Reply to comments on “Impacts of condensable particulate matter on atmospheric organic aerosols and fine particulate matter ($PM_{2.5}$) in China” by Mengying Li et al.

We thank you for all the constructive comments and suggestions. We have adopted all of the suggestions in our revised manuscript. The followings are our point-to-point responses to the reviewer’s comments. The responses are shown in brown and bold fonts, and the added/rewritten parts for the revision are presented in blue and bold fonts.

Reply to Reviewer #2

Organic aerosol, especially secondary organic aerosol (SOA), is a major component of the overall aerosol loading in various environments around the globe and significantly influences the air quality and climate. However, there is a significant gap between observed and modeled SOA. One possible reason is the incomplete information on emissions and properties of SOA precursors. This work modified the aerosol emission inventory by including condensable particulate matter (CPM) and tested this inventory by simulating some observations in Beijing and Beijing-Tianjin-Hebei region. The manuscript is overall well-written and fits the scope well of ACP. However, I have a few comments to be addressed before this manuscript can be published.

Response: We truly appreciate all the constructive comments and suggestions of the reviewer. We have adopted all the suggestions in our revised manuscript.

1. Estimations of CPM emissions (Line 160 – 198): It is reasonable to assume that CPM contains many condensable substances, such as nitric acid, ammonia, S/IVOCs, and the gas-particle partitioning of which is regulated by temperature. Therefore, have many of the CPM components already been considered in the gas emissions inventory? This may lead to repeated consideration of many compounds and further overvaluing of emission intensity; please clarify how organic (gas + particle) emissions were considered in this study?

Response: We thank the reviewer for the good suggestion. Since the current measurement methods for PM in stationary exhaust sources in China (GB/T 16157-1996) have only involved the collections of FPM, the CPM components have not been considered in the emission inventory. First， for inorganic gases, the gas emission inventory contained SO_2, NOx, CO and NH_3, thus the condensable substances in CPM, such as nitric acid and ammonia have not been included in the current emission inventory. Then for organic gases, the current gas emission inventory only contained gaseous NMVOCs directly emitted by the exhaust sources, which was different from the CPM components. For organic (gas + particle) emissions in this study, the organic CPM is a mixture of components which is semi-volatile (SVOCs, $10^0 \leqslant C^* \leqslant 10^3$ µg m^{-3}), or has intermediate volatility (IVOCs, $10^3 < C^* \leqslant 10^6$ µg m^{-3}), thus it is definitely different from the organic gases with higher volatility ($C^* \geqslant 10^7$ µg m^{-3}) in the current emission inventory. Considering that organic FPM from the stationary combustion and mobile sources mainly contained low volatile matter, so all of these emissions were

assigned to the LVPO1 species in the CMAQ. Then the organic CPM components were assigned to the CMAQ species of different volatility bins (LVPO1, SVPO1, SVPO2, SVPO3 and IVPO1). As mentioned in Sect. 2.1, organic CPM is composed of organic matter which is semi-volatile or has intermediate volatility, thus the first bin which represents nonvolatile organic matter should be set to zero. These mixed species underwent gas-particle partitioning and multi-generational gas-phase photochemical oxidations to generate SOA. Therefore, the inclusion of organic CPM did not lead to repeated consideration of many compounds and overvaluing of emission intensity.

2. The model configuration (Line 200 – 246): The emitted CPM can be further oxidized in the atmosphere to produce SOA. How does the model consider this process? It seems there is no component information of the CPM. Therefore, it is not clear to me how CPM performs atmospheric chemical reactions.

Response: We thank the reviewer for the suggestion. As stated in the Sect. 4 Conclusions, one of the limitations in this study was that there was no explicit component information and volatility characterization of primary organic CPM species available. Thus the organic CPM emissions were lumped into the original surrogate volatility species (LVPO1, SVPO1, SVPO2, SVPO3, and IVPO1) in the CMAQ model for representing the SOA formation from CPM. These mixed species underwent gas-particle partitioning and multi-generational gas-phase photochemical oxidations of organic vapors by OH radicals to generate successively lower volatility and more-oxygenated species, and then produce SOA.

Added/rewritten part in Sect. 2.3 Design of sensitivity simulation cases: the emissions of organic CPM were mapped to surrogate species for different volatility bins (LVPO1, SVPO1, SVPO2, SVPO3, and IVPO1) in the CMAQ model for representing the SOA formation from CPM. These mixed species underwent gas-particle partitioning and multi-generational gas-phase photochemical oxidation of organic vapors by OH radicals to generate successively lower volatility and more-oxygenated species, and produce SOA.

3. Line 241: Please provide the reasons for setting the emissions to be reduced by 30%.
Response: We thank the reviewer for the suggestion. We have provided the reasons for setting the emissions to be reduced by 30% in the revision.

Added/rewritten part in Sect. 2.2 The model configuration: Based on the observed reductions in the concentrations of $PM_{2.5}$, SO_2, NO_2, and CO during APEC in Beijing and its surrounding cities (Li et al., 2017e, 2019; Wen et al., 2016), and 28% contribution of the emission control measures to the reduction of $PM_{2.5}$ concentrations (Liang et al., 2017), thus the approximate emission reduction of 30% was conducted during the above time period for the region with two municipalities (Beijing and Tianjin) and five provinces (Hebei, Shanxi, Henan, Shandong, and Inner Mongolia).

4. Table 4: This study can be seen as an improvement to the emission inventory, but it seems that the model simulates SOA better than POA. Please clarify this.
Response: We thank the reviewer for the good question. Indeed, organic CPM contributes to the primary emissions of OA in the emission inventory, but it is composed of organic matter which is semi-volatile (SVOCs, $10^0 \leqslant C^* \leqslant 10^3$ µg m^{-3}), or has intermediate volatility (IVOCs, $10^3 < C^* \leqslant 10^6$ µg m^{-3}), and it can go through the gas-particle partitioning and aging reactions to generate SOA. Therefore, the organic CPM can contribute to both POA and SOA. Based on better simulations of SOA than POA, it further suggests that CPM has a greater impact on SOA than POA.

5. Line 338-339: how to make the estimation of uncertainties?

Response: We thank the reviewer for the suggestion. The estimation of uncertainties related to variabilities in the ratio of E_{OM}(CPM) to $E_{PM2.5}$(FPM) was described in Sect. 2.3 as follows "We carried out bootstrapping and Monte Carlo simulations to obtain the mean and uncertainty ranges of E_{OM}(CPM)/$E_{PM2.5}$(FPM) for stationary combustion sources including power plant (PP), industry combustion (IN), steel (IR) (see Table 3). First, the optimal probabilistic distributions and uncertainty ranges were determined for each source category. Then the statistical bootstrap simulation was applied to calculate the mean and 95% confidence interval of emission ratios for each source category. Finally, the uncertainties of these parameters were propagated to calculate the total uncertainty of emission by running Monte Carlo simulations for 10,000 times. Notably, the estimated uncertainties were only related to variabilities in the ratio of $E_{OM(CPM)}$ to $E_{PM2.5}$(FPM), but did not necessarily represent the overall uncertainties of organic CPM emissions."

6. Line 82: the reference here should be Huang et al., 2014.

Response: We thank the reviewer for the suggestion. We have revised Huang et al. (2015) to Huang et al. (2014).

7. The distinction between SOA and POA based on OC/EC measurements is debatable. I recommended using some AMS data, published a lot during the last decades in the Beijing-Tianjin-Hebei region, to validate.

Response: We thank the reviewer for the suggestion. Actually, the observation data of OA in 2014 was measured by an Aerodyne high-resolution time-of-flight aerosol mass spectrometer (HR-AMS). Then three POA factors and two SOA factors were identified using positive matrix factorization (PMF) with the AMS measurement data. Therefore, the SOA and POA data were not based on OC/EC measurements, and can be used to validate the simulation results. This information has been added to the Sect. 2.4 Observational data.

Added/rewritten part in Sect. 2.4 Observational data: To distinguish between SOA and POA, Aerosol Mass Spectrometer (AMS) measurements and the method of Positive Matrix Factorization (PMF) used by Xu et al. (2015) identified three POA factors from coal combustion, biomass burning and cooking, and two SOA factors of semi-volatile and low-volatility oxygenated OA used in this study.