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 #1

Li et al. constructed an emission inventory for condensable particulate matter for China and evaluated its impact on the simulation of organic aerosol and $PM_{2.5}$. The study provides useful information on how CPM emissions (which are conventionally not measured in emission studies) affect ambient concentrations. However, I find the paper difficult to follow mainly because the method descriptions are not well organized. For example, the authors did not explicitly state their operational definitions for OA, POA, SOA, FPM, CPM, OMlsi, OM(C*<100), SVOC, and their relationships, which cause confusion. For example, I am confused about whether CPM emissions are accounted for as only primary emissions or also as secondary emissions in the emission inventory? And whether this inventory assumes that all CPM are organic? Whether E_OA include E_OMlsi (Eq.1-3) or do they represent non-overlapping components? Clearly describing what the authors actually did and meant would definitely help assess the scientific value of this study.

Response: We truly appreciate all the constructive comments and suggestions of the reviewer. We have adopted all the suggestions in our revised manuscript. We clearly recognized that the methods descriptions were not well organized and might cause confusions. Therefore, we have revised the descriptions in the Materials and methods section and also supplemented a table to explicitly state our operational definitions for OA, POA, SOA, FPM, CPM, OM_{lsi}, OM(C*<100), SVOC, and other used acronyms (see Table 1). The serial numbers of other tables have been adjusted. This will help readers better understand what we actually did and meant in this study.
For these points of confusions, our answers are as follows: CPM emissions are accounted for only primary emissions in the emission inventory; This study only concentrated on organic CPM emissions, so the constructed inventory only included the emissions of organic CPM; In Eq.1-3, E_OM_{si} included E_OM_{si}, and E_OM_{lsi} has been revised to E_OM_{si}.

Added/rewritten part in Sect. 2 Materials and methods: CPM contained both organic and inorganic fractions, but this study only concentrated on organic CPM emissions.
In addition, the component information of organic CPM is important to model the participation of organic CPM in atmospheric chemical reactions. The organic CPM mainly contains alkanes (with C_{10}-C_{30} being the major n-alkanes), esters, and polycyclic aromatic hydrocarbons (PAHs) (Li et al., 2017c, d; Song et al., 2020; Zheng et al., 2018). Based on the relationship between carbon number of n-alkanes and saturation concentrations (C^*) following Lu et al. (2018), it is reasonable to speculate that organic CPM 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}), combined as OM_{si} (CPM). It denotes a collective term for a range of organic matter with different volatilities in CPM. Since the volatility characteristics of organic CPM from these stationary combustion sources have not been accurately determined in relevant measurement studies, the emissions of OM_{si} (CPM) were scaled to emissions of OM (CPM) in this estimate as shown in Eq. (3), that is, the total emissions of OM (CPM) were distributed in different volatility bins. E_{OMsi} (CPM) denotes the emission rate of OM_{si} in CPM; C_{OMsi} (CPM) denotes the concentration of OM_{si} in CPM. The specific partition coefficients for different volatility bins in the model will be discussed in the following Sect. 2.3.

For the base scenarios, the simulations were performed with the inputs of the previous emission inventory without the newly constructed organic CPM emissions. Considering that organic FPM from stationary combustion and mobile sources mainly contained low volatile matter, so all of these emissions should be assigned to the CMAQ species of LVPO1 and other volatility bins should be assigned a scale factor of 0, and the rests were kept at the default settings in the model. In addition, different volatility distributions could be chosen for different emission sources, but this was not our study focus and did not interfere with the results of CPM contributions. For the cases including CPM emissions from stationary combustion and mobile sources, 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 oxidations of organic vapors by OH radicals to generate successively lower volatility and more-oxygenated species, and then condensed to produce SOA. Due to the unavailable volatility distribution information of OM_{si} (CPM), different scaling factors of volatility bins were employed under each emission scenario to discuss the uncertainties of CPM effects. In this study, we tested two kinds of scaling factors for the five volatility bins: fac1 (0.09, 0.09, 0.14, 0.18, 0.5) (Grieshop et al., 2009) and fac2 (0.40, 0.26, 0.40, 0.51, 1.43) (Shrivastava et al., 2011). As mentioned in Sect. 2.1, organic CPM was composed of organic matter which was semi-volatile or had intermediate volatility, thus the first bin which represents nonvolatile organic matter should be set to zero. Here, the original partition coefficient of the first bin was added to the following bin, so the fac1 (0, 0.18, 0.14, 0.18, 0.5) and fac2 (0, 0.66, 0.40, 0.51, 1.43) were applied in the sensitivity simulation cases. The fac2 estimated total SVOCs emissions as 3 times POA emissions to consider missing OM_{si} (CPM) emissions. Then the fac3 (0, 0.42, 0.27, 0.345, 0.965) which was the average of fac1 and fac2, was also tested for the five volatility bins.

Table 1 Definitions of some acronyms used in this study.

Acronyms	Definitions
FPM	Primary-emitted filterable particulate matter which is in liquid or solid phases in flue
CPM	Primary-emitted condensable particulate matter which is in gas phase at flue gas temperature but condenses or reacts in the ambient air to form solid or liquid PM
OM (CPM)	Organic matter measured in CPM
OM_{si} (CPM)	Organic matter in CPM which is semi-volatile (SVOCs, $10^0 \leq C^* \leq 10^3$ μg m^{-3}), or has intermediate volatility (IVOCs, $10^3 < C^* \leq 10^6$ μg m^{-3}) are combined as OM_{si} (CPM)
OM ($C^* \leq 100$)	Organic matter with the saturation concentrations (C^*) below 100 μg m^{-3}
SVOCs	Primary-emitted semi-volatile organic compounds
IVOCs	Primary-emitted intermediate-volatility organic compounds
S/IVOCs	SVOCs + IVOCs
POA	Atmospheric organic aerosols from primary-emitted organic matter or formed by condensation of organic vapors before photochemical reactions

SOA	Atmospheric secondary organic aerosols generated by photochemical reactions and condensation of organic vapors after photochemical reactions
ASOA	SOA generated by photochemical oxidations of anthropogenic volatile organic compounds
BSOA	SOA generated by photochemical oxidations of biogenic volatile organic compounds
SISOA	SOA generated by photochemical oxidations of primary S/IVOCs
OA	POA + SOA

Minor comments:

1. Line 76: ambiguous meaning of "negative impact". Change to "negative radiative forcing".
Response: We thank the reviewer for the suggestion. "a negative impact on radiative climate forcing, air quality and human health" has been changed to "a negative radiative forcing and adverse impacts on air quality and human health" in the revision.

2. Line 98: unclear what is "inapplicability of parameter localizations". Do you mean there is a lack of local emission factors?
Response: We thank the reviewer for the suggestion. "inapplicability of parameter localizations" means that the parameters which characterize SOA yields in models were not sufficiently localized, and not necessarily applied to all the regions. This has been changed to "lack of localized parameters" in the revised manuscript.

3. Line 109: "totally" -> "completely"

Response: We thank the reviewer for the suggestion. "totally" has been changed to "completely".

4. Line 134: the ambiguous expression "more than 50% of organic composition were measured in CPM". Please rephrase.
Response: We thank the reviewer for the suggestion. This has been rephrased to "CPM contained more than 50% organic components".

5. Line 148: "largely" -> "greatly" or "substantially"
Response: We thank the reviewer for the suggestion. "largely" has been changed to "greatly".

6. Table S3 should list the measurement methods that these studies used.
Response: We thank the reviewer for the suggestion. The measurement methods have been listed in the "test method" column of Table S3. The column name has been changed to "Measurement methods".

7. Line 172 & Line 188-193: Please explain what is the difference between OA(CPM) and OM_lsi(CPM). Do you consider OA(CPM)-OM_lsi(CPM) as POA(CPM)?
Response: We thank the reviewer for the suggestion. OA(CPM) denotes the primary organic matter measured in CPM. To differentiate organic CPM emissions from simulated OA concentrations, OA(CPM) has been changed to OM(CPM) as shown in the supplemented Table 1. OM_{lsi} (CPM) has been changed to OM_{si} (CPM) in the revised manuscript. OM_{si} (CPM) denotes a collective term for a range of organic matter with different volatility in CPM, containing organic matter which was semi-volatile (SVOC, $10^0 \le C^* \le 10^3$ μg m^{-3}), or had intermediate volatility (IVOC, $10^3 < C^* \le 10^6$ μg

m^{-3}). Thus OM (CPM) included OM_{si} (CPM). Since the volatility characteristics of organic CPM have not been accurately determined in relevant measurement studies, the emissions of OM_{si} (CPM) were scaled to OM (CPM) emissions in this estimate (see Eq.1-3), that is, the total emissions of OM (CPM) were distributed in different volatility bins.

8. Line 196: what is the relative importance of stationary combustion vs. vehicles?
Response: We thank the reviewer for the suggestion. As shown in Fig. 2-4, the emissions of organic CPM from stationary combustion sources and the contributions of CPM from these sources to atmospheric OA concentrations were much greater than that from vehicle sources.

9. Line 216-217 & 270-273: what are the bases of these scaling factors? I thought you already derived emissions based on Eq.1-3. Why do you need to scale with respect to POA emissions?
Response: We thank the reviewer for the suggestion. Although the POA emissions from CPM has been derived based on Eq.1-2, the emissions of organic matter with different volatility cannot be acquired on the basis of the current measurement results. Therefore, a list of scaling factors should be applied to POA emissions to calculate the emission rates of these organic matter, then to represent model species in CMAQv5.32. 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 condensed to produce SOA.

10. Line 318-319: C*<=100 or C*<=10? Also state the unit of C*.
Response: We thank the reviewer for the suggestion. It should be C*<=100. The unit of C* is μg m^{-3} and has been stated in the revised manuscript.

11. Line 322: The OA emissions reported here are for what geographical region?
Response: We thank the reviewer for the suggestion. The OA emissions reported here are for the whole China. It has been changed to "the emissions of OA over mainland China were 3664.6 Gg" in the revision.

12. Section 3.2. These evaluations are not relevant and are just distractive. I'd suggest removing the section or putting it in the supplementary material.
Response: We thank the reviewer for the suggestion. The Sect. 3.2 has been moved to the supplementary material.

13. Line 369-372. Use "episode" instead of "process".
Response: We thank the reviewer for the suggestion. "process" has been changed to "episode" in the revision.

14. Section 3.3. How do the observations distinguish POA and SOA? Their operational definition should be introduced, as sometimes it is not so straightforward to compare to simulations.
Response: We thank the reviewer for the suggestion. We have supplemented the operational definition to distinguish between POA and SOA in Sect. 2.4.

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) were used by Xu et al. (2015), identifying three POA factors from coal combustion, biomass burning and cooking, and two SOA factors of semi-volatile and low-volatility oxygenated OA.

15. Line 461-462: Why do you think it is more likely due to meteorological factors, rather than that your emissions are still underestimated and that there are still missing SOA pathways?

Response: We thank the reviewer for the good suggestion. Indeed, we ignored the influence of other factors. This has been changed to "the underestimation of our estimated CPM emissions, effects of meteorological factors and other missing SOA formation pathways" in the revision.

16. Figure 1. Explain in the caption what the color shading represents

Response: We thank the reviewer for the suggestion. The color shading represents the regional altitude. We have supplemented the explanation in Fig.1 and caption in the revision.

Added/rewritten part in Figure 1: Figure 1. (a) Map of the modeling domain and location of each target city in model evaluation. (b) The locations of BTH2+26 cities, denoted as the red frame in (a). The color shading represents the regional altitude.