Comments from Reviewer #1

General comments

This manuscript investigated the sources of black and brown carbon in Northeast China, by integrating observational and simulation results. An agreement between observed and modeled $PM_{2.5}$, especially with respect to the source-resolved chemical compositions, is essential to design efficient air pollution control strategies. Such comparisons have rarely been made for Northeast China, which possessed distinct primary sources and meteorological conditions compared to other regions in China. The authors observed efficient SOA formation at low temperatures during winter and abundant open-burning POA in spring, both of which could not be properly reproduced by CMAQ. The results contribute to the understanding of haze pollution in China. I think this manuscript could be considered for publication after addressing my following concerns.

Major comment

My major concern is that for the comparison of observational and modeling results, an interesting point was missing, i.e., the relationship between EC_{mod} and EC_{obs} during the fire episodes. The authors argued that open burning emissions and thus EC_{mod} were underestimated, whereas EC_{obs} were biased high due to fire-induced BrC. Then it would be expected that EC_{mod} and EC_{obs} should have larger differences for the fire episodes compared to other periods, but the authors did not show the comparison. If this inference did not hold, the major conclusions would be questionable.

Our responses: We thank the reviewer for the suggestion. In the revised manuscript, EC_{mod} and EC_{obs} were compared for the fire episodes as suggested, and their discrepancies were indeed more significant than the other periods in spring **(see lines 542-547)**:

"*It was also noticed that the mean bias in elemental carbon (EC_{mod} – EC^*) was more significant for the fire episodes (–1.26 μgC/m^3) compared to other periods in spring (–0.44 μgC/m^3). This pattern could be attributed to two factors, including the underestimation of open burning emissions by the inventory and the fire-associated overestimation of elemental carbon mass by EC^*. In other words, both EC_{mod} and EC^* were subject to larger uncertainties for the fire episodes, resulting in more significant model vs. observation discrepancies in elemental carbon concentration*".

Minor comments

(1) Lines 16-18. I guess what the authors want to say is "the understanding on the abundances and sources of light-absorbing carbon is still subject to non-negligible uncertainties". In other words, the "abundances and sources" themselves don't have uncertainties. This sentence needs to be re-organized.

Our responses: The sentence was rewritten as suggested: "*However, their abundances and sources remain poorly constrained, as can been seen from the frequently-identified discrepancies between the observed and modeled results*" **(see lines 16-18)**.

(2) Line 92. It should be "light-absorbing".

Our responses: The change was made as suggested **(see line 95)**.

(3) Line 206. I assume the authors mean "2020-2021", not "2021-2022".

Our responses: The typo was corrected **(see line 210)**.

(4) Line 223. Provide quantitative result for "close to zero".

Our responses: The change was made as suggested **(see line 227)**.

(5) Line 287. Why did TC decreased after extraction for the blank filters? Is the difference significant?

Our responses: The blank TC decreased slightly after the extraction (from 0.61 ± 0.23 to 0.44 ± 0.21 $\mu gC/cm^3$; the difference was statistically significant), with no EC detected for either the untreated or extracted filters. A possible explanation for the decrease was the dissolving of organic compounds, which constituted the TC of the untreated blank filters, into the solvent. The discussions above were incorporated into the revised manuscript **(see lines 290-295)**.

(6) Line 392. Provide quantitative description for "a considerable number".

Our responses: The change was made as suggested **(see line 399)**.

(7) Line 467. Clarify whether the mean bias was calculated as model – observation.

Our responses: The change was made as suggested **(see line 476)**.

(8) Figure 9. It would be better to use log scale for the y-axis of the upper panel.

Our responses: The change was made as suggested **(see lines 511-514)**:

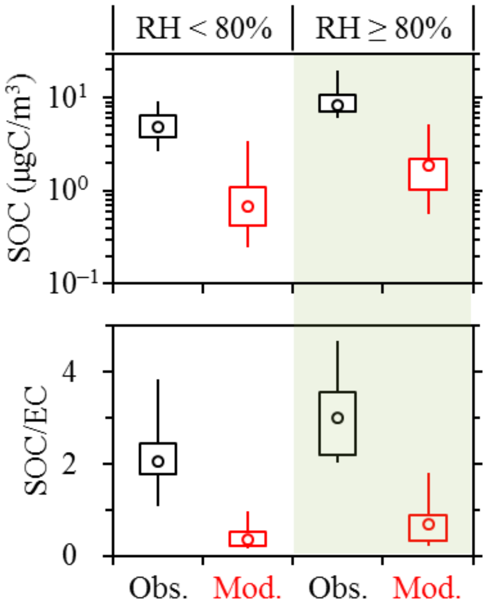

Figure R1. Comparisons of the modeled and observed SOC concentrations (upper panel) and SOC to EC ratios (lower panel) for the 2020–2021 winter. The comparisons were performed for the RH ranges of below and above 80% separately. Open burning impact was negligible for this period. This figure was presented as Figure 9 in the revised manuscript.

(9) Caption of Figure S12. OC^* and EC^* should be used for the equation of EC-tracer method. In addition, the firework periods should be clearly shown in the figure, i.e., clarify whether all the events without SOC results were associated with fireworks.

Our responses: The figure was revised as suggested:

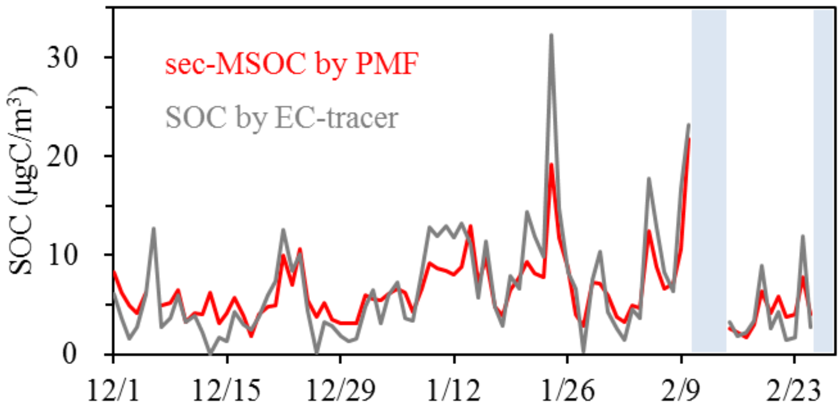

Figure R2. Variations of SOC derived from different approaches for the 2020–2021 winter. SOC was determined as secondary MSOC (i.e., sec-MSOC) based on PMF. In addition, SOC was also estimated by the EC-tracer method as OC – EC × $(OC/EC)_{min}$, where $(OC/EC)_{min}$ indicates the minimum OC to EC ratio; OC and EC results measured by the untreated samples deploying IMPROVE-A were used for the calculation. SOC resolved by the two approaches showed similar patterns of temporal variation and comparable mass concentrations, leading to a strong linear correlation ($r = 0.91$). As indicated by the shadowed periods, SOC was not estimated for the samples strongly impacted by firework emissions during the Chinese New Year Period. This figure was presented as Figure S12 in the revised manuscript.