We thank the reviewer for their valuable comments, which have greatly helped improve our study. In response to these comments, we have carefully revised the manuscript. For clarity, the reviewer's comments are presented in black, our responses are highlighted in cyan, and the specific revisions made to the main text are marked in red.

Reviewer #1

Lin et al. investigated the growing role of secondary organic aerosol in PM2.5 pollution in Shijiazhuang by both field observations and modelling. They found that the contribution of SOA to the total OA exceeded that of POA during the winter haze in 2024 compared to 2014, and aqueous oxidation of POA might explain the increased SOA. The response of OA to emission reduction was also discussed. The results are interesting, and the manuscript is well-represented and organized. It is publishable after the following questions have been well addressed.

Response: We thank the reviewer for the positive comments. Please see our point-to-point response to each comment below.

In lines 105-106, the 2024 emission data is constructed based on the MECI 2020 and the 2024 measurement, but it is not clear for this scale.

Response: We have now clarified that we scaled MEIC 2020 by a factor of 0.5 to enable a better comparison between measurements and model results.

In line 114, it now reads, "... Since the emission inventories are not up-to-date for 2024, the MEIC data were downscaled from the 2020 MEIC version. A scaling factor of 0.5 was applied to enable more accurate comparisons between model outputs and 2024 measurements (Fig. S2)..."

In line 160, the PM1 components in 2014, but PM2.5 in 2024, will cause uncertainties when comparing the growth role of SOA. A reasonable discussion should be added.

Response: Thank you for pointing this out. We have now discussed the uncertainties in the size cut.

In line 179, it now reads, "... Considering the differences in size cuts (i.e., PM_1 from ACSM in 2014 and $PM_{2.5}$ from AMS in 2024) and the distinct sampling sites between the two campaigns (Huang et al., 2019), a direct comparison of the two campaigns may introduce limitations. 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."

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.

Response: Thank you for pointing this out. We have now discussed the impact of ALWC on SOA formation.

In line 158, it now reads, "...Moreover, changes in PM components are known to drive

differences in aerosol liquid water content (Kuang et al., 2020), and such changes may also alter the formation pathways of SOA..."

In Figure 2, although the correlation between SOA and RH is meaningful to understand the role of aqueous reaction in SOA formation, it should be more reasonable to discuss it using the fraction of ALWC in PM2.5 because ALWC should be related to RH, temperature, and chemical composition of aerosol.

Response: We agree that ALWC is closely related to RH, temperature, and aerosol chemical composition. However, our intent was to emphasize two key points: first, that POA concentrations reached a plateau under high RH conditions; second, that POA was not necessarily internally mixed with the inorganic components that play key roles in regulating ALWC (beyond the influence of RH itself). Given this context, we intend to retain the analysis of the correlation between SOA and RH, as this helps highlight the distinct responses of POA and SOA to RH.

OA was reduced by 79% in 2024 compared to 2014, with a 78% reduction during the most polluted period. It is meaningful to separate the reduction of POA and SOA.

Response: We have now added the reduction of POA and SOA.

In Line 193, it now reads, "... On average, ncPOA was reduced by 93% in 2024, while SOA was reduced by 45%..."

In lines 185-187, aqueous oxidation might promote SOA formation, while oxidation of SVOC and IVOCs might also be enhanced in 2024 because of increased oxidation capacity.

Response: Thank you for pointing this out. We have now added that the oxidation of S/I VOCs might also be enhanced in the revised manuscript.

In line 212, it now reads, "... Additionally, the oxidation of semi-volatile and intermediate volatile organic compounds (SVOCs and IVOCs) may have been enhanced due to increased oxidation capacity (Huang et al., 2025)..."

In Figure 3, the mean values of observed ncPOA and SOA concentrations should be shown for a better understanding of the performance of the model.

Response: We have now added the mean values of observed ncPOA and SOA in the updated Figure 3.

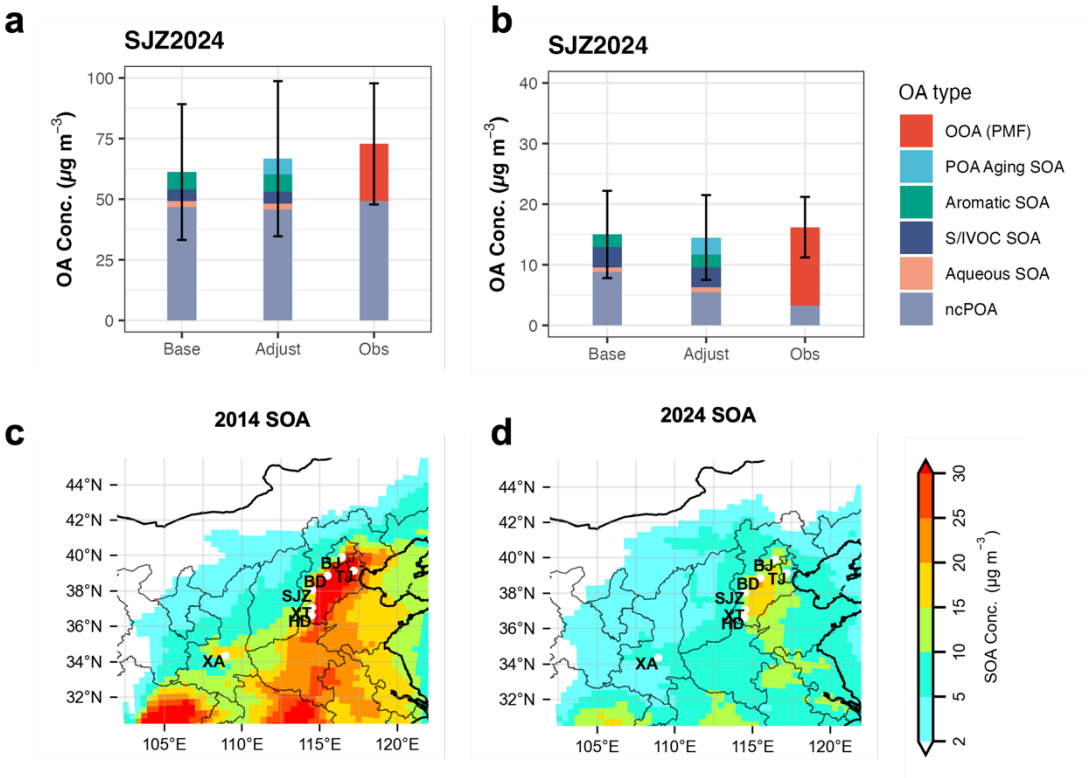

Figure 3. SOA Underestimation in the Model. (a) the base run (Base), and the adjusted (Adjust) CMAQ model for SJZ in 2014, along with (b) the corresponding data for 2024 as a comparison to the observation (Obs) from PMF result. (c) Distribution of the adjusted SOA in 2014 and (d) in 2024 during the pollution period. Error bars indicate one standard deviation.

In lines 205-206, the data in Figure 3b can not support this contribution (80) of SOA to OA. Please clarify. And it can give the detailed data (such as a table) for the cities in Figures 3c and 3d to make it easier to understand.

Response: We have now incorporated the observation data into Figure 3b to support this contribution. Additionally, a detailed table listing the cities featured in Figures 3c and 3d has been added.

In line 233, it now reads, "...In 2024, SOA constituted approximately 60% of the total OA for the adjusted model (Table S2) and 80% of the total OA for the observed OA (Fig. 3b), highlighting its dominance over POA..."

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 μg 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=21.1	2.1 + 3.3 + 0.8 + 2.8 = 9.0
Beijing (BJ)	6.5 + 3.7 + 2.6 + 4.7 = 17.6	2.7 + 3.2 + 1.3 + 2.0 = 9.2
Tianjin (TJ)	11.1 + 5.9 + 4.1 + 9.3 = 30.3	1.8 + 2.7 + 1.0 + 2.0 = 7.6
Baoding (BD)	6.2 + 4.7 + 2.1 + 7.1 = 20.3	2.1 + 3.4 + 0.9 + 3.0 = 9.4
Xingtai (XT)	7.5 + 4.5 + 2.3 + 5.4 = 19.6	1.7 + 3.1 + 0.7 + 2.2 = 7.8

Handan (HD)	11.7 + 6.5 + 3.4 + 10.6 = 32.3	2.2 + 4.5 + 0.9 + 3.3 = 11.0
Xi'an (XA)	5.2 + 3.9 + 2.0 + 4.9 = 16.1	1.2 + 2.5 + 0.6 + 1.3 = 5.7

In lines 206-210 and Figure 3, another concern is that the contributions of biogenic sources (such as isoprene and pinenes) and aliphatics to SOA are not considered, which will introduce some uncertainties to the aging of POA.

Response: We agree that biogenic sources contribution to SOA are not considered which may introduce uncertainties in the model. This is now discussed in the revised text.

In line 239, it now reads, "... Note that biogenic SOA was not considered in the CMAQ configuration. However, given a reduced emission of biogenic VOCs in winter, SOA formation potential from biogenic SOA was less important in winter than in summer (Dou et al., 2025), and may have a minor impact on the overall contribution of the anthropogenic sources..."

Why does the contribution of local emissions of ncPOA in SJZ increase greatly in 2024 compared to 2014?

Response: We have revisited the data, and in fact, the local emissions of ncPOA were similar between 2014 and 2024, accounting for 61% and 65% respectively.

In line 224, it now reads, "... CMAQ simulations indicate that, on average, 39% of ncPOA in 2014 was from regional transport, while 61% originated from local emissions in SJZ. In 2024, 35% was from local emissions and 65% from regional transport. However, during pollution episodes ($PM_{2.5}$ >75 μg m^{-3}), approximately 50% of ncPOA was sourced from regional transport in both 2014 and 2024 (Fig. S7). Given the large contribution from regional ncPOA, accounting for the aging of transported ncPOA into SOA could help resolve these model-measurement discrepancies..."

In lines 227-235, Figure S4 can not support this expression and conclusion. And the scenario setting in Section 3.3 is confusing, and it should be clarified. And the legend in Figure 3c should be checked. I think the difference shows the emission rather than meteorology.

Response: Thank you for pointing this out. Figure S4 is now changed to Figure S8. The difference shows emissions. We have now updated Table S1 to clarify the scenario settings in Sect 3.3.

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. 4)
MEIC 2014 + Meteorology 2024	Emission diff (Fig. 4)
MEIC 2024 + Meteorology 2014	Meteorological diff (Fig. S8)
MEIC 2024 (zero SJZ) + Meteorology 2024	Regional vs local (Fig. S7, S9)

MEIC 2014 (zero SJZ) + Meteorology 2014	Regional vs local (Fig. S7, S9)

In Figures S3 and S4, the performance of SOA modelling is terrible, particularly in 2014. The diurnal curve can not be correctly captured by the model. So, how are you sure about the SOA formation from aqueous oxidation of POA?

Response: In this study, we propose a POA aging in the aerosol phase can reduce the gap between simulated and measured SOA. The diurnal pattern of SOA was simulated relatively well in 2024, whereas the model missed the midday peak of SOA in 2014, suggesting that daytime SOA formation was more important in 2014 than in 2024 (Fig. S4). In line 131, it now reads, "... The diurnal pattern of SOA was simulated relatively well in 2024, whereas the model missed the midday peak of SOA in 2014, suggesting that daytime SOA formation was more important in 2014 than in 2024 (Fig. S4)...."

Figure S10 is not cited.

Response: It is now cited in Line 282.

The formatting problems should be checked and revised, such as the upper and lower marks (lines 84, 130-134, etc.), the fonts (lines 37-45, 62-70), and reference information, like journal, volume, pages, and doi (lines 343-345, 366-369, etc).

Response: The formatting is checked and revised for the entire text.

Reviewer #2

This manuscript investigated the differences in aerosol mass concentration and chemical composition between 2014 and 2024 for a large city in the North China Plain. The two campaigns were performed using a Q-ACSM and SP-AMS, respectively. The major finding was that with large reductions in primary emissions, the contribution of SOA to OA increased significantly, from 27% in 2014 to 87% in 2024. The results contribute to the understanding of long-term variations in ambient aerosols in China. I think this manuscript could be considered for publication if the following concerns could be properly addressed.

Response: We thank the reviewer for the positive comments. Please see our point-to-point response to each comment below.

First, the 2014 and 2024 measurements were conducted at different sites (i.e., urban and suburban, respectively), indicating that results from the two campaigns were not directly comparable. For example, it is not surprising that the SOA contribution was higher for the suburban site (i.e., the 2024 campaign), even if the anthropogenic emissions were unchanged. To explain the observed variations of SOA, the influences of different factors (e.g., primary emissions and the types of measurement site) should be properly distinguished. This is essential for an ACP paper.

Response: we agree that a direct comparison of the two campaigns may introduce limitations. However, given the strong time-series correlation in bulk $PM_{2.5}$ concentrations across cities in the NCP (Figure 5), this comparison can still reflect an overall trend. We have clarified these uncertainties in the revised text.

In line 179, it now reads, "...Considering the differences in size cuts (i.e., PM_1 from ACSM in 2014 and $PM_{2.5}$ from AMS in 2024) and the sampling sites between the two campaigns (Huang et al., 2019), a direct comparison of the two campaigns may introduce limitations. 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...."

Second, the CMAQ performance was questionable. As indicated by Figure S3, the observed and simulated SOA results in fact had little correlation. I don't think this level of agreement could properly support the related discussions in the main manuscript.

Response: Figure S3 has been updated to show that simulated SOA and POA tracked measured values with moderate correlations (R = 0.4–0.7). This comparison indicates that the model performance was acceptable.

In line 131, it now reads, "... As a result, 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 missed the midday peak of SOA in 2014, suggesting that daytime SOA formation was more important in 2014 than in 2024 (Fig. S4)..."

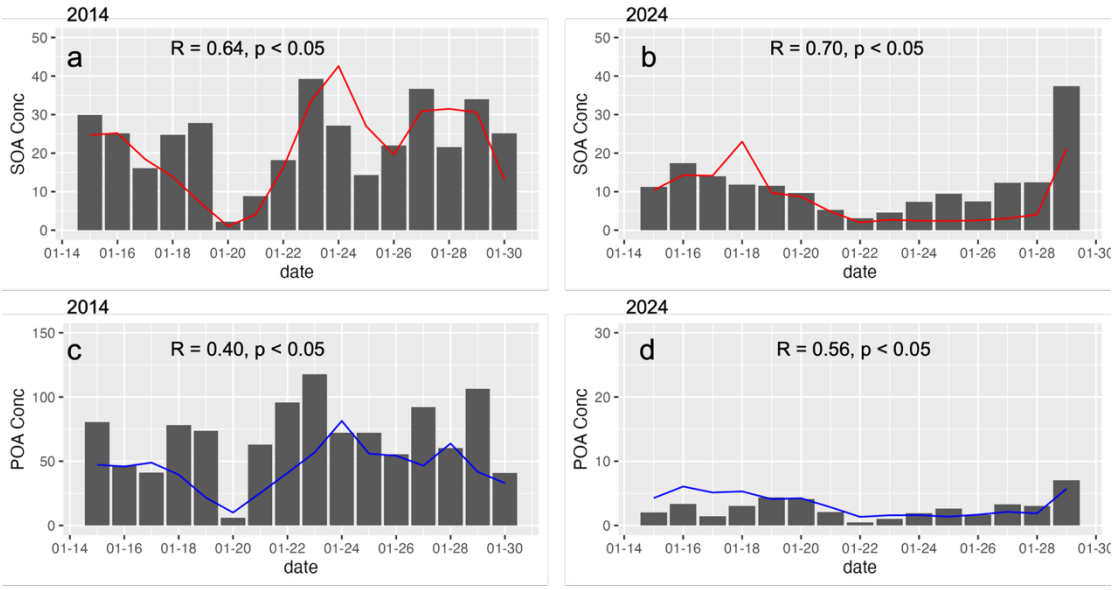

Figure S3. Comparison of simulated SOA and POA with observed values for 2014 and 2024. Bars represent measured daily values, while lines represent the corresponding modeled values.

Third, the comparison between CMAQ and AMS (or ACSM) results must also be performed for POA. A reasonable agreement is the precondition for the related discussions in section 3.2.

Response: We have added the comparison of POA to the revised Figure S3. (please see our previous reply for specific text edits)