Response to RC#2:

Dear Editor and anonymous referee #1:

We greatly appreciate your consideration and the reviewer's constructive comments on the manuscript of "Estimation of Secondary $PM_{2.5}$ in China and the United States using a Multi-Tracer Approach" (acp-2021-683). We have carefully revised the manuscript to address all the comments as described below. Reviewer comments are shown in blue. Our responses are shown in black. The revised texts are shown in italics.

The manuscript demonstrates the multi-tracer estimation algorithm (MTEA), to identify the primary and secondary components from routine observation of $PM_{2.5}$ and validates the method by comparing the long-term and short-term measurements of aerosol chemical composition in China and a network from the United States. This method provides a useful and uncomplicated way to estimate primary and secondary PM, using routine observation species and emission inventories. This manuscript aims to address important questions quantifying primary and secondary aerosols and is within the scope of ACP.

However, regarding the method itself, the method should be carefully introduced with more details. The validation part is a bit weak and should be strengthened in the next version. It is vital because only with good validation can one trust the result from the model. In addition, in the result and discussion part, the discussion is superficial, which needs to be improved in depth, and backed up by more scientific evidence and/or publications.

As a conclusion, the manuscript provides a novel algorithm in primary and secondary particle concentrations, however, the manuscript is not carefully written from the perspective of science and scientific writing, with certain degree of improvement for publication in ACP. Therefore, this manuscript needs a major revision in terms of major context and English language.

Response: We thank the reviewer for the comments. According to the reviewer's helpful and insightful comments, we have revised our manuscript and the point-by-point responses to the specific comments were given subsequently. We sincerely hope these revisions are able to address the reviewer's concerns.

1. Introduction: the introduction is poorly written and need to be re-write. If I were you, I would write the introduction based on this outline: 1) introduction of atmospheric aerosols, including sources, type, chemical composition and impacts on air quality, human health and climate, 2) summaries other studies, you must state what has been achieved and what is the current challenging, 3) what is your paper about, how this paper can narrow the gap.

In the current version, the point 1) is addressed, but should be introduced in smoother way. The author is trying to address the point 2), but the studies mentioned in the paragraph 3 in page 3 look not very relevant. For example, the author summarizes the online and offline studies, which is good, and people can see the drawbacks of field and lab measurement to study the PPM and SPM, so the next paragraph should state to overcome these drawbacks, people use model to study the PPM and SPM, and should also state what these model studies have achieved and/or the drawbacks of these method. Finally, this paragraph can lead the final paragraph in the introduction, namely, introduce this study and how this study advances the model studies on PPM and SPM estimation.

Response: Thanks for your constructive suggestions and rigorous attitude to scientific research. We do think it is necessary and important to rephrase the structure of this part. Following the suggestion, we have rewritten the introduction section. The detailed description of this part has been corrected in the revised manuscript as follows.

Revision in Section 1:

Fine particulate matter ($PM_{2.5}$, aerodynamic diameter less than 2.5 μm) can be categorized into primary and secondary $PM_{2.5}$ according to its formation processes.

Primary $PM_{2.5}$ (PPM), including primary organic aerosol (POA), elemental carbon (EC), sea salt and mineral dust, is the product of direct emission from combustion of fossil/biomass fuel, dust blowing and sea spray. Secondary $PM_{2.5}$ (SPM) mainly generates from the further oxidation of gaseous precursors emitted by anthropogenic and biogenic activities (Zhu et al., 2018; Wang et al., 2019). SPM consists of secondary organic aerosol (SOA) and secondary inorganic aerosol (SIA, including sulfate, nitrate and ammonium). The primary and secondary components of $PM_{2.5}$ have different environmental impacts on air quality, human health and climate change. For example, as a typical PPM, EC can severely reduce atmospheric visibility and greatly influence weather and climate due to its strong absorption of solar radiation (Bond et al., 2013; IPCC, 2013; Mao et al., 2017). Sulfate, a critical hygroscopic component of secondary $PM_{2.5}$ (SPM), can be fast formed under high relative humidity conditions and further leads to grievous air pollution (Cheng et al., 2016; Guo et al., 2014; Quan et al., 2015). Furthermore, the sulfate and other hygroscopic $PM_{2.5}$ have considerable influences on climate change mostly by changing cloud properties (Leng et al., 2013; von Schneidemesser et al., 2015). In addition, different $PM_{2.5}$ components also have various deleterious impacts on human health for their toxicities (Hu et al., 2017; Khan et al., 2016; Maji et al., 2018).

To understand the severe $PM_{2.5}$ pollution characteristics in China over the past several years (An et al., 2019; Song et al., 2017; Yang et al., 2016), many observational studies have been conducted on $PM_{2.5}$ components. The basic methods of these studies are offline laboratory analysis and online instrument measurement such as aerosol mass spectrometer (AMS). The observational studies are crucial to exactly identify the aerosol chemical compositions. For offline approach, it is the most widely used method (Ming et al., 2017; Tang et al., 2017; Tao et al., 2017; Dai et al., 2018; Gao et al., 2018; Liu et al., 2018a; Wang et al., 2018; Zhang et al., 2018; Xu et al., 2019; Yu et al., 2019) and is successfully applied to investigate the inter-annual variations of different aerosol chemical species (Ding et al., 2019; Liu et al., 2018b). In terms of online approach, AMS is the state-of-the-art method for analyzing different chemical species with high time resolution, which has great application value in diagnosing the causes of haze events in China over the past decade (Huang et al., 2014; Quan et al., 2015; Guo et al., 2014; Yang et al., 2021; Gao et al., 2021; Hu et al., 2021; Zhang et al., 2022).

Nevertheless, both the online and offline measurements require a high level of

manpower and economic cost, and for this reason, these methods are expensive and rarely applied in large-scale regions or long-term periods.

Chemical transport model (CTM) is another useful tool to identify the composition characteristics of $PM_{2.5}$. The simulation predicted by CTM is featured as high spatio-temporal resolution (Geng et al., 2021). Meanwhile, it also provides vertical profiles of diverse chemical species (Ding et al., 2016). However, the CTM results are largely dependent on external inputs such as emission inventories, boundary conditions, initial conditions, etc. The internal parameterizations of itself significantly influence the final model results as well (Huang et al., 2021), which leads to uncertainty in the simulated $PM_{2.5}$ and its composition. In addition, the burden of high requirement in computational cost and storage also makes CTM hard to universally use.

In this study, we develop a novel method, Multi-Tracer Estimation Algorithm (MTEA), with the aim of distinguishing the primary and secondary compositions of $PM_{2.5}$ from routine observation of $PM_{2.5}$ concentration. Different from traditional CTMs, MTEA proposed by this study is based on statistical assumption and works in a more convenient way. This algorithm and its application are tested in China and the United States. In Section 2, we introduce the structure and principle of MTEA. In Section 3, we evaluate the MTEA results comparing with three $PM_{2.5}$ composition data sets, (1) short-term measurements in 16 cities in China from 2012 to 2016 reported by previous studies, (2) continuous long-term measurements in Beijing and Shanghai from 2014 to 2018, and (3) IMPROVE network in the United States during 2014 and 2018. Additionally, we also compare MTEA model with one of the most advanced datasets from CTM in China. Subsequently, in Section 4 we investigate the spatio-temporal characteristics of PPM and SPM concentrations in China, explain the unexpected haze event in several cities of China during the COVID-19 lockdown and discuss the complicated correlation between PM and O_3. This study is different from previous works as follows: (1) we develop an efficient approach to explore PPM and SPM with low economy-/technique-cost and computation burden, (2) we apply this approach to observation data from the MEE network, offering an unprecedented opportunity to quantify the $PM_{2.5}$ components on a large space and time scale.

2. Methodology: the methodology part is written in a reasonable logic, but the author needs to pay more attention to specify the technical details, e.g., the definition of some terms.

Response: Thanks for your kind reminding and rigorous attitude to scientific research. We have carefully checked all technical details and revised them for a more proper expression in Section 2.

Revision in Section 2:

The multi-tracer (marked as X) is defined to represent multiple primary contributions to $PM_{2.5}$, mainly resulting from incomplete combustion of carbonaceous material and flying dust.

We select the typical combustion product CO as one tracer to represent the combustion process, and the particles in coarse mode (PM_{coarse}, marked as PMC, PMC = $PM_{10} - PM_{2.5}$) as the other tracer to track flying dust.

However, this investigative coefficient for quantifying primary sulfate and nitrate emissions might be relatively higher compared to empirical coefficients (0.01-0.05) used in previous simulation studies.

They estimated primary and secondary organic carbon (marked as POC and SOC) concentrations by adopting a proper POC/EC ratio when SOC correlated with EC worst.

3. Model validation: this part straightforwardly delivers the good validation result between model and observation. Good correlation is shown in this part, suggesting good model performance. However, this part also requires more interpretation on the model's over/underestimation behavior compared to observation, which is now absent. Ideally, the author should focus most on this part, because only when the model is reasonable validated can we trust the result and make the further interpretation on the result.

Therefore, from my own perspective, the author should strengthen this part.

Response: Thanks for your conducive comment. We have enhanced the discussion in the model evaluation part as you suggested.

Revision in Section 3.1.2:

However, we find that there are still a few discrepancies between the estimated and observation-based results. For example, we overestimated the secondary proportions of $PM_{2.5}$ in cities such as Haikou, Lanzhou and Lhasa. Though all of them show a considerable overestimation of over 20%, the causes lead to this kind of bias may be quite different. In coastal city Haikou, we may attribute this discrepancy between MTEA and observation to the neglect of the contribution of sea salt aerosols. The $PM_{2.5}$ offline measurements in 2015 exhibited that the contribution of sea salt aerosols to ambient $PM_{2.5}$ mass concentration in Haikou is 3.6-8.3% (Liu et al., 2017). Secondly, the overestimation phenomenon in Lanzhou, which is a typical inland city located in northwestern China, can be explained by overlooking the contribution of natural dust to $PM_{2.5}$ speciation. Generally, both sea salt and natural dust are categorized into non-anthropogenic processes, and are not accounted for by anthropogenic emission inventory, resulting in the underestimation of representing primary process intensity. Finally, for Lhasa, the observation-based results which are derived from too few samplers also pose controversial comparison against MTEA model.

4. Result and discussion: this part also very straightforwardly and logically reports the results. However, the interpretation of results should be more comprehensive and backed up by previous studies and/or solid evidence, which is absent now and needs to be added. In addition, the discussion of the result is very superficial, lacking depths, which should also be improved.

Response: Thanks for your conducive comments and rigorous attitude to scientific research. To enrich our discussion as the reviewer mentioned, we have carefully revised the related texts in the result part.

Revision in Section 4:

We used the MTEA approach and the MEE observation data to estimate PPM and SPM concentrations in China for the period of 2014-2018. The observations during severe haze events (top 10% CO and PMC polluted days) were excluded to avoid the influence of unfavorable meteorological conditions and extreme high primary emission cases. Unfavorable meteorological conditions are major causes for haze events. PPM under these unfavored meteorological conditions may have considerable high co-linear relationship with total $PM_{2.5}$. The concentration of SPM from complicated formation pathways is then underestimated. Therefore, we excluded these polluted days to focus more attention on general characteristics of PPM and SPM concentration.

Revision in Section 4.3:

To explore this unexpected air pollution, we find that the enhanced secondary pollution could be the major factor, which even offset the reduction of primary emissions in the BTH region during the lockdown. With the help of MTEA, we tracked variations of the secondary proportions of $PM_{2.5}$ in East China before and during the COVID-19 lockdown (Fig. 9 d-f). The specific emission reductions owing to the national lockdown were derived from Huang et al. (2020). Based on the bottom-up dynamic estimation, provincial emissions of CO, NO_x, SO_2, VOC, $PM_{2.5}$, BC and OC decreased by 13-41%, 29-57%, 15-42%, 28-46%, 9-34%, 13-54%, and 3-42%, respectively during the lockdown period. The secondary proportions in the BTH region show an evident increase, at the level of 7%-34%, which highlights the importance of the secondary formation during the lockdown. Our result is consistent with recent observation and simulation studies (Chang et al., 2020; Huang et al., 2020; Le et al., 2020), which suggested that the reduced NO_2 resulted in O_3 enhancement, further increasing the AOC and facilitating the formation of secondary inorganic aerosols such as ammonium sulfate, ammonium nitrate. In addition, another cause of the air pollution is the unfavorable atmospheric diffusion conditions. CO, a nonreactive pollutant, was increased by 22% in Beijing during the lockdown even under considerable reduction on its emission.

Revision in Section 4.4:

A series of recent studies have focused on the correlation between $PM_{2.5}$ and O_3, and many of them agreed that the correlation varies greatly in different regions of China. Specifically, the statistical correlation is stronger positive in southern cities compared to that in northern cities (Chu et al., 2020). Because of this significant difference, a question raises: is the difference mostly caused by PPM, or SPM, or both of them? To address this question, we compare the correlations between daily PPM, SPM and total $PM_{2.5}$ versus O_3 in Beijing-Tianjin-Hebei (BTH) and Yangtze River Delta (YRD) region during the study period, with the help of META approach. The O_3 diurnal formation regime can be destroyed because of the suppressed radiative condition under precipitation. The local O_3 concentration level is mainly dominated by background fields. Here we would like to focus our attention on the secondary formation relationship between daily $PM_{2.5}$ and O_3. Therefore the cases when precipitation took place were removed to avoid the cleaning impacts of wet deposition on MDA8 (maximum daily 8-h average) O_3 concentrations. Precipitation data is based on the ERA5 reanalysis database from the European Centre for Medium-Range Weather Forecasts (ECMWF, https://www.ecmwf.int/, last access, 1 August 2021).

Revision in Section 4.5:

Thirdly, current bottom-up emission inventories are generally outdated with a time lag of at least 1-2 years, mainly due to the lack of timely and accurate statistics. Consequently, the adjoint uncertainty in MTEA estimation is inevitable.

To evaluate the uncertainty, a comparison test was conducted by adjusting the apportioning coefficient (the a and b in Eq. 1) with a disturbance of ±0.1. Firstly, we decreased the value of a in each populous city by 0.1. Meanwhile, the coefficient b increased by 0.1. This scenario indicates an overestimation in contribution of combustion-related process to primary $PM_{2.5}$ or underestimation in contribution of dust-related process. Secondly, we increased the value of a in each populous city by 0.1 (decreased b by 0.1) for checking the opposite case. The results are presented in Table

S5 and point out that the estimated secondary proportions of $PM_{2.5}$ varied less than ±3% in most populous cities caused by the changes of the apportioning coefficient. This sensitivity experiment highlights that the apportioning coefficients depending on emissions has limited impacts on the final estimation results. Generally, the uncertainty of apportioning coefficient is one of two factors that directly affect the tracer X. The other one is the concentration of CO and PMC itself. Hence, we also conducted a similar test to check the impacts of tracer X on the model estimation by changing the tracer concentrations mentioned in Eq.1. Specifically, we (1) increased CO concentration by 10% as well as decreased PMC concentration by 10% and (2) decreased CO concentration by 10% as well as increased PMC concentration by 10%. Both sets of adjustment show changes within ±2% in the estimated secondary proportions of $PM_{2.5}$ in all cities except for Urumqi (Table S6). This phenomenon from the perspective of tracer concentration also supports that the impacts of the tracer X on the final model results are limited. In summary, we believe that the most determinative stuff for the final results of our model is the principle of the minimum correlation between PPM and SPM but not the tracer X which relies on emissions or concentrations.

5. Conclusion: it summarizes the significance of the study, but one or two paragraph need to be re-written, based on the revised context in Section 4.

Response: Thank you for your comments and we have added the related texts to the manuscript.

Revision in Section 5:

We also discussed the uncertainties of the MTEA method. MTEA may pose overestimation on the secondary fractions of $PM_{2.5}$ in those regions which are near to desert or sea by ~20% for failing taking natural dust into consideration. In addition, the sensitivity experiment through imposing reasonable disturbance on emissions and tracer concentrations also show the limited impacts on final estimation. Overall, the

most determinative stuff for our model estimate is the principle of the minimum correlation between PPM and SPM.

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