We are grateful to the reviewer for taking the time to re-review our manuscript. We have revised the manuscript accordingly, which has helped improve the quality and clarity of this study substantially. Below is our point-by-point response to each comment. The reviewer's comments are in black and our replies in cyan.

Lin et al. did a good job in addressing some of my comments and the concerns raised by the reviewers. The manuscript has been improved, but I still have some comments and concerns on the replies and revised manuscript. It is recommended to publish after the following questions have been well addressed.

Response: We thank the reviewer for his/her careful re-review of our manuscript. Below is a point-by-point response to the comments.

For the response to comment 1 of reviewer 1: It is surprising that the author just gives a scaling factor of 0.5 for 2020 MEIC data to obtain MIEC 2024 to enable more accurate result. It means that the emission intensity was cutoff a half in 2024 compared to 2020? It sounds not reasonable and unrealistic. As shown in Figure S2, the modeled and observed data did not match well. These indicated that random scaling factor was selected. The author should give more reasonable, available and convinced analysis and discussion.

Response: We agree that scaling the 2020 MEIC data may introduce substantial uncertainties and cause confusion. To avoid unrealistic scaling of the emission inventory from 2020 for 2024, we have rerun the model and now adopted the latest 2023 MEIC data (no scaling applied) as the base emission inventory for 2024. As shown in the updated Figure S2, CMAQ results with the 2023 MEIC can simulate ambient $PM_{2.5}$ with reasonable performance. Figure S2 and other relevant figures have been updated accordingly.

In line 113, it now reads, "... The emission inventory for 2014 was derived from the Multiresolution Emission Inventory for China (MEIC) (http://meicmodel.org.cn; last access: May 1, 2025) (Geng et al., 2024), while the 2023 emission inventory was used as the base emission data for 2024 (Fig. S2)..."

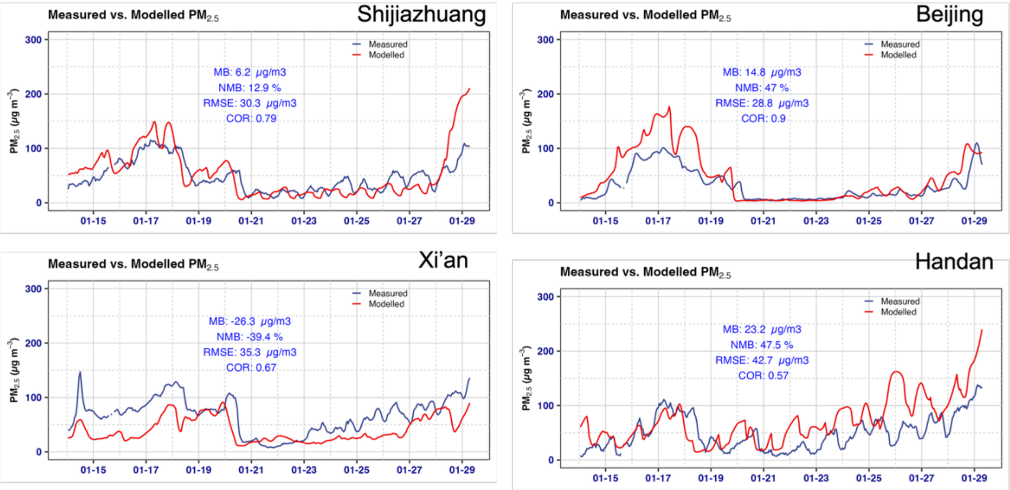

Figure S2. Time series of CMAQ-modeled $PM_{2.5}$ and measured $PM_{2.5}$ in Shijiazhuang, Beijing, Xi'an, and Handan. The Mean Bias (MB), Normalized Mean Bias (NMB), Root Mean Square Error (RMSE), and correlation coefficient (COR) are displayed in blue text.

For the response to comment 2 of reviewer 1: The author just mentioned it existed limitation and uncertainty when directly comparing PM1 and PM2.5. What is the detailed uncertainty of SOA concentration or percentages in PM1 and PM2.5? How is the growth role of this uncertainty? It should give specific discussion.

Response: We have now discussed the PM_1 fraction in $PM_{2.5}$ by comparing PM_1 (within the AMS PM_1 size range) with PM $PM_{2.5}$ from CMAQ simulation. The results show that PM_1 accounted for an average of 80% of $PM_{2.5}$. Directly comparing 2014 PM_1 ACSM data with 2024 $PM_{2.5}$ AMS data may lead to the underestimation of PM species and OA factor reduction over this decade.

Line 207, it now reads, "... PM_1 mass typically accounts for 50–80% of $PM_{2.5}$ mass concentration in ambient air (Elser et al., 2016), and simulations from the CMAQ model indicate that PM_1 (within the AMS/ACSM PM_1 size range) constitutes approximately 80% of total $PM_{2.5}$ mass (Fig. S10). Therefore, comparing 2014 PM_1 data (which correlates to a higher $PM_{2.5}$ level) directly with 2024 $PM_{2.5}$ data would underestimate the actual reduction in $PM_{2.5}$ concentrations between the two periods. However, given the strong time-series correlation in bulk $PM_{2.5}$ concentrations across cities in the NCP (as discussed in Section 3.4), this comparison can still reflect an overall trend..."

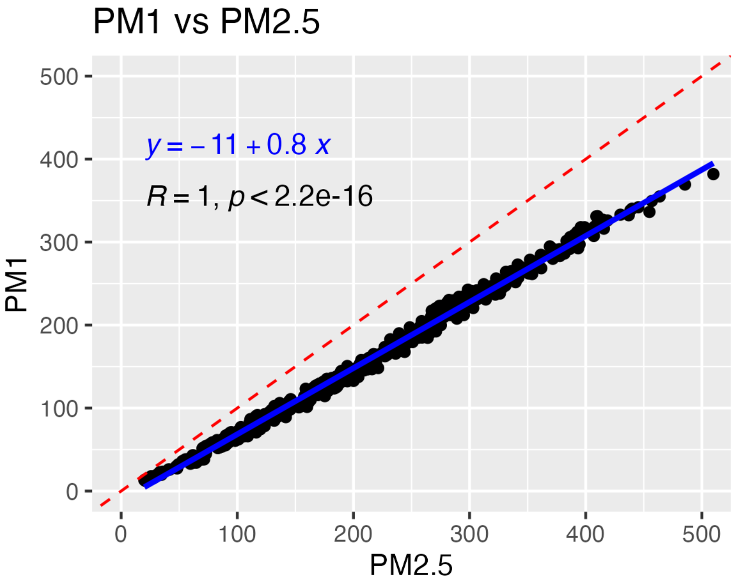

Figure S10. Scatter plot of PM_1 (AMS size cut) versus $PM_{2.5}$ from CMAQ. The red dashed line represents the 1:1 ratio line, while the blue line denotes the linear fit. Text annotations indicate the corresponding linear fit equation and correlation coefficient.

For the response to comment 3 of reviewer 1: The author did not address this question. In Figures 1a and b, the composition of PM2.5 has changed greatly in 2024 compared to 2014. In particular, the fraction of nitrate exceeded that of sulfate. This means aerosol acidity and aerosol liquid water content (ALWC) might have greatly changed at different pollution levels, which should influence aerosol formation, including both inorganic and organic aerosols. Those factors should be accounted for when you discuss SOA formation. However, the author just mentioned that it may alter the formation of SOA. What is the specific influence? How to affect the SOA formation.

Response: We agree that ALWC is important in driving SOA formation. We have now added discussion on the impact of ALWC on SOA formation. ALWC plays an important role in SOA formation given that elevated ALWC enhances the dissolution of gaseous precursors (e.g., VOCs and OVOCs) into the aerosol liquid phase, where they undergo acid-catalyzed oxidation, oligomerization, and other heterogeneous reactions. The observed decrease in ncPOA with rising ALWC implies ncPOA can undergo further oxidation and chemical modification, reducing their volatility and transforming them into SOA species.

We have added descriptions of ALWC calculation using the ISORROPIA II thermodynamic model (Fountoukis and Nenes, 2007), and updated Figure 2 to present the relationship between SOA and ALWC. The relevant discussion section has also been revised accordingly.

Line 146, it now reads, "... ALWC was calculated using the ISORROPIA II thermodynamic model in forward mode (Fountoukis and Nenes, 2007), with input parameters including the inorganic aerosol components (sulfate, nitrate, ammonium and chloride) quantified by AMS or ACSM, as well as RH and temperature from the field campaign. The forward mode of ISORROPIA II enables the thermodynamically consistent prediction of aerosol liquid water content by simulating the equilibrium partitioning of inorganic species between the gas and condensed phases, leveraging the constraint of measured inorganic composition and ambient RH/temperature to yield ALWC estimates..."

Line 239, it now reads, "... Figure 2 shows that as ALWC increased, OOA in both 2014 and 2024 exhibited an increasing trend. In 2024, when ALWC rose from 5 μg m^{-3} to over 100 μg m^{-3}, OOA increased from 18.4 μg m^{-3} to 35.7 μg m^{-3}. In 2014, OOA increased from 12.6 to 28.9 μg m^{-3}. In contrast to OOA, ncPOA showed a plateau or decreasing trend at higher ALWC. ALWC plays an important role in SOA formation: elevated ALWC enhances the dissolution of gaseous precursors (e.g., VOCs and OVOCs) into the aerosol liquid phase, where they undergo acid-catalyzed oxidation, oligomerization, and other heterogeneous reactions. The observed decrease in ncPOA with rising ALWC implies ncPOA can undergo further oxidation and chemical modification, reducing their volatility and transforming them into SOA species..."

For the response to comment 4 of reviewer 1: I agree with the author that POA was not necessarily internally mixed with the inorganic components that play key roles in regulating ALWC, but the SOA formation would be affected according to aqueous chemistry. It would be reasonable replacing RH with ALWC.

Response: We agree that replacing RH with ALWC can better capture the combined impacts of RH and aerosol composition on SOA formation. Figure 2 has been updated to illustrate the relationship between SOA and ALWC, while the original Figure 2 has been relocated to the supporting information (Figure S10).

We have also revised the discussion section related to ALWC. Specific edits to the main text are provided in our response to the preceding comment.

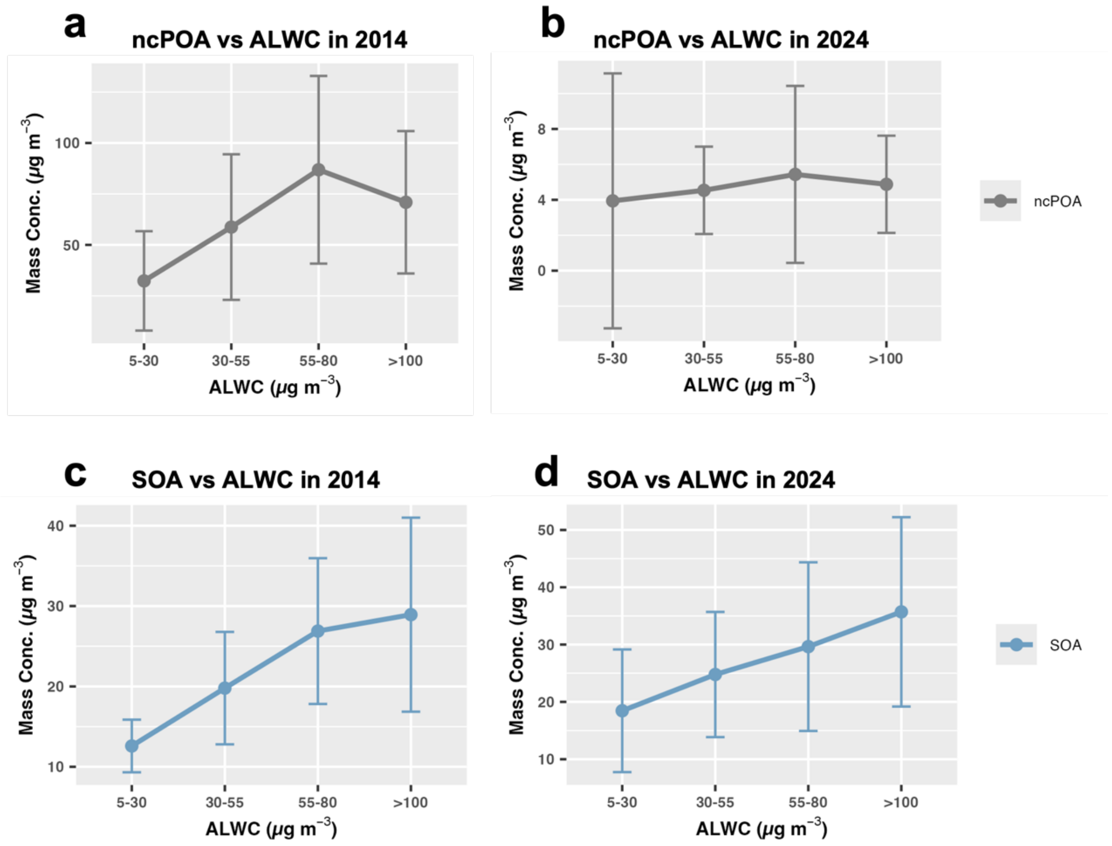

Figure 2. Organic Aerosol Subtypes vs ALWC. Variation of non-cooking primary organic aerosol (ncPOA) and secondary organic aerosol (SOA) across different ALWC ranges in 2014 (a) and (b) and in 2024 (c) and (d). The error bars represent one standard deviation. Only RH from 35% to 95% was considered.

For the response to comment 12 of reviewer 1: I think the author did not address this question well. The author just mentioned that the daytime SOA formation was more important in 2014 than in 2024. First, this conclusion was not reasonable due to the terrible model performance. Second, the author should present the contribution of different process in CMAQ model in 2014 and in 2024 to check the model performance. Meanwhile, any process contributing to SOA formation lack should be added to the model rather than using the bad result. This model validation was the fundamental of the further analysis and conclusion. The more analysis and discussion are necessary to get a good model performance, then differentiate the SOA formation from aqueous oxidation of POA.

Response: We agree that examining the contribution of distinct SOA formation pathways is critical to understanding the daytime SOA underestimation for the 2014 simulation. In the revised text, we have added more discussion on the daytime formation of SOA in 2014. The model underestimated the midday peak of SOA in 2014, suggesting that daytime SOA formation (including SOA formation from aromatics, S/IVOCs, and aqueous pathway; Fig. S5 and Fig. S6) was likely underestimated in the 2014 model run. This underestimation stems from underestimated precursor emissions and/or low yields of these reactions during the day. In particular, SOA formation from aromatics, SVOCs/IVOCs, and the aqueous pathway exhibited a daytime increasing trend, while POA-aging SOA showed higher nighttime concentrations (Fig. S5 and S6). The current POA aging scheme assumes POA conversion occurs both day and night (Fig. S5), consistent with observations of bulk SOA during the 2024 field campaign. Our previous winter study in Shijiazhuang identified an OOA-NO_3 factor (Lin et al., 2020), which accounted for the majority (38–48%) of OOA formation pathways, demonstrating that daytime processes can produce fresh SOA with formation pathways and volatility similar to those of inorganic nitrate. Other identified OOA factors include sulfate-related OOA and aged POA from biomass burning and fossil fuel combustion (Lin et al., 2020).

Line 137, it now reads, "... As a result of this addition of the simplified SOA formation scheme, the model SOA and POA tracked the observed ones with a moderate correlation coefficient (Fig. S3). The diurnal pattern of SOA was simulated relatively well in 2024, whereas the model underestimated the midday peak of SOA in 2014 (Fig. S4), suggesting that daytime SOA formation (including SOA formation from aromatics, S/IVOCs, and aqueous pathway; Fig. S5 and Fig. S6) was likely underestimated in the 2014 model run. This underestimation stems from underestimated precursor emissions and/or low yields of these reactions during the day. In particular, SOA formation from aromatics, SVOCs/IVOCs, and the aqueous pathway exhibited a daytime increasing trend, while POA-aging SOA showed higher nighttime concentrations (Fig. S5 and S6). The current POA aging scheme assumes POA conversion occurs both day and night (Fig. S5), consistent with observations of bulk SOA during the 2024 field campaign (Fig. S4). As evidence of the underestimation of daytime SOA formation, our previous winter study in Shijiazhuang using comprehensive PMF identified a daytime-dominant OOA-NO_3 factor (Lin et al., 2020), which accounted for the majority (38–48%) of OOA formation pathways. This demonstrates that daytime processes can produce fresh SOA with formation pathways and volatility similar to those of inorganic nitrate. Other identified OOA factors include sulfate-related OOA (i.e., regional and low-volatility; 25–26% of total SOA) and aged POA (26–37%) from biomass burning and fossil fuel combustion (Lin et al., 2020). In contrast, direct PMF analysis in 2024 revealed that more oxidized oxygenated organic aerosol (MOOOA; 35% of total SOA) and POA aging-derived SOA (34%) were more important than less oxidized, relatively fresh OOA (LOOOA; 30% of SOA; Fig. S7). While SOA accounted for approximately 75% of total OA in 2024, its contribution was only 27% in 2014 (Fig. S7). Therefore, this comparison indicates that regional transport and POA aging during transport outweighed fresh SOA formation in 2024..."

Line 279, it now reads, "... As a result, the aging of POA under high RH, which leads to the formation of SOA, contributed to 25-50% of the total SOA in 2014-2024 (Fig. 4). Additionally, SOA generated from the oxidation of intermediate and semi volatile compounds comprised 19-24% of the total. Aromatic precursor oxidation accounted for 8-11% of the SOA, while SOA resulting from aqueous phase chemistry involving glyoxal and methylglyoxal contributed 22-38%. Compared to 2014, the contribution of the POA aging to total SOA increased substantially in 2024, becoming the dominant component (approximately 50%) in most cities, while the proportions of the aqueous reactions, S/I VOCs and Aromatic oxidation all declined to varying degrees (Fig. 4)..."

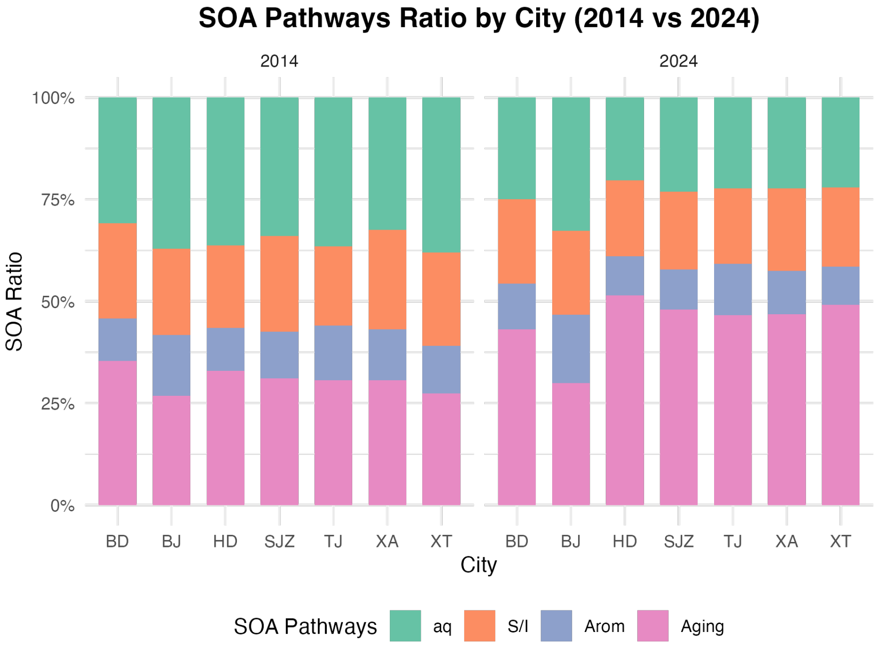

Figure 4. Fraction of different SOA formation pathways in cities of Baoding (BD), Beijing (BJ), Handan (HD,), Shijiazhuang (SJZ), Tianjin (TJ), Xi'an (XA), and Xingtai (XT) in 2014 vs. 2024.

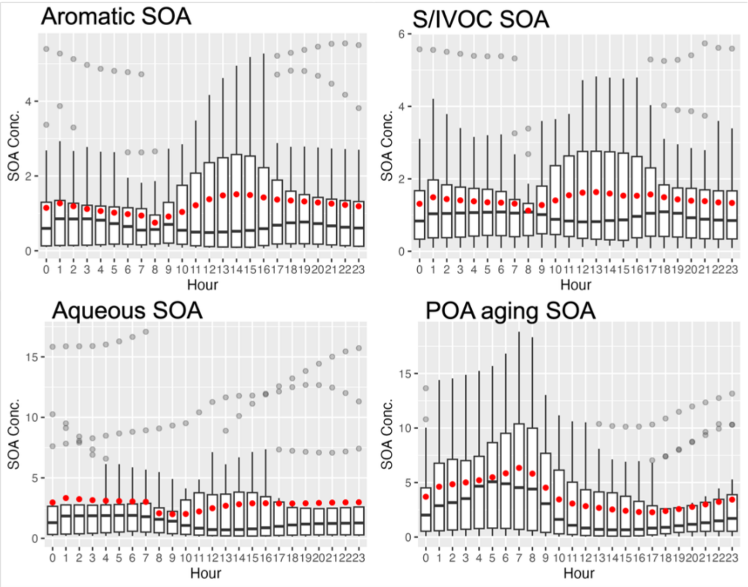

Figure S5. Diurnal variations in distinct SOA formation pathways in 2024, including SOA derived from aromatic oxidation, SVOCs/IVOCs oxidation, the aqueous pathway, and POA aging. Box plots display the median, 25th, and 75th percentiles; whiskers extend to 1.5 times the interquartile range (IQR). Grey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

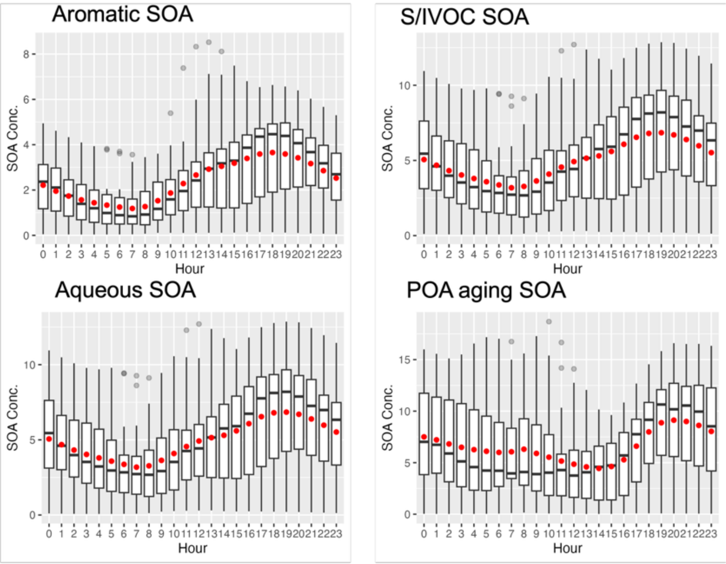

Figure S6. Same as Figure S5, but for 2014.

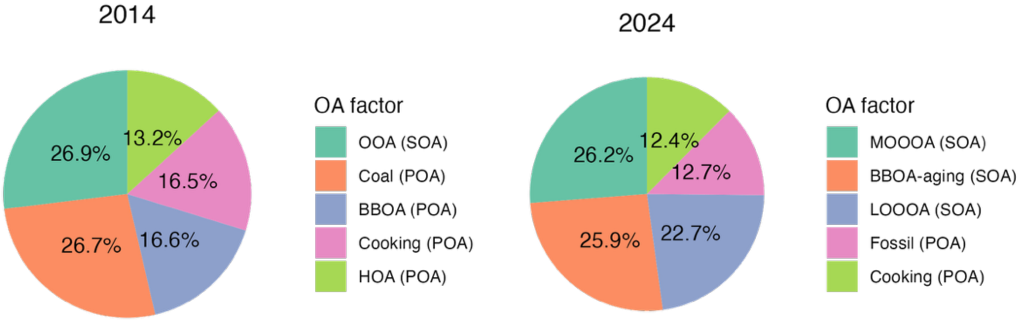

Figure S7. OA factor contributions in Shijiazhuang (2014 vs 2024). Non-cooking POA (ncPOA) represents the sum of all other POA fractions. SOA surrogates were resolved only as bulk OOA for 2014, whereas three distinct SOA types were identified for 2024, attributed to the use of high-mass-resolution AMS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

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