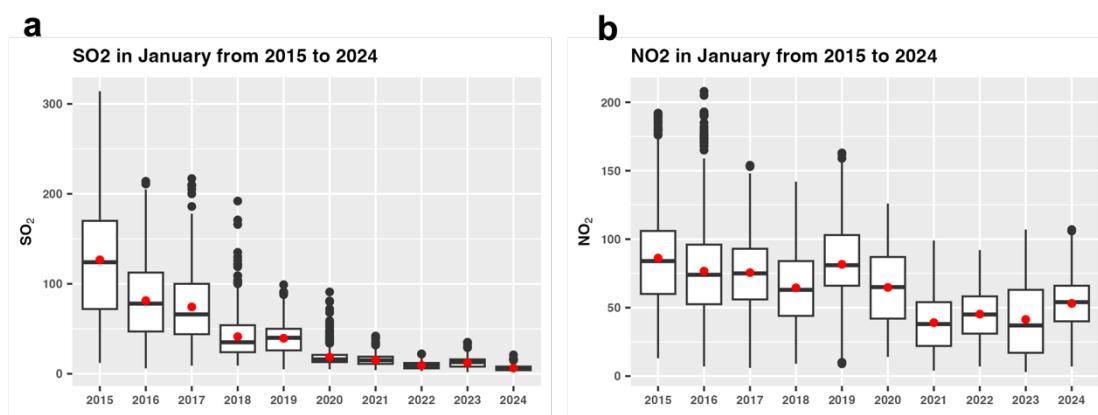
Figure S5. Box plot of SO_2 (a) and NO_2 (b) concentrations in January from 2015 to 2024. The box plot displays the 50th percentile (median), 25th percentile, 75th percentile, and the values extending to 1.5 times the interquartile range. The red dot represents the mean concentration, while the black dots outside the box indicate the outliers.
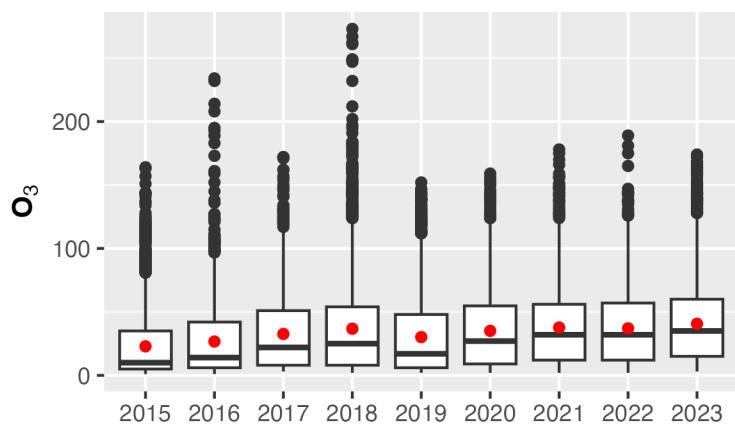
Figure S6. Box plot of O₃ concentrations in winter from 2015 to 2024. The box plot displays the 50th percentile (median), 25th percentile, 75th percentile, and the values extending to 1.5 times the interquartile range. The red dot represents the mean concentration, while the black dots outside the box indicate the outliers.
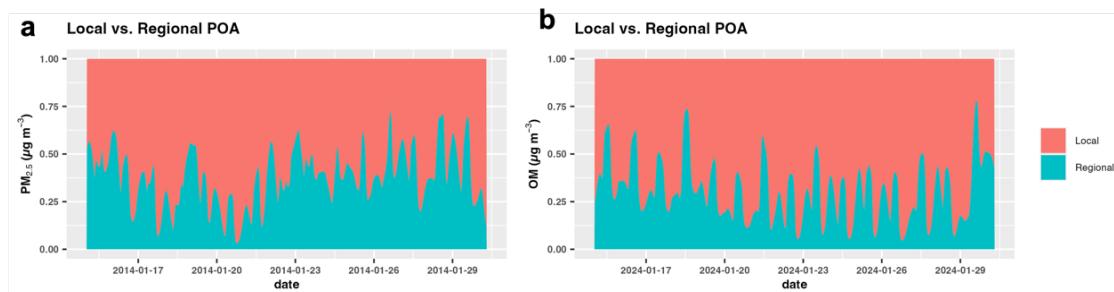
Figure S7. Time series of the fraction between local and regional contributions for primary organic aerosol (POA).
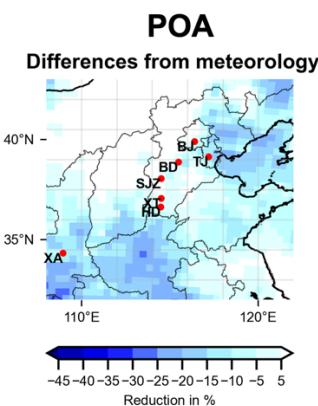
Figure S8. Differences in CMAQ simulations attributed to meteorological variations, comparing 2024 with 2014 while using the same emission inventory.
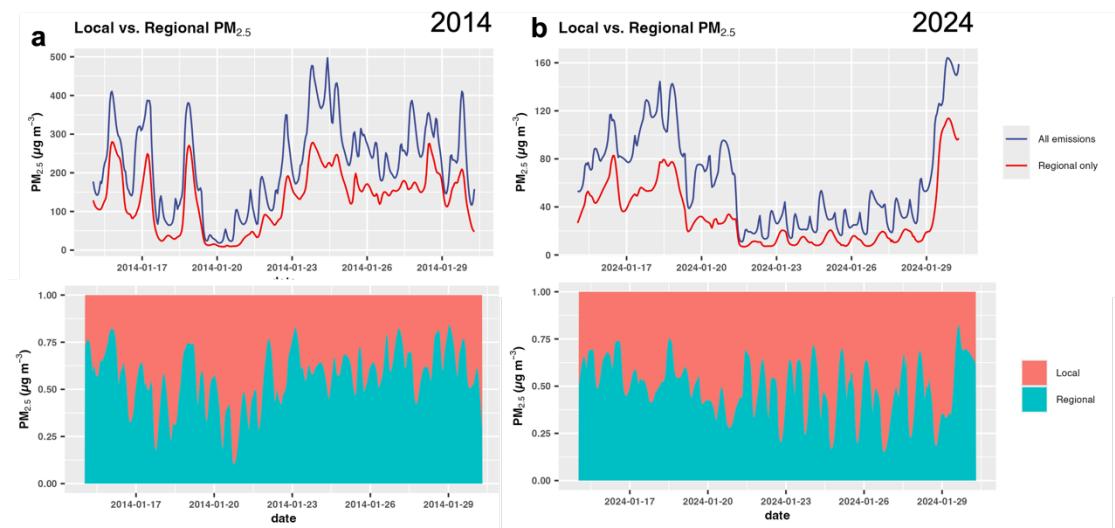
Figure S9. Time series of PM$_{2.5}$ in January 2014, comparing scenarios with all emissions considered (i.e., All Emissions) and with emissions from Shijiazhuang zeroed out (i.e., Regional Only). The difference between "All Emissions" and "Regional Only" represents local emissions. The lower panel displays the time series of the fraction between local and regional contributions.
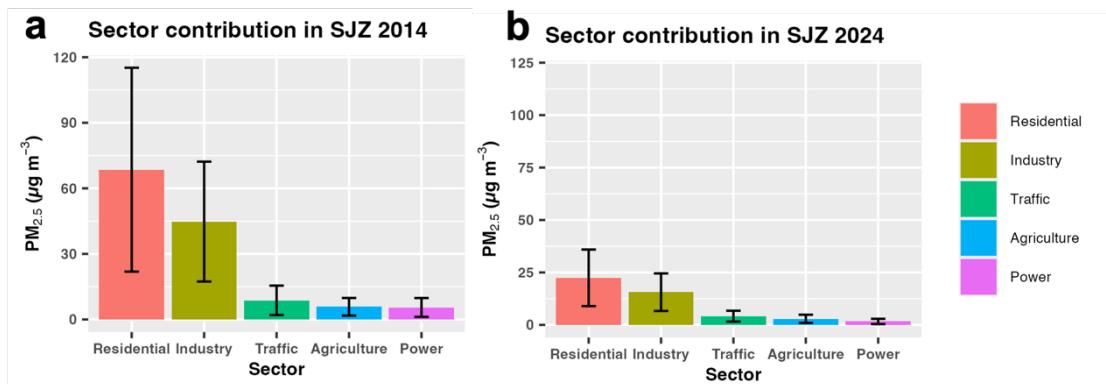
Figure S10. The contribution of five major sectors in MEIC, i.e. residential combustion, industry, traffic power generation, agriculture to the total PM2.5 in Shijiazhuang (SJZ) in (a) 2014 and (b) 2024.

Table S1. Model set-up in different model runs

Scenario	Run #
Without Considering RH-mediated POA conversion	Base
Considering RH-mediated POA conversion	Adjust
MEIC 2024 + Meteorology 2024	Emission diff
MEIC 2014 + Meteorology 2024	Emissions diff
MEIC 2024 + Meteorology 2014	Meteorological diff
MEIC 2024 (zero SJZ) + Meteorology 2024	Regional vs local
MEIC 2014 (zero SJZ) + Meteorology 2014	Regional vs local