Response to RC#1:

Dear Editor and anonymous referee #3:

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.

This study developed a new method to determine the portion of primary and secondary $PM_{2.5}$ using some basic measurements and inventory. They evaluated this new approach through the comparison with lots of observations in China and US. In addition, they analyzed the temporal and spatial variation as well as correlation between O_3 and $PM_{2.5}$ using the results from their new method. Although their evaluation looks very well, I think their results were not enough convincing because of unclear statement of their method and defect of this method. I would suggest major revision before reconsideration. My detail comments are following.

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 the revisions are able to address the reviewer's concerns.

1. Eq (1) and Eq (2): These equations are the core of their method. They regarded CO as one tracer to represent the combustion process and assumed the combustion emission sources are same for CO, OC and EC. This assumption is mostly correct, but the emission factor/emission ratio of CO, OC and EC from different combustion sources are different. I think it is unconvincing to use one single coefficient without

the influence of diversity of sources to standard for all conditions. I may misunderstand something, please discuss this uncertainty or make this clear.

Response: Thanks for the conducive comments. We also do believe that the emission factors of CO, OC and EC from different sources are various as well. Our method tracks the combustion process, which produces OC and EC, by regarding CO as the tracer. However, the correlation between different sources of diverse carbonaceous matter is hard to find out with the aid of current routine observations of CO. The coefficients in Eq. 1 are aimed at representing the relative contribution of combustion process and flying dust to primary $PM_{2.5}$. We constrained the uncertainty of both coefficients by setting up a set of sensitivity tests. The specific discussion about this uncertainty is in Section 4.5. The specific configuration issue your concerned about the sensitivity experiment will be clarified in the following 3rd point. The final experiment result indicates that the adjustment of coefficient for CO (*a*) within 0.1 does not obviously affect the estimated secondary proportions of $PM_{2.5}$ (< 3%). To make this point clearer, the detailed description of this part has been corrected in the revised manuscript as follows.

Revision in Section 2.1:

As shown in Eq. 1, we use a and b to quantify the relative contributions of combustion and dust process to PPM. Given that the complicated process such as the combustion from multiple sources is hard to represent via current routine CO observations, we avoid considering the correlation among these sources but focus on the relative weights of combustion process and flying dust. Meanwhile, the uncertainty resulting from the apportioning coefficient a and b will be further discussed in Section 4.5.

2. Eq (2): why did you name b as emission of fine dust? To my knowledge, MEIC does not include the emission of dust even urban dust.

Response: Thanks for your concerns. The dust emissions are not specifically

separated from $PM_{2.5}$ emissions in MEIC. In fact, the composition of $PM_{2.5}$ emission in MEIC includes EC, OM, sulfate, nitrate and other trace elements such as Al, Ca, Si, Fe, Mg, K and other species etc. (Li et al., 2017a). Trace elements are usually related to the flying dust from constructions and onroad traffic transportation. In the MTEA approach, we would like to represent the dust-related part of PPM with the emissions of the mineral dust in fine mode particulate matter. We calculated the dust-related emissions by deducting the emissions of EC, OM, sulfate and nitrate from total $PM_{2.5}$ emissions. We revised the relevant texts for a clearer statement.

Revision in Section 2.1:

Coefficient b is aimed at reflecting the activity intensity of fine mode dust by counting its emissions. However, MEIC does not directly provide fine mode dust emissions. It is included in the emissions of total $PM_{2.5}$ (Li et al., 2017a). Thus we inferred the fine mode dust emission by deducting the emissions of EC, POA, sulfate and nitrate from the $PM_{2.5}$ emissions.

3. I did not understand how you did the sensitivity experiment to examine the uncertainty in the inventories. Page 16, you said you changed the emission coefficient with 10%. If so, how can you keep a+b=100%? According to my understanding on this new method, the results should have large dependence on the inventory of $PM_{2.5}$, OC, EC even the factor you used to decide OA, SO_4^{2-} and NO_3^-. I would strongly suggest setting up more comprehensive and scientific sensitivity experiments to discuss the dependence on the inventory.

Response: Thank you for your conducive comments and rigorous attitude to scientific research. Coefficients *a* and *b* are determined by calculating the relative ratio between EC+POA to dust as Eq. 1-2. Hence the uncertainty of emission inventory can lead to the changes of the ratio *a* to *b*. In Section 4.5, we tested the adjoint changes of the final estimated secondary proportions of $PM_{2.5}$ by adjusting the coefficient *a*. The adjustive test includes two parts. Firstly, we increased the value of *a*

in each city by 0.1 to check the model results in the case of underestimating the contributions of combustion process (or overestimating the contributions of dust process). Under this circumstance, the coefficient *b* which represents dust process should be decreased by 0.1. On the contrary, we also decreased the value of *a* in each city by 0.1 to check the model results in the case of overestimating the contributions of combustion process (or underestimating the contributions of dust process). Meanwhile, the coefficient *b* which stands for dust process is increased by 0.1. The sum of *a* and *b* is still 100%. The sensitivity experiment results indicate that the disturbance of coefficient *a* (±0.1) lead to changes in the secondary proportions of $PM_{2.5}$ within ±3% (refer to Table S5 in the supplementary material). In addition, the discussion about the uncertainty of the primary sulfate and nitrate emissions also reveals that the predicted results are not sensitive to their emissions (refer to Section 2.1 and Table S1 in the supplementary material). Therefore, we indeed agree that the emission inventory can pose impacts on our model estimation, but the effects are not obvious.

The assumed tracer of PPM (i.e. X, see Eq. 1) is one of the cores of MTEA approach. However, the most determinative stuff for the final results of our model is the principle of the minimum correlation between PPM and SPM but not only the value of the tracer X. To prove this view, we also carried out another kind of test in adjusting X by changing the concentrations of CO and PMC. 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 demonstrate changes within ±2% in the estimated secondary proportions of $PM_{2.5}$ in all cities except for Urumqi (Table R1). This phenomenon also supports that the impacts of the tracer X on the final model results are not obvious. To clearly state the point mentioned by the reviewer, we have rephrased the relevant texts in the manuscript.

Revision in Section 2.1:

We evaluated the potential effect of the coefficient, by conducting a set of

comparative simulation with the coefficient of 0.03, and found that the final estimated SPM was not sensitive to this coefficient (Table S1). Thus we concluded that the uncertainty of primary sulfate and nitrate emissions did not significantly influence the final estimation of MTEA model. For other uncertainties of X which are dependent on emission intensities or tracer concentrations, we would conduct discussions in the later Section 4.5.

Revision in Section 4.5:

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

142 *correlation between PPM and SPM but not the tracer X which relies on emissions or*
143 *concentrations.*

144 **Table R1.** Impacts of tracer concentration uncertainty on the estimated secondary proportion of
145 $PM_{2.5}$ [1] in China (Unit: %).

City	Secondary proportion of $PM_{2.5}$	Change of secondary proportion of $PM_{2.5}$	
		1.1 * CO concentration & 0.9 * PMC concentration	0.9 * CO concentration & 1.1 * PMC concentration
Beijing	40.3	-0.01	0.01
Tianjin	61.9	-0.32	-0.52
Shijiazhuang	44.8	-0.26	-0.28
Taiyuan	43.1	0.22	0.17
Hohhot	48.6	-0.03	-0.01
Shenyang	48.7	-0.06	-0.06
Changchun	47.9	0.03	0.04
Harbin	66.9	0.22	-0.59
Shanghai	68.0	-1.51	-1.90
Nanjing	50.3	0.00	0.03
Hangzhou	45.6	-0.42	-0.46
Hefei	65.4	-1.57	-1.73
Fuzhou	64.8	-0.25	-0.44
Nanchang	62.5	-0.33	-0.42
Ji'nan	54.6	-0.04	-0.02
Zhengzhou	54.6	0.14	0.14
Wuhan	61.5	-1.45	-1.49
Changsha	65.9	-1.60	-1.74
Guangzhou	65.2	0.00	-0.28
Nanning	65.2	-0.22	-0.47
Haikou	65.9	-0.15	-0.09
Chongqing	62.7	-0.23	-0.31
Chengdu	45.3	0.42	0.44
Guiyang	65.6	-0.22	-0.50
Kunming	70.4	-0.40	-0.69
Lhasa	56.1	0.07	0.05
Xi'an	52.6	-0.04	-0.01
Lanzhou	60.0	0.15	0.02
Xining	59.1	-0.56	-0.60

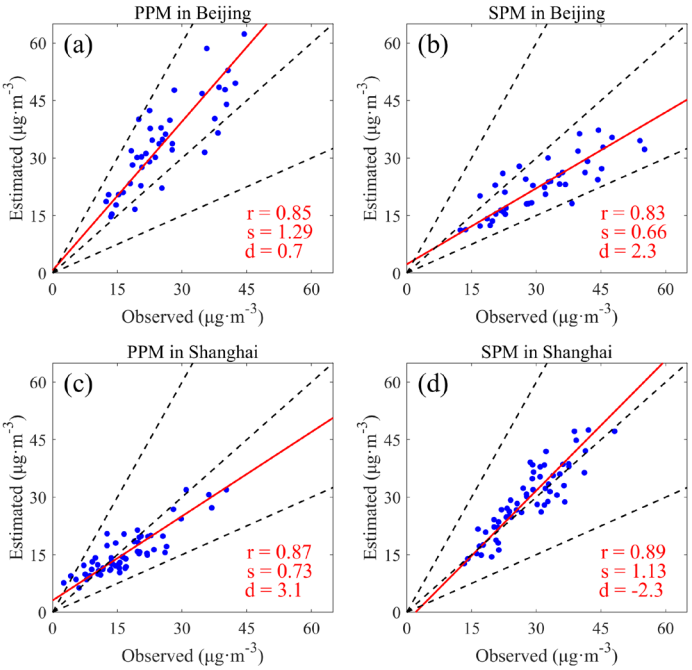
Yinchuan	59.5	0.02	-0.06
Urumqi	72.1	-2.70	-2.85

[1] Based on the MEE observations in 2016.

4. Figure 3, as I saw, the largest concentration is < 60 μg/m3. Why not short the range of axis to spread those dots?

Response: Thanks for your highly careful reminding. We have reduced the range of axis from 130 to 65 for aesthetics. And the revised figure is shown below.

Revision in Fig. 3:



5. P8L7: Why did you remove the heavy pollution cases here as well as in Section 4? As you stated at P10L25, you would like to avoid the influence of extreme high primary emission cases. However, mostly heavy pollution cases are caused by unfavored meteorological condition but not caused by sudden high primary emission (except the biomass burning cases). I would be curious that how your method applied to analyze the heavy pollution cases. In general, it is more important to understand the contribution of secondary particles to heavy pollution cases than the general

conditions.

Response: Thanks for your highly conducive comments and rigorous attitude to scientific research. The data preprocessing in P8L7 and P10L25 are different. The data preprocessing in Section 3.1.1 is aimed at removing the gap between long-term measurements of $PM_{2.5}$ at a single site and routine observation of $PM_{2.5}$ from national network for further evaluation.

However, the data preprocessing in Section 4 is prepared for the usage of data from MEE. To address reviewer's concern, we take estimation in 2016 as an example and make a comparison. MTEA method shows that the estimated secondary proportions of $PM_{2.5}$ without excluding the heavy polluted cases are 2.0-13.7% lower than that including the data preprocessing (Fig. R1). We agree with the reviewer that unfavorable meteorological conditions are major causes for haze events. Under these unfavored meteorological conditions, the assumed tracer X may have extremely high co-linear relationship with total $PM_{2.5}$. Thus the PPM concentrations may be falsely overestimated. Here we excluded these days to avoid the incorrectly estimation and focus more attention on the common characteristics of PPM/SPM during the general periods. We revised the statement in Section 3.1.1 and Section 4 for a clearer version.

Revision in Section 3.1.1:

Given the discrepancy in $PM_{2.5}$ concentrations between in-situ measurements of a single site and multiple MEE national sites, we firstly preprocessed the data for further evaluation. In data preprocessing, we removed the in-situ daily measurements whose value was over 30 $\mu g \cdot m^{-3}$ higher than the city average (from MEE).

Revision in Section 4:

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.

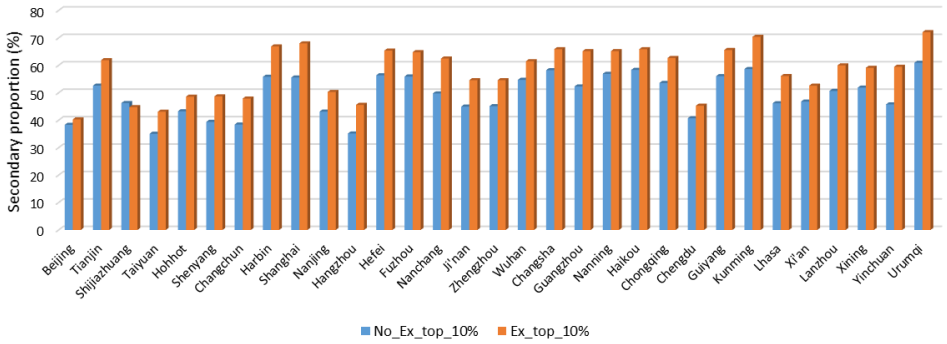


Figure R1. The estimated secondary proportions of $PM_{2.5}$ in case of including (No_Ex_top_10%) and excluding top 10% polluted days (Ex_top_10%) in 2016.

6. P10L30: Could you explain what is regional background cities you defined here? Usually, cities are not background.

Response: Thank you for pointing this out. We agree that cities usually are not categorized as background regions. We are aimed at disclosing the discrepancy in PPM/SPM among diverse cities which depend on different levels of anthropogenic activity. The 19 regional background cities in this study are chosen because they suffered the least $PM_{2.5}$ pollution during 2014-2018. The averaged mean $PM_{2.5}$ concentration of each city is less than 35.0 μg m^{-3} (National Ambient Air Quality Standard level II of China, NAAQS) except for Guyuan, Ningxia Province (refer to Table S3 in the supplementary material). We believe that these selected cities can generally reveal the PM pollution characteristics of the regions which are under sparse anthropogenic emissions. For a clearer expression, we have revised the related texts in the manuscript.

Revision in Section 2.2.1: *31 among the 50 cities are provincial capital cities, employed to represent populous cities, while the rest 19 relatively small cities are categorized as regional background cities (Table S3). The mean $PM_{2.5}$ concentration*

of each regional background city is less than 35 μg m^{-3} (National Ambient Air Quality Standard level II of China, NAAQS) except for Guyuan, indicating that they are slightly impacted by anthropogenic activities. By comparing populous cities with regional background cities, we could reveal the discrepancy in PPM and SPM among those regions which suffer from different levels of $PM_{2.5}$ pollution.

7. Section 4.2.1: I think the seasonal variation of PPM and SPM is largely depend on the seasonal variation of emissions you applied.

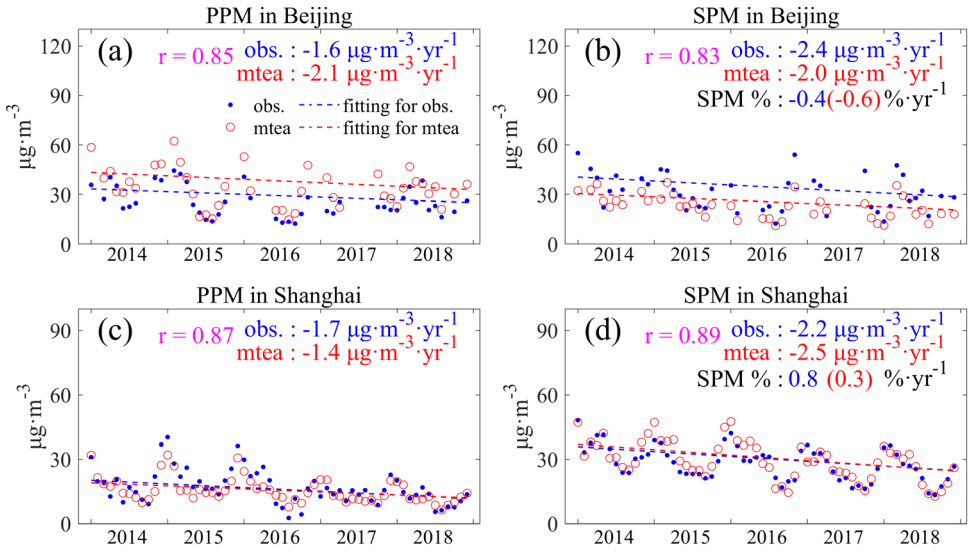
Response: Thank you for your comments. We indeed agree with the reviewer that the seasonal pattern of the estimated PPM and SPM concentration can be attributed to the seasonal variations of emissions. Taking Shanghai as an example, we tested the impacts of the seasonal variations of emissions on the estimated PPM and SPM concentrations by comparing two cases (i.e. seasonal emissions in this study and homogenous emissions in the ideal sensitivity experiment). As listed in Table R2, though the seasonal maxima/minima of PPM and SPM concentration still happen in the wintertime/summertime, but the specific concentrations significantly change. The maximum of relative change can be 10% (PPM in DJF, changes from 15.8 μg·m^{-3} to 14.3 μg·m^{-3}).

Table R2. Comparison of seasonal PPM and SPM concentrations between applying seasonal emissions or homogenous emissions in Shanghai (Unit: μg·m^{-3}).

		MAM	JJA	SON	DJF
PPM	Seasonal emissions (This study)	12.4	11.1	11.7	15.8
	Homogenous emissions (Ideal study)	12.8	11.7	12.2	14.3
SPM	Seasonal emissions (This study)	29.5	22.5	20.8	25.4
	Homogenous emissions (Ideal study)	29.2	21.9	20.3	26.8

8. Section 4.2.2: Did you use the emission inventory for specific year here? China conducted a large reduction on $PM_{2.5}$ emission since 2014. If you did not use the specific inventory, the estimated trend of PPM and SPM would not make sense, even though they agreed with observations. In addition, could you show the correlation coefficient between the observation and estimation here?

Response: Thanks for your concern. We indeed agree with the reviewer's opinion that the emission inventory should be matched for each year. For anthropogenic emissions from 2014 to 2017, we utilized the MEIC emission inventory (v1.3) developed by Tsinghua University, which is publicly offered at their website (http://meicmodel.org/) (Li et al., 2017a; Li et al., 2017b). In terms of emissions after 2017, we also accessed from MEIC support team (Zheng et al., 2021). For the correlation coefficient between the observation and estimation in Section 4.2.2, we have followed the suggestion from the reviewer and showed it both in the related figure and the related texts in the manuscript.



Revision in Section 4.2.2:

Applying the MTEA model to this case, we are delighted to find that our model not only successfully reproduces the consistent decreasing trends of PPM and SPM in Beijing and Shanghai (correlation coefficient r of observation versus estimation ranges from 0.83 to 0.89), but also captures the different trends in secondary

proportions of $PM_{2.5}$ in the two cities (–0.6% yr^{-1} in Beijing and 0.3% yr^{-1} in Shanghai).

9. Section 4.3: The same issue as above. Did you update the inventory to the lockdown condition? If yes, please state the inventory you used here and the decrease in the emission of $PM_{2.5}$, CO, OC, EC.

Response: Thank you for your concern. We used the emission reduction ratio in of various air pollutants during the COVID-19 lockdown from Huang et al. (2020). The specific emission reduction ratios of various air pollutants are listed in Table R3. Meanwhile, we modified the related texts in the manuscript to make it clearer.

Revision in Section 4.3: *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. 8 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.*

Table R3. Estimation of provincial emission reduction ratio (%) of CO, NO_x, SO_2, VOC, $PM_{2.5}$, BC, OC due to COVID-19 lockdown in China.

Province	CO	NO_x	SO_2	VOC	$PM_{2.5}$	BC	OC
Beijing	22%	45%	26%	45%	18%	46%	8%
Tianjin	21%	38%	20%	41%	14%	22%	6%
Hebei	15%	45%	16%	36%	12%	17%	5%
Shanxi	18%	40%	20%	33%	16%	19%	10%
Inner Mongolia	14%	29%	15%	34%	13%	16%	6%
Liaoning	21%	40%	28%	36%	16%	28%	8%
Jilin	16%	39%	23%	34%	13%	18%	5%
Heilongjiang	17%	37%	27%	28%	13%	15%	7%
Shanghai	35%	48%	42%	45%	34%	54%	42%
Jiangsu	23%	50%	26%	41%	16%	35%	7%

Zhejiang	41%	50%	29%	45%	30%	49%	20%
Anhui	14%	56%	22%	31%	11%	22%	4%
Fujian	29%	51%	30%	42%	19%	31%	7%
Jiangxi	24%	53%	21%	43%	19%	30%	9%
Shandong	23%	50%	25%	39%	19%	35%	9%
Henan	23%	57%	22%	41%	18%	35%	8%
Hubei	19%	55%	23%	35%	16%	23%	10%
Hunan	22%	51%	25%	36%	20%	24%	15%
Guangdong	38%	50%	33%	46%	27%	42%	13%
Guangxi	24%	50%	28%	39%	17%	27%	5%
Hainan	24%	44%	25%	36%	14%	25%	4%
Chongqing	18%	53%	32%	37%	14%	20%	4%
Sichuan	16%	50%	27%	33%	9%	15%	3%
Guizhou	24%	39%	25%	30%	22%	25%	20%
Yunnan	24%	51%	25%	41%	18%	21%	8%
Tibet	16%	35%	15%	35%	14%	14%	5%
Shaanxi	19%	45%	18%	34%	13%	22%	5%
Gansu	13%	47%	16%	29%	9%	13%	3%
Qinghai	23%	46%	22%	39%	20%	20%	7%
Ningxia	24%	36%	24%	39%	20%	23%	8%
Xinjiang	16%	35%	15%	35%	14%	14%	5%

10. Section 4.4: How did you decide the diurnal variation of emission? Was your result sensitive to the diurnal pattern? Because the diurnal pattern of O_3 concentration is almost constant.

Response: Thank you for your careful concerns. MEIC provides the bottom-up anthropogenic emission inventory with monthly time resolution. Based on the fixed total emissions, we further distributed them with specific diurnal variation patterns of each sector, including power plants, industry, transportation and residential sources (Fig. R3a). This kind of preprocessing is also adopted for preparing emission input for other air quality model studies and is proved to be reasonable (Li et al., 2021; Zhang et al., 2021).

We used the processed emissions as input for MTEA method and found that the

model results show obvious diurnal pattern as well. The diurnal patterns are characterized by two peaks in the day, one occurring at 10:00-15:00 (local time, UTC+8) and the other appearing at 22:00-00:00. The 10:00-15:00 peak can be explained by the elevating emissions of $PM_{2.5}$ precursors, such as NO_x and SO_2, as well as strong solar radiation. The intensive solar radiation around noon can promote production of hydroxyl (OH) radical, and further oxidizes substantial precursors to form secondary particles. However, the 22:00-00:00 peak is mostly attributed to the other two factors. Firstly, the primary $PM_{2.5}$ obviously is decreased due to the reduction of emission activities at night. Meanwhile, the secondary $PM_{2.5}$ requires some time to generate and accumulate, thus lagging behind changes in emission. Secondly, nitrate particles can also be produced via N_2O_5 heterogeneous reactions in the nighttime.

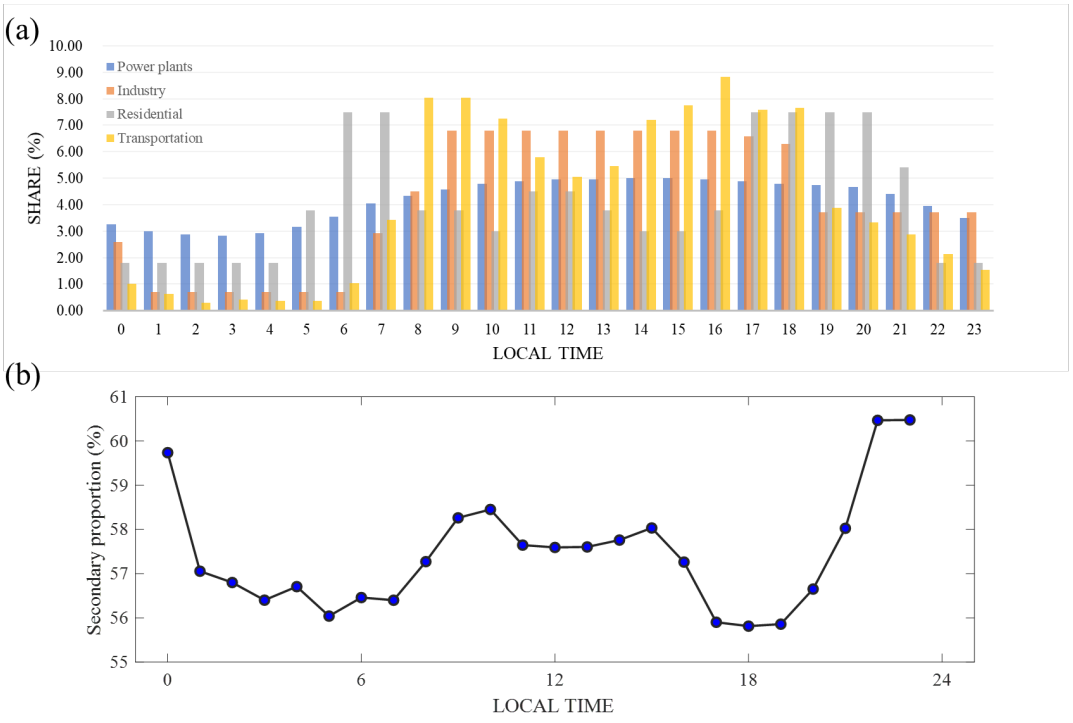


Figure R3. (a) The diurnal distribution of anthropogenic emissions from power plants, industry, residential and transportation (Unit: %). (b) The diurnal variation of the estimated nationwide secondary proportion of $PM_{2.5}$ (Unit: %).

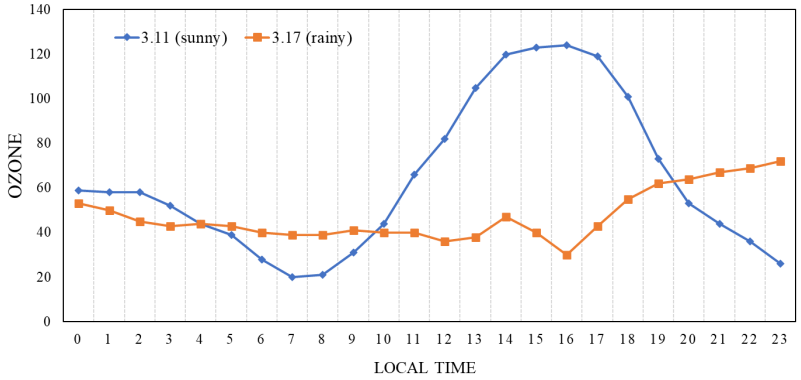
11. Section 4.4: Why did you exclude the wet deposition case here but include in

other sections? I would suggest adding the application condition for your method somewhere.

Response: Thank you for your highly careful reminding. Section 4.4 is aimed at discussing the statistical correlation between ozone versus $PM_{2.5}$. We used the daily concentration of these two variables as inputs for further investigation. For $PM_{2.5}$, the 24-h mean concentration can be applied to representing its daily level. The maximum daily 8-h average ozone concentration (MDA8) is usually adopted for describing its concentration level on the daily time-scale. As the reviewer said in the 10^{th} point, ozone is a kind of typical secondary air pollutant with distinctive diurnal pattern (Wang et al., 2017). As shown in Fig. R3, the precipitation process can destroy this diurnal pattern because of the extremely weak radiative condition on rainy days. Meanwhile, ozone concentration level under this condition is mainly affected by background fields. Therefore, MDA8 of rainy days can reveal the background concentration characteristics but not the intensity of secondary formation. To explain the relationship between PM and O_3 from the aspect of chemical generation, removing the background dominated cases of O_3 concentrations which under precipitation is necessary. We have followed your suggestion to add the explanation for using this preprocessing and rephrase the related texts in Section 4.4.

Revision in Section 4.4:

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.



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329 **Figure R3.** The diurnal variations of O_3 concentration in Shanghai on 11 Mar (sunny weather) and
330 17 Mar (rainy weather), 2022 based on the observations from MEE.

331

332 12. The general method to calculate the portion of secondary $PM_{2.5}$ is chemical
333 transport model using bottom-up inventory. It's better to examine the difference in the
334 result between your method and CTM with same inventory.

335 **Response:** Thanks for your highly conducive comments and rigorous attitude to
336 scientific research. It is really an awesome suggestion. We completely agree that
337 chemical transport model (CTM) is another useful tool to reveal the aerosol
338 compositions. It is interesting to conduct a parallel comparison between two kinds of
339 modeling methods. To examine the difference in result between the MTEA approach
340 and traditional CTM, we adopted the monthly simulated PPM/SPM concentrations
341 from a data fusion system developed by Tsinghua University. This system, which is
342 named Tracking Air Pollution in China (TAP), integrates ground measurements,
343 satellite remote sensing retrievals, emission inventories (MEIC), and CTM
344 simulations (WRF/CMAQ) based on machine learning algorithms. More descriptions
345 of this dataset can be found at http://tapdata.org.cn/ (Geng et al., 2021; Geng et al.,
346 2017). We treated the PPM and SPM concentrations from TAP as the state-of-the-art
347 model representation. Then we showed comparisons between MTEA and TAP in
348 terms of PPM, SPM concentrations and their annual trends in 31 populous cities of
349 China (Fig. R4). In general, comparisons indicate that MTEA estimation has a good

agreement with the CTM simulation. To add this part in the manuscript suggested by reviewer, we introduced the TAP dataset in Section 2.3 and showed the related comparisons in Section 3.1.3.

Revision in Section 2.3:

2.3 PPM and SPM estimated by CTM

Apart from evaluating PPM and SPM with various composition measurements, we also compared MTEA estimation with CTM results. Here we utilized the $PM_{2.5}$ composition gridded dataset with a spatial resolution of 10 km×10 km developed by Tsinghua University for further comparisons. This dataset is named Tracking Air Pollution in China (TAP, available at http://tapdata.org.cn/, last access 15 Mar 2022) (Geng et al., 2021; Geng et al., 2017). TAP is directly calculated by Community Multiscale Air Quality (CMAQ) model. In terms of methodology, based on machine learning algorithms, TAP integrates surface measurements, satellite remote sensing retrievals, emission inventories (MEIC) with CMAQ simulations. Moreover, it is also constrained by ground aerosol composition measurements. We collected the monthly mean concentrations of aerosol species during 2014-2018 from TAP, including SO_4^{2-}, NO_3^-, NH_4^+, OM, BC and total $PM_{2.5}$. SOA was further calculated from OM by EC-tracer model (Ge et al., 2017). SPM concentrations were inferred by summing SO_4^{2-}, NO_3^-, NH_4^+ and SOA. PPM concentrations were then obtained via deducting SPM from $PM_{2.5}$.

Revision in Section 3.1.3:

3.1.3 Comparison with the CTM simulation

In addition to evaluating our model via PPM and SPM measurements in China, we also provided a comparison between MTEA estimation and CTM simulation in 31 populous cities based on the monthly mean PM concentrations. As shown in Fig. R4 a-b, the correlation coefficient r for TAP versus MTEA is 0.86 in terms of PPM concentration and 0.91 in terms of SPM concentration, showing a strongly positive correlation between the two models. At the same time, both slopes (1.26 and 0.89) and

intercepts (–3.7 μg m^{-3} and 1.9 μg m^{-3}) of the regression about PPM and SPM illustrate that most of the scattering spots distribute around 1:1 ratio line.

Moreover, we further compared the long-term varying trends between MTEA versus TAP in averaged PPM and SPM concentration of 31 populous cities (Fig. R4 c-d). Both of them exhibit a descending interannual trend in PPM concentration, with a rate of –2.0 μg m^{-3} yr^{-1} for MTEA and –1.9 μg m^{-3} yr^{-1} for TAP. In terms of SPM concentration, the decline rates are –2.9 μg m^{-3} yr^{-1} and –2.8 μg m^{-3} yr^{-1}, respectively. Meanwhile, the statistical correlations between two interannual variations are 0.98 (PPM) and 0.99 (SPM), which are quite close to 1, showing a good agreement.

Thus, the comparisons about PPM/SPM concentration magnitudes and interannual variations between two kinds of models suggest that statistical model can infer similar estimation with traditional CTM. Meanwhile, it is again highlighted that our model is capable of capturing reasonable PPM and SPM concentrations. Furthermore, it is also shown that MTEA can track primary and secondary component of $PM_{2.5}$ by using proxy at a much lower cost when compared to traditional air quality model simulations.

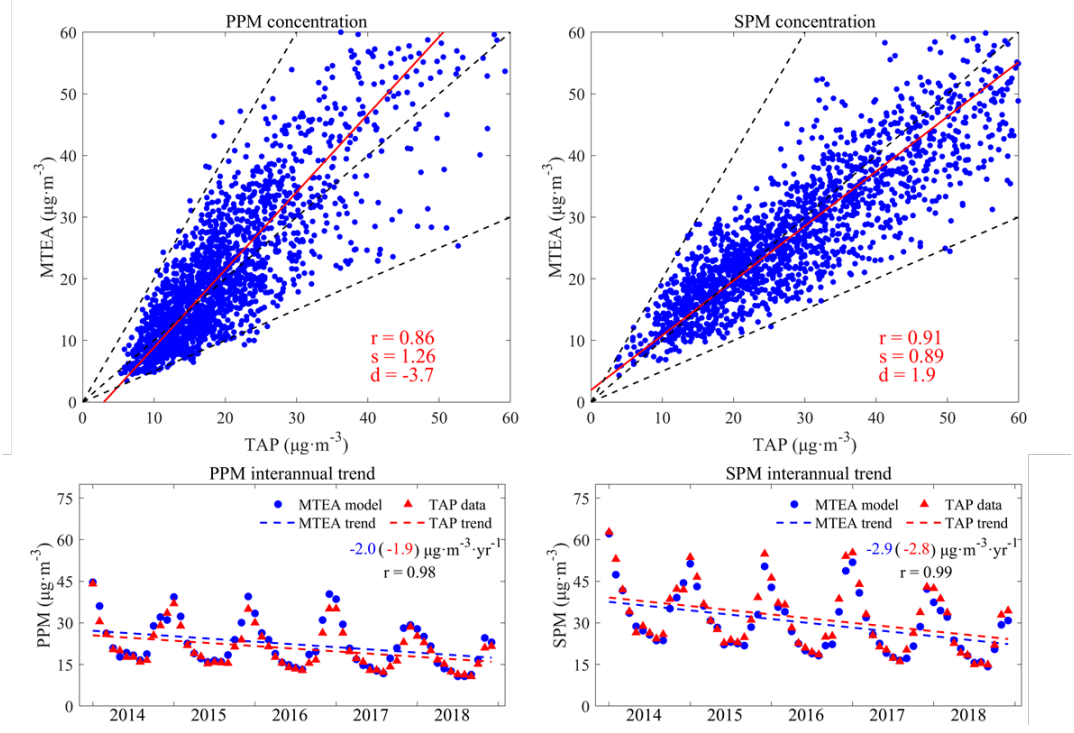

Figure R4. Comparisons between MTEA and TAP in terms of PPM, SPM concentrations and

their annual trends from 2014 to 2018 in 31 populous cities of China. In panel (a) and (b), each blue solid dot stands for a monthly mean concentration of PPM or SPM in one of 31 populous cities. The number of samples is 1860 (60×31). The metrics r, s and d represent correlation coefficient, slope and intercept of fitting line, respectively. The fitting method follows the Reduced Major Axis (RMA) regression. In panel (c) and (d), MTEA and TAP are marked by blue circles and red triangles. Each dot represents the mean PPM/SPM concentration of 31 cities. The colorful numbers stand for the annual trends of PPM and SPM concentrations during 2014-2018. At the same time, the correlation coefficient (r) between MTEA versus TAP is also provided.

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