

Comments from Reviewer #2

General comments

The manuscript by Cheng et al. explored the characteristics and sources of carbonaceous aerosols during three successive winters in Harbin. Samples were collected and analyzed for a variety of species, e.g., brown carbon, elemental carbon and levoglucosan. The authors evaluated the loss of EC during methanol extraction of filter samples, a long-lasting debate on the measurement method for BrC mass concentration. This artifact was suggested to be unimportant based on indirect evidences, providing valuable implications for future studies. The authors also explained the EC discrepancies between different analytical methods, and identified the OC/EC ratios (i.e., OC and EC results) that were in reasonable accordance with secondary aerosol formation. The authors then performed source apportionment of BrC and EC using the measurement results, and finally compared the observation-based attributions with those predicted by an air quality model. Overall, the results were properly interpreted and presented. However, as listed below, there are some major concerns on top of writing problems.

Major comments

(1) The authors only broadly stated that the high RH conditions in winter should favor heterogeneous formation of secondary aerosols. This statement could be more specific. For example, did the heterogeneous reactions occur in aerosol water like Beijing or on the surface of frozen particles due to the low temperature?

Our responses: We expanded the discussions on heterogeneous chemistry as suggested: *“In addition, aerosol water could remain supercooled at the typical temperatures during winter in Harbin, which were down to about $-25\text{ }^{\circ}\text{C}$ in terms of daily average (Rosenfeld and Woodley, 2000). For the frigid atmosphere in Northeast China, therefore, heterogeneous reactions in aerosol water were expected to prevail as long as RH reached sufficiently high levels. The mechanisms of low-temperature chemistry, which may differ from those in the relatively warm regions (e.g., Beijing), merit further investigations”* (see lines 505-510).

(2) Line 300. Suggesting toning down the statement, as this study did not have robust evidence for heterogeneous chemistry. Please note that the RH-dependent increases of SOR + NOR (Figures 1b and 1c) and SOC/EC (Figures 3 and 4) should only be

considered indirect evidences.

Our responses: We agree with the reviewer that this study lacks direct observational evidence for heterogeneous chemistry. In the revised manuscript, the sentence was rewritten as: “*The 2019–2020 campaign was characterized by unusually high levels of RH in winter, which were expected to favor heterogeneous chemistry*” (see lines 306-307).

(3) Please clarify whether it was acceptable to approximate MSOC as untreated OC. This point is important, given that thermal-optical analysis of the extracted filters could be laborious and time-consuming.

Our responses: We thank the reviewer for the suggestion. Our results suggested that it was acceptable to approximate MSOC as untreated OC measured by IMPROVE-A. This point was clarified in the revised manuscript: “*Results from the untreated samples using IMPROVE-A were found to provide OC to EC ratios in reasonable accordance with secondary aerosol formation.....the corresponding OC (OC*) approximately equaled MSOC, the determination of which was laborious. This equivalence supported the simplification of MSOC as OC* for further studies*” (see lines 560-566).

Minor comments

(1) Line 18. Suggest clarifying that EC is a measure of black carbon.

Our responses: The change was made as suggested (see line 19).

(2) Lines 25 and 27. Change SOA to SOC, as no SOA result was presented throughout the manuscript.

Our responses: The change was made as suggested (see lines 26 and 28).

(3) Line 77. Suggest providing an example for the “possible artifacts”.

Our responses: An example was provided as suggested: “*when determining BrC mass as the difference in total carbon concentration between untreated and extracted filters, the result could be biased high due to the loss of insoluble BC during extraction*” (see lines 78-80).

(4) Line 102. Full name should be given for PM_{2.5}.

Our responses: The change was made as suggested (see line 105).

(5) Line 142. Use “Results and discussion”.

Our responses: The change was made as suggested (see line 145).

(6) Line 160. Suggest adding a “the” before “filter”.

Our responses: The change was made as suggested (see line 163).

(7) Line 183. Suggest changing the first “as” to “since”.

Our responses: The change was made as suggested (see line 186).

(8) Line 249. Suggest re-writing this sentence as “Importantly, as shown in Figure 2a, Δ ATN were negligible.....”

Our responses: The sentence was rewritten as: “ *Δ ATN were negligible after excluding these two distinct cases (Figure 2a), suggesting that the loss of insoluble carbon (e.g., EC) should be minimal during our extraction procedures*” (see lines 253-255). We think this statement was more precise than the original description.

(9) Line 281. Suggest adding an “in turn” before “supported”.

Our responses: The change was made as suggested (see line 285).

(10) Line 325. Change “is” to “was”.

Our responses: The change was made as suggested (see line 332).

(11) Line 334. Suggest adding a “the” before “winters”.

Our responses: The change was made as suggested (see line 341).

(12) Lines 394-396. The statements did not hold for all the Harbin samples.

Our responses: The sentences were rewritten as: “*It is also noteworthy that for NIOSH, ~40% of the samples showed R_{NIOSH} values above 1, indicating that their $EC_{untreated}$ was even lower than $EC_{extracted}$. A possible explanation was that when using NIOSH, the NIOSH-based $EC_{untreated}$ also frequently underestimated the elemental carbon mass, and the underestimation could be more significant than that by $EC_{extracted}$* ” (see lines 398-404).

(13) Line 480. Remove the “the” before “seasonal”.

Our responses: The change was made as suggested (see line 490).

(14) Line 542. Suggested adding a “(i.e., MSOC)” after “BrC mass”.

Our responses: The change was made as suggested (**see lines 563-564**).