Dear Editor,

Manuscript number: acp-2021-522

Title: Dramatic changes in Harbin aerosol during 2018–2020: the roles of open burning policy and secondary aerosol formation

Many thanks to you and the referees for the valuable comments and suggestions. We have considered the points raised and revised our manuscript accordingly. Our detailed responses and relevant changes are presented below.

Comments from Reviewer #1

General Comments:

The manuscript by Cheng et al. investigated the characteristics of haze pollution during two sequential heating seasons in the central city of the Harbin-Changchun (HC) metropolitan area, with a focus on the major drivers responsible for the observed inter-annual variations of aerosol composition. Compared to traditional hotspots of air pollution studies in China (e.g., the North China Plain), HC is unique with respect to both meteorological conditions (e.g., the extremely cold winter) and anthropogenic emissions (e.g., those from the intensive energy use for central and space heating). Therefore, observational results from this region may provide additional insights into haze pollution in China. However, $PM_{2.5}$ in HC remains largely unexplored with limit studies, which may explain the relatively slow improvement of air quality in HC compared to the North China Plain. In this context, the authors did a good job of unfolding the chemical compositions and sources of $PM_{2.5}$ in HC. Many in the community of atmospheric science would be interested in the results presented. My overall assessment is that this manuscript could be considered for publication in ACP given the following concerns (mostly technical) could be properly addressed.

Specific comments

(1) Line 54, suggest changing “aerosol” to “aerosols”.

Our responses: The change was made as suggested **(see line 54)**.

(2) Line 58, “its effects on SOA…” should be better.

Our responses: The change was made as suggested **(see line 58)**.

(3) Line 97, suggest removing the "the" after "with"; in addition, references should be provided for "limited studies".

Our responses: The changes were made as suggested **(see lines 98-99)**.

(4) Line 177, as indicated by the PMF source profiles, it seems that zero EC was apportioned into the two factors representing secondary aerosols. Please confirm and revised the description if necessary.

Our responses: Yes, zero EC was apportioned into the secondary-aerosol factors. This point was clarified in the revised manuscript **(see line 168)**.

(5) Lines 207-208, I think the "during 2019-2020" here is redundant and unnecessary.

Our responses: The phrase was removed as suggested **(see lines 274-275)**.

(6) Lines 227-228, rephrase "the same OC average concentrations".

Our responses: The phrase was revised to "the same OC levels" **(see lines 294-295)**.

(7) Line 235, references should be provided for the ISORROPIA model.

Our responses: References were added as suggested **(see line 148)**.

(8) Line 249, suggest adding a "usually" before "not".

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

(9) Line 261, suggest adding a "the" before "positive".

Our responses: The change was made as suggested **(see line 344)**.

(10) Line 287, the meaning of "those" is not clear enough.

Our responses: "Those" was changed to "the corresponding values" for clarity **(see line 370)**.

(11) Line 291, references are necessary for this statement.

Our responses: Reference was added as suggested **(see line 375)**.

(12) Line 307, suggest adding an "In addition" before "an obvious difference…"

Our responses: The change was made as suggested **(see line 391)**.

(13) Line 318, suggest changing "emission" to "emissions".

Our responses: The change was made as suggested **(see line 402)**.

(14) Line 332, please note that the more rapid decrease of SO_2 emissions compared to NO_2 is valid only for recent years in China.

Our responses: This point was clarified in the revised manuscript: "*...which typically indicated a more rapid decrease of SO_2 emissions compared to NO_2 during recent years in China*" **(see lines 416-417)**.

(15) Line 637, change "contribution" to "contributions".

Our responses: The change was made as suggested **(see lines 755-756)**.

(16) Line 647, suggest adding an "an" before "ambient", and changing "with" into "at".

Our responses: The changes were made as suggested **(see line 765)**.

(17) Annotation in Figure 8, suggest changing "Stronger impact of…" to "Stronger impacts of…"

Our responses: The change was made as suggested **(see Figure 8)**.

(18) Caption of Figure S4, suggest using "increasing strengths of biomass burning impact".

Our responses: The change was made as suggested **(see Page 9 in supporting information)**.

(19) Caption of Figure S6, I guess something was missing after "Relationship between OC/EC".

Our responses: It should be "Relationship between OC/EC and sulfate". This mistake was corrected in the revised manuscript **(see Page 11 in supporting information)**.

(20) Caption of Figure S7, rephrase the sentence "For the 2019-2020 campaign, $(OC/EC)_{pri}$ and OC^* were determined as the slope…"

Our responses: This sentence was re-written as: "*For the 2019–2020 campaign, $(OC/EC)_{pri}$ and OC^* were determined based on linear regression of OC on EC (r = 0.98), with $(OC/EC)_{pri}$ as the slope (2.13) and OC^* as the intercept (3.11), respectively, using low-RH samples (i.e., those with RH below 60%)*" **(see Page 12 in supporting information)**.

(21) Captions of Figures S9 and S10, it is unnecessary to repeat the definition of "D" and "H" conditions.

Our responses: The duplicated definitions were removed as suggested **(see Pages 14 and 15 in supporting information)**.

(22) Caption of Figure S11, suggest changing "temperature" to "temperatures".

Our responses: The change was made as suggested **(see Page 16 in supporting information)**.

Comments from Reviewer #2

General Comments:

This work investigates the aerosol chemical composition in Harbin in the past 2 years based primarily on in-situ measurement. Generally, I find that this manuscript may be more like a measurement report since that it does not provide sufficient new insight into atmospheric chemistry but the dataset seems unique and presents some new perspectives. One main problem with this work is that it entirely attributed the variations of secondary aerosol to the chemical processes, but aerosol does have a lifetime of 1~2 weeks and regional transport could largely contribute to the temporal variation at one specific site. Overall, I think this work fits the scope of ACP but there are some issues that need to be addressed to improve this work.

Our responses: We thank the referee for the constructive comments. The referee raised one major point regarding the roles of meteorology and regional transport, and further expanded in the specific comment #2. Please refer to our responses to that specific comment #2 for details.

Major Comments:

(1) The method ought to be detailed in the main text. Section 2 is too sketchy, in which some basic information like the geographic location of the campaign site, its representativeness, the main emission sources in the surrounding areas, the instrumentation and the QAQC should be included. Also, other analysis like AWC, Positive Matrix Factorization (PMF) results discussed in Section 3, which is not from the direct observation, should be described in this section. This article itself missed a lot of necessary information on the method and analysis tools. I suggest moving the supplementary method to the main text. Another issue related to the method is that PMF analysis usually needs a large amount of data to support the factorizations since that it basically is statistical analysis. According to the Supporting Information, the sampling number is around 200~300, which is not enough to get a reliable result.

Our responses: Two points were raised in this comment. The first one was that the Methods section (i.e., Section 2) should be expanded substantially, by including all the necessary information. Following this suggestion, (i) detailed descriptions of the filed campaign, which was originally presented as supplementary information, was moved to the main manuscript, and (ii) procedures of the ISOROPPIA and PMF analyses were

also moved to the Methods section. In the revised manuscript, Section 2 was re-organized as:

2.1 Field observation and additional data sets used

Two campaigns were conducted at an urban site located in the campus of Harbin Institute of Technology (HIT; 45°45'24" N, 126°40'49" E) during the heating seasons of 2018–2019 (from 16 October, 2018 to 14 April, 2019; N = 180) and 2019–2020 (from 16 October, 2019 to 4 February, 2020; N = 112), following the same sampling and analytical procedures. As described for the 2018–2019 campaign (Cheng et al., 2021a), a low volume sampler (MiniVol; Airmetrics, OR, USA) operated at a flow rate of 5 L/min was used to collect airborne $PM_{2.5}$ onto pre-baked quartz-fiber filters (2500 QAT-UP; Pall Corporation, NY, USA), and the measured species included organic carbon (OC), elemental carbon (EC), organic tracers for biomass burning (levoglucosan and mannosan) and water-soluble inorganic ions (sulfate, nitrate, ammonium, etc.). Briefly, OC and EC were determined by a thermal/optical carbon analyzer (DRI-2001; Atmoslytic Inc., CA, USA), using the IMPROVE-A temperature protocol with transmittance charring correction. Precision of the carbon analyzer was investigated by analyzing the samples using another protocol (NIOSH). Comparisons of total carbon and optical attenuation results between the two protocols suggested good repeatability for both the carbon and transmittance measurements (Figure R1). Levoglucosan and mannosan were detected by a Dionex ion chromatography (IC) system (ICS-5000$^+$; Thermo Fisher Scientific Inc., MA, USA), using the high-performance anion-exchange chromatography coupled to pulsed amperometric detection (HPAEC-PAD) method. In addition, the IC was also used to measure the inorganic ions. Precision of the IC was evaluated by analyzing selected solutions 5–10 times, and the relative standard deviations were found to be within 5% for all the water-soluble species detected, either organic or inorganic. Based on the observed aerosol components, $PM_{2.5}$ mass was reconstructed as the sum of organic matter (determined as 1.6 × OC), EC and inorganic ions. The reconstructed $PM_{2.5}$ will be specified as $(PM_{2.5})^*$ in the flowing discussions.

Air quality data including $PM_{2.5}$, sulfur dioxide (SO_2), nitrogen dioxide (NO_2), carbon monoxide (CO), etc. were obtained from China's National Urban Air Quality Real Time Publishing Platform (http://106.37.208.233:20035/). They were measured at monitoring sites operated by the China National Environmental Monitoring Center (CNEMC), and could be accessed with a time resolution of 1 hour. There are a total of

12 CNEMC sites in Harbin. Results from the nearest one to the filter sampling site (~2.4 km apart), i.e., Taiping Hongwei Park, were used in this study. In addition, hourly meteorological data including temperature and relative humidity (RH) were obtained from Weather Underground (https://www.wunderground.com).

Using levoglucosan as the reference component, the relative abundances of water-soluble potassium (K^+) were found to increase substantially for five samples collected during the Chinese New Year periods in February of 2019 (N = 2; Cheng et al., 2021a) and in January of 2020 (N = 3; Figure S2), pointing to significant influence of firework emissions. Given that such emissions may result in primary sulfate and nitrate which are difficult to quantify, the firework events were excluded, and the remaining sulfate and nitrate were considered secondary in the following discussions. Then taking together observational results from the filter sampling and CNEMC sites, the sulfur oxidation ratio (SOR) was determined as the molar ratio of sulfate to the sum of sulfate and SO_2, and the nitrogen oxidation ratio (NOR) was determined similarly based on nitrate and NO_2.

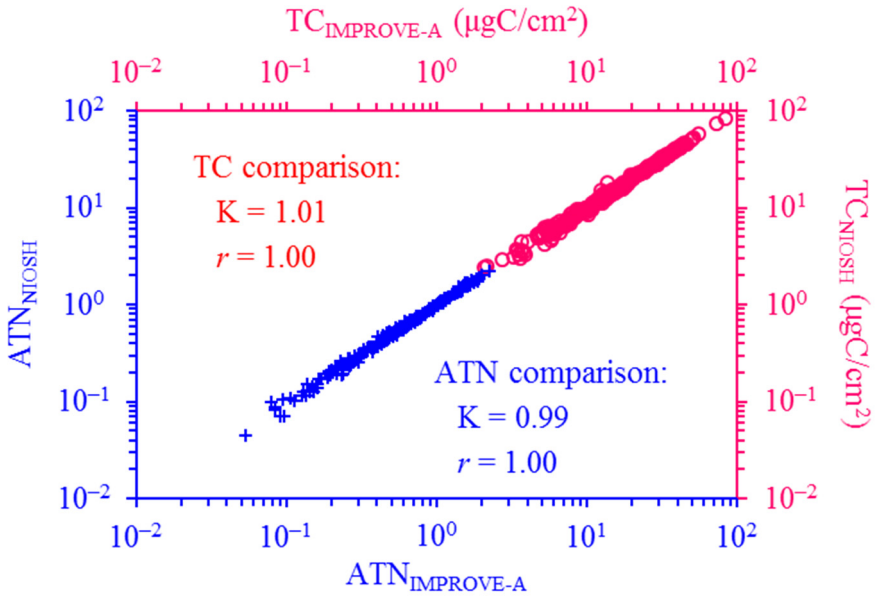

Figure R1. Comparisons of total carbon (TC) and optical attenuation (ATN) measured by different temperature protocols. Results from both campaigns are involved. ATN is calculated as $\ln(I_{final}/I_{initial})$, where $I_{initial}$ and I_{final} indicate filter transmittance signals measured at the beginning (i.e., when the loaded filter has not been heated) and end (i.e., when all the deposited carbon has been combusted off the filter) of thermal-optical analysis, respectively. Linear regression results are shown with K as slope (intercept was set as zero). TC and ATN agreed well between different protocols, demonstrating good precisions for both the carbon and transmittance measurements.

2.2 Thermodynamic simulation

The ISORROPIA-II model (Fountoukis and Nenes, 2007) was used to predict aerosol water content (AWC) and aerosol pH. The calculations were conducted in two ways, namely the "reverse" and "forward" modes. For the "reverse" mode, the measured aerosol-phase data were deployed as input to derive AWC and pH directly. For the "forward" mode, the model was run in an iteration way (Liu et al., 2021a). Briefly, we used the measured aerosol-phase data as initial input, ran ISORROPIA-II in the "forward" mode to predict gas-phase concentrations of semi-volatile species (e.g., ammonia and nitric acid), and used the sum of predicted gas-phase and measured aerosol-phase concentrations as the input for next round. The calculations were repeated until the simulated results were stable and in line with the observational data. Although the "reverse" and "forward" mode simulations showed comparable AWC levels for this campaign (Figure S3), the latter approach has been suggested to give more accurate and robust estimation of pH (Guo et al., 2017; Song et al., 2018). Thus, AWC and pH results predicted by the iteration approach were used in the following discussions.

2.3 Source apportionment

Source apportionment was performed using EPA's Positive Matrix Factorization (PMF) model (version 5.0), with times series of OC, EC, levoglucosan, chloride, nitrate, sulfate and ammonium from both campaigns as inputs. A total of five factors were resolved, and their profiles were shown in Figure S4. Two factors (BB-1 and BB-2) were strongly associated with primary biomass burning emissions, since almost all the levoglucosan (~90%) were apportioned to these two factors whereas neither of them was a major contributor to secondary ions. Another two factors were inferred to represent secondary aerosols (SA-1 and SA-2), as they had zero EC but the majority of nitrate and sulfate. The last factor (non-BB_{pri}) was attributed to primary emissions from non-BB sources, because more than 50% of EC but little levoglucosan was found in this factor.

The second point raised in this major comment was about the reliability of the PMF analysis. As pointed out by the reviewer, a large number of samples are usually required to get reliable source apportionment results, while the User Guide of PMF recommends that a minimum of 100 samples should be involved in the analysis. In this study, a total of 292 samples were collected, with 180 and 112 from the 2018-2019 and

2019-2020 campaigns, respectively. After excluding those heavily impacted by firework emissions ($N = 5$), all the remaining samples ($N = 287$) were used for the PMF analysis. We think this number of samples could generally be considered acceptable. In addition, we compared the source profiles derived by this study with those obtained by Cheng et al. (2021a), which were based only on the 2018-2019 campaign. As shown by Figure R2, the profiles were quite similar between the two studies. We think this consistency could to some extent support the reliability of the PMF results.

The discussions above were reflected in the revised manuscript **(see lines 107-194, 237-246, 318-319, Figures S1 and S4)**.

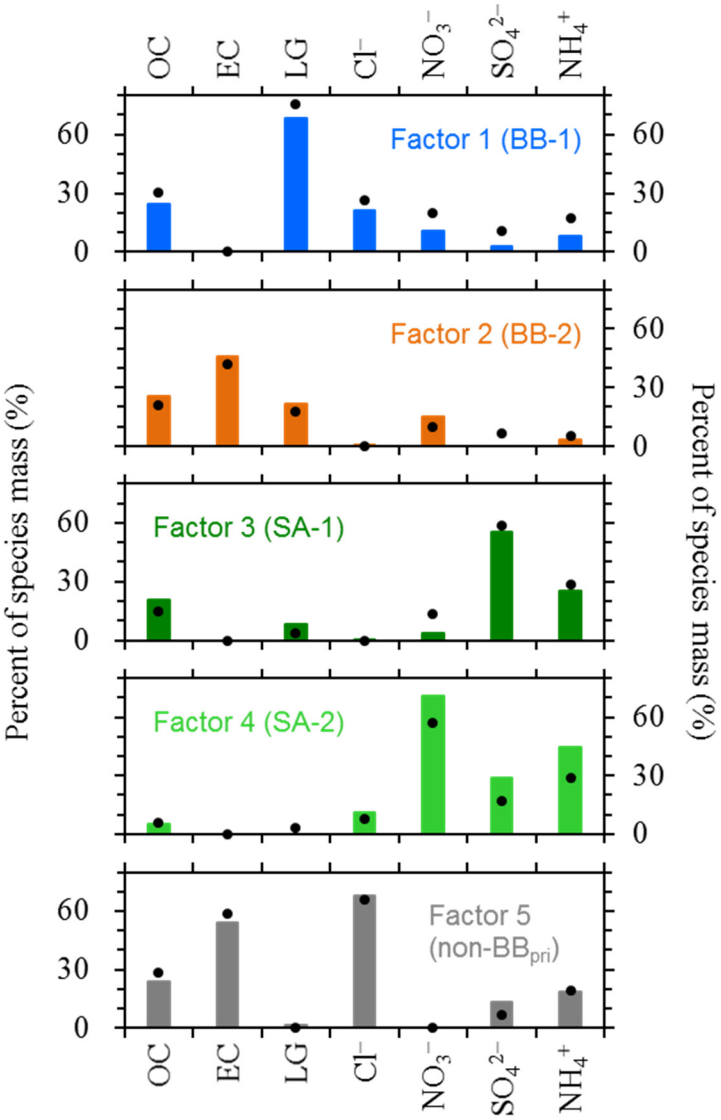

Figure R2. Source profiles resolved by PMF. The solid bars and circles indicate results obtained by this study and Cheng et al. (2021a), respectively. For this study, aerosol compositions measured during the 2018–2019 and 2019–2020 campaigns were

combined and used as the PMF inputs, whereas Cheng et al. (2021a) was based only on the former campaign. In general, similar profiles were resolved by the two studies, despite the different measurement periods covered.

(2) The discussion part is too descriptive and the majority of the main text is just describing the variation of different aerosol components. In some places, the authors jumped to some conclusions without careful investigation. For instance, the RH-dependent increase of OC was attributed to heterogeneous reactions without investigating the cloud-water chemistry and diffusion/dispersion analysis (Line 236-238). Additionally, the author concluded that the inter-annual (I would not call it inter-annual since it is just a two-year comparison) variation of OC was related to RH levels without other vital information of the regional transport pattern and other meteorological parameters. Besides, the higher threshold RH for the sharp increase of SOR in Harbin is not certainly indicative of the fact that the heterogeneous formation of sulfate was less efficient there. Many previous works have pointed out that the polluted air mass from the southern area of Beijing also brings about humidity, rather than heterogeneous chemical reactions at a local/city scale. Thus, some in-depth analysis and rigorous arguments need to be added to this work.

Our responses: We agree with the referee that some discussions in the original manuscript were not sound enough, especially regarding the RH-dependent increase of secondary OC (OC_{sec}), the inter-campaign variation of OC_{sec}, and Harbin's higher threshold RH for sharp increase of SOR compared to Beijing. As pointed by the reviewer, a major problem was that all the observed phenomena were attributed to the influence of atmospheric chemistry, i.e., the roles of other factors such as meteorology and transport were ignored.

In general, air quality modeling is usually required to quantitatively evaluate the contributions of various factors to the variations of $PM_{2.5}$ concentration and composition. If applied to this study, simulations need to be performed for six scenarios: (A–B) actual emissions and meteorological conditions of the 2018–2019 and 2019–2020 measurement periods, respectively; (C) 2018–2019 emissions with 2019–2020 meteorological conditions; (D) 2019–2020 emissions with 2018–2019 meteorological conditions; (E–F) zero emissions from Harbin for the two periods, respectively. The A vs. C and B vs. D differences indicate the influence of meteorology; the A vs. D and B vs. C differences can be attributed to the influence of emissions; the A vs. E and B vs. F differences point to the influence of regional transport. Of course, a precondition for

these simulations is that the model could properly reproduce the observational results. Actually, we have performed the simulation for the 2018–2019 measurement period (i.e., scenario A), using a revised CMAQ model (Hu J. et al., 2016). Compared to the original version (5.0.1), the revised model involved a modified Statewide Air Pollution Research Center (version 11; SAPRC-11) photochemical mechanism, which includes the isoprene epoxydiols (IEPOX) and methacrylic acid epoxide formation pathways, and allows predictions of glyoxal and methylglyoxal formed by oxidation of various precursors including isoprene. In addition, heterogeneous pathways were incorporated into the revised CMAQ to account for secondary inorganic and organic aerosols formed through reactive uptake of gaseous species on aerosol surfaces. The simulations were performed over East Asia with a horizontal resolution of 36×36 km. The meteorological inputs were retrieved from the Weather Research and Forecasting (WRF) model. The emission inputs were generated by combining various inventories, e.g., the Multi-resolution Emission Inventory for China (MEIC; http://www.meicmodel.org/) was used to derive anthropogenic emissions of OC, EC, volatile organic compounds (VOCs), SO_2, NO_2, etc., whereas the satellite-based Fire INventory from NCAR (FINN; Wiedinmyer et al., 2011) was used for open burning emissions. When comparing the observational and modeling results, large gaps were observed for two cases. The first one was agricultural fire episodes. For example, the simulated $PM_{2.5}$ accounted for only ~30% of the observed concentrations during February 25 to March 2, 2019, when intensive fire counts were detected (Figure R3a). In addition, the large differences in $PM_{2.5}$ could be attributed mainly to the underestimation of organic aerosol (OA, determined as 1.6×OC) mass by the model, and eventually pointed to the underestimation of open burning emissions by the FINN inventory. The second case was high RH conditions, when enhanced formation of secondary aerosols was evident. Such a period occurred during 12 to 15 January, 2019, showing increased RH (exceeding 80%) and elevated levels of SOR and NOR. The simulated sulfate were substantially lower than the observed concentrations during this period (with averages of ~8 and 27 $\mu g/m^3$; Figure R3b), although the model involved both in-cloud and heterogeneous pathways for sulfate formation. The agricultural fire-related underestimation of OA could be resolved by adjusting the open burning emission (Uranishi et al., 2019); however, the RH-related underestimation of sulfate by the revised CMAQ has not been addressed. Thus, we prefer not to conduct additional simulations (e.g., for scenarios B–E) at the current stage.

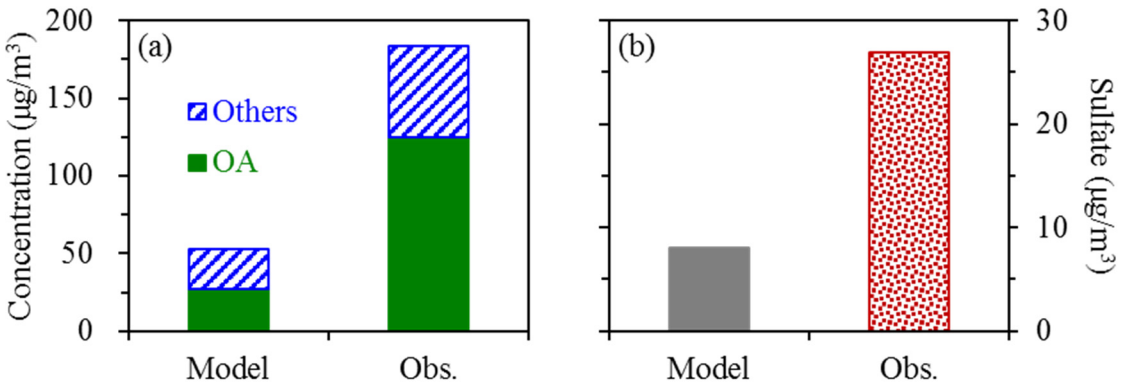

Figure R3. (a) Comparison of modeled and observed $PM_{2.5}$ for a typical agricultural fire episode, with concentrations of OA and other species shown separately. **(b)** Comparison of modeled and observed sulfate concentrations for a high RH period.

Given that variation of meteorological conditions could indeed influence the absolute concentrations of a given species (e.g., OC_{sec}) but will not change its relative abundances (e.g., the OC_{sec} to OC ratio and the OC_{sec} to EC ratio), in the revised manuscript, the comparisons among different cases (e.g., low-, medium- and high-RH conditions of 2019–2020) were primarily performed using various ratios and fractions. In addition, it was also clearly stated that our discussions on heterogeneous reactions did not exclude the presence of in-cloud chemistry or indicate secondary aerosol formation on a city scale. Specifically, the following changes were made:

(i) The discussions on the OC_{sec} (the second and third paragraphs of section 3.2 in the original manuscript) were reorganized as **(see lines 296-329)**:

As shown in Figure 3, OC_{sec} exhibited a positive dependence on RH, with an explosive increase of OC_{sec} after RH exceeded 80%. Only ~6% of the 2018–2019 samples (10 out of 180) experienced such humid conditions, whereas this fraction was as high as ~37% for 2019–2020 (corresponding to 42 out of the 112 samples). Thus the potential influence of RH on SOA formation was primarily investigated based on results from the 2019–2020 campaign. Figure 4 compares OC source apportionment results across different RH ranges (< 60%, 60–80% and > 80%), which are termed low-, medium- and high-RH conditions, respectively, for this measurement period. Not only OC_{sec} but also its contribution to OC (f_{sec}) increased significantly from the low- through high-RH conditions, by factors of 9.8 and 2.6, respectively. Although the 2019–2020 campaign experienced much lower ambient temperatures (as low as –20 °C) compared to Beijing's winter (~0 °C), the f_{sec} of Harbin reached 42% for the RH range of > 80%, generally comparable with the typical range of oxygenated organic aerosol (OOA)

contribution (~35–60%) determined under humid winter conditions in Beijing (Sun et al., 2013, 2014, 2018; Hu et al., 2016; Xu et al., 2019). Given the considerable AWC levels predicted for the high-RH conditions (typically above 50 μg/m^3; Figure 3), it was inferred that heterogeneous reactions might be at play in the RH-dependent increase of f_{sec}.

(ii) The statement arguing the less efficient sulfate formation in Harbin was removed **(see lines 372-373)**:

However, the threshold RH for sharp increase of SOR was higher in Harbin (80%) than that in Beijing (~40–70%), and the SOR in Harbin with RH above 80% (averaging 0.2) were at the lower end of those the corresponding values observed during winter in Beijing (typically with averages of ~0.2–0.6) (Sun et al., 2013; Zheng et al., 2015b; Zhang et al., 2018; Li et al., 2019; Liu et al., 2020). A likely cause for these differences was the relatively low temperatures during the measurement period, which would reduce the rate coefficients of relevant aqueous-phase reactions (Cheng et al., 2016).

(iii) Conclusions and implications were presented in separate sections, with the limitations of this study (i.e., the future work that needs to be done) being clearly stated **(see lines 469, 495 and 514-523)**:

It should be noted that the discussions on heterogeneous formation of SOA and SNA did not necessarily exclude the reactions in fog/cloud water. Actually, based on the observational results available, we could not robustly distinguish the relative importance of various aqueous-phases pathways for secondary aerosol formation. To address this problem, air quality modeling with a focus on HC should be conducted in future studies, which could also quantitatively evaluate the contributions of various factors (e.g., meteorology, emissions and regional transport) to long-term trends of $PM_{2.5}$ concentration and chemical composition. An essential precondition is that the model could properly re-produce the observational results, which appears a substantial challenge for Harbin as indicated by the limited inter-comparison studies, especially for the periods with intensive agricultural fires or high RH levels (Cheng et al., 2021b).

(3) There are also some inconsistent arguments that needed to be checked. For instance, in the abstract, the authors claimed that "we found that open burning activities were actually not eliminated ……", but Line 168 stated that "indicating that agricultural fires

were almost completely eliminated during the measurement period". It doesn't seem very clear.

Our responses: Regarding the open burning activities under the "strict prohibition" policy, the message we would like to convey was actually that "although not occurred during the 2019–2020 measurement period, agricultural fires broke out within a short period before crop planting in spring of 2020". All the inconsistent/misleading descriptions were corrected in the revised manuscript. In addition, the use of "inter-annual" was avoided following the suggestion raised in the major comment #2. **(See lines 36, 234, 315 and 472)**.

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Again, we thank the referees very much for their valuable comments and suggestions.

Sincerely yours,

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