Response to RC#3:

Dear Editor and anonymous referee #4:

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.

1. The manuscript presents a method for estimating the relative contributions of primary and secondary PM by proxy. The observed input parameters are CO, PM_{10} and $PM_{2.5}$, however, the method also relies on estimated emissions of OA, EC, OC, fine dust, $PM_{2.5}$, sulfate and nitrate, from emission inventories. The authors develop a proxy for secondary particulate matter on the basis of the observed parameters and estimated emissions. The motivation is presented as the need for a low cost, operational method for monitoring the contributions of secondary aerosols to the total $PM_{2.5}$ levels.

The method appears to have some use for informing operational air quality management or for informing policy, but the scientific value of the method is not convincingly presented. It relies on assumptions and inventories that are not universal, and the manuscript does not present a convincing argument for its use, other than that it is cheaper than source apportionment methods based on chemical speciation. But it does not present comparative estimates of primary-secondary contributions with those methods.

It is questionable if this method has any value. It requires a big body of inputs, as other chemical transport models, but also relies heavily on assumptions and coefficients that are externally adjusted, even tuned to fit the model.

Response: Thank you for the comments. The traditional methods to identify

$PM_{2.5}$ compositions include observational and simulation methods. The observational method is currently the most common and useful way for quantitatively investigating the $PM_{2.5}$ chemical compositions. Moreover, chemical transport model (CTM) is another useful tool to identify the composition characteristics of $PM_{2.5}$. However, the CTM results are largely dependent on external inputs as the reviewer mentioned 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).

Different from CTM, the MTEA model developed in this study is a statistical model, which does not suffer from the burden of high requirement in computational cost and storage. MTEA is positioned as a low economy-/technique-cost tool to conveniently estimate the primary and secondary $PM_{2.5}$ in both scientific and practical areas, although concomitantly it is slightly inferior to the two traditional methods in terms of identifying detailed $PM_{2.5}$ compositions and capturing high temporal variation.

The aim of this study, by using MTEA, is to reveal the general characteristics of primary and secondary $PM_{2.5}$ pollution over wide spatio-temporal coverages. The evaluation between MTEA estimation versus various measurements in terms of monthly mean value shows a satisfying performance (Section 3). At the same time, the reasonable spatio-temporal patterns of PPM and SPM concentrations disclosed by our model also inform that MTEA is a promising tool for illustrating general pollution patterns. Thus, for studies which would like to distinguish primary and secondary $PM_{2.5}$, MTEA model can serve as a potential option. In the future, we also hope to cooperate with the team which focuses on observational studies to broaden the application of MTEA and reduce the uncertainty. Thanks again and we have rephrased our texts in the manuscript for a clearer description in terms of the scientific value of our method in Section 1.

Revision in Section 1:

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. The manuscript describes comparisons between estimated and observed primary particulate matter. Categorization of measured historical data into secondary and primary aerosols for comparison with the MTEA seems to be based on chemical compositions, but this process is not clearly described and the criteria are vague. There has been no attempt to verify the MTEA estimates for ppm by comparing with published estimates based on receptor modelling, CTMs or AMS studies. There are many studies in the literature that have produced estimates that can be easily compared with the outcomes of the MTEA approach, but that has not been done.

Response: Thank you for pointing this out. We have added the description about categorizing the concentrations of measured historical aerosol chemical species into PPM and SPM concentrations in Section 2.2.2 and 2.2.3.

The estimation from MTEA model is based on the routine $PM_{2.5}$ observation. However, the measurements from literature we summarized in Section 3.1.2 rely on sampling at different locations. The measurements may be quite different though the observational campaigns were conducted in the same city. Thus it is difficult to directly compare PPM concentrations predicted by MTEA with that in various literature. Therefore, we mainly focus on the comparison in terms of secondary proportions of $PM_{2.5}$ between the MTEA method versus various previous studies. Please refer to Table S4 in the supplementary material for the specific comparisons. Moreover, we also revised Table S4 to clearly show the method applied by these previous studies (offline sampling or AMS instrument).

To examine the difference in result between the MTEA approach and traditional CTM, we adopted the monthly simulated PPM/SPM concentrations from a data fusion system developed by Tsinghua University. This system, which is named Tracking Air Pollution in China (TAP), integrates ground measurements, satellite remote sensing retrievals, emission inventories (MEIC), and CTM simulations (WRF/CMAQ) based on machine learning algorithms. More descriptions of this dataset can be found at http://tapdata.org.cn/ (Geng et al., 2021; Geng et al., 2017). We treated the PPM and SPM concentrations from TAP as a typical model representation. To add this part in the

manuscript suggested by reviewer, we introduced TAP dataset in Section 2.3 and showed comparisons between MTEA and TAP in terms of PPM, SPM concentrations as well as their annual trends in 31 populous cities of China in Section 3.1.3.

Revision in Section 2.2.2:

After accessing the chemical compositions, we categorized them into PPM and SPM for further evaluation. Specifically, SOA was roughly identified from OM by EC-tracer model (Ge et al., 2017). SPM concentrations were calculated via summing SO_4^{2-}, NO_3^-, NH_4^+ and SOA concentrations. Then PPM could be calculated though deducting SPM from $PM_{2.5}$.

In addition, we investigated observation-based $PM_{2.5}$ component analyses in 16 cities of China during 2012-2016 from 32 published studies. This survey offered an opportunity to compare the estimation by MTEA with the past measurements in the terms of the secondary fraction of $PM_{2.5}$. SPM concentrations in literature were roughly estimated by multiplying OM from 0.5 because of the limit of data source. Meanwhile, it is noted that the factor which converts OC to OM is dependent on the definition of each observation study itself.

Revision in Section 2.2.3:

The specific aerosol chemical compositions include ammonium sulfate, ammonium nitrate, organic/elemental carbon and soil/mineral dust. The categorization for PPM and SPM in IMPROVE dataset is similar to the process in Section 2.2.2. The only difference is that SPM concentration is the sum of ammonium sulfate, ammonium nitrate and SOA.

Revision in Section Table S4:

#Please see below#

Table S4. List of $PM_{2.5}$ component measurements (μg m^{-3}) of China in previous studies.

City	Period	$PM_{2.5}$	SO_4^{2-}	NO_3^-	NH_4^+	SOA[1]	SPM/$PM_{2.5}$	Mean SPM/$PM_{2.5}$	Method	References
Beijing	2012 Summer	103	20.6	15.8	8.3	10.1	53.2%	**31% ~ 80%**	Offline	Tian et al., 2015
	2012 - 2013	72	9.3	11.9	5.3	9.6	50.3%		Offline	Liu et al., 2018b
	2013 Winter	159	25.4	19.0	15.6	32.2[a]	58.0%		Offline	Tao et al., 2015
	2013 Winter	143	23.9	20.2	16.5	15.4	53.1%		AMS	Huang et al., 2014b
	Jan 2014	153	9.6	12.1	6.7	33.8[c]	40.6%		Offline	Gao et al., 2018
	Apr 2014	115	10.7	10.7	11.4	15.2[c]	41.6%		Offline	Gao et al., 2018
	Jul 2014	96	25.6	25.6	14.1	11.1[c]	79.7%		Offline	Gao et al., 2018
	Oct 2014	139	21.1	45.5	13.9	23.0[c]	74.5%		Offline	Gao et al., 2018
	2014 Winter	138	21.0	26	14.1	17.4	51.8%		Offline	Lin et al., 2016
	Jun 2014 - Apr 2015	100	14.3	17.1	11.5	12.4[b]	55.6%		Offline	Huang et al., 2017
	May 2015 - Apr 2016	114	8.6	11.1	5.2	10.1	30.9%		Offline	Yu et al., 2019
	Jul 2015 - Apr 2016	81	9.6	12.4	8.6	7.7	47.1%		Offline	Xu et al., 2019
	Oct 2016	95	16.8	16.8	12.5	12.3[b]	61.5%		Offline	Zhang et al., 2018
	MTEA estimation [2]							**41%**	-	This study
Tianjin	Jun 2014 - Apr 2015	106	16.6	16.2	13.7	10.4[b]	53.8%	**41% ~ 54%**	Offline	Huang et al., 2017
	Jul 2015 - Apr 2016	86	12.1	13.9	10.5	7.3	51.0%		Offline	Xu et al., 2019
	Jul 2014	113	12.2	16.2	9.3	11.0[c]	43.0%		Offline	Gao et al., 2018
	Oct 2014	101	12.8	9.9	8.2	11.1[c]	41.4%		Offline	Gao et al., 2018
	2014 Winter	183	19.5	40.7	15.1	21.8[c]	53.1%		Offline	Gao et al., 2018
	May 2015 – Apr 2016	120	18.1	20.3	8.5	10.0	47.4%		Offline	Liu et al., 2018a

	MTEA estimation						**63%**			This study
Shijiazhuang	Jun 2014 - Apr 2015	155	25.5	23.4	18.8	17.7[b]	55.0%	**51% ~ 55%**	Offline	Huang et al., 2017
	Jul 2015 - Apr 2016	105	16.8	14.9	12.3	9.6	51.0%		Offline	Xu et al., 2019
	MTEA estimation						**49%**			This study
Shanghai	2012 Spring	70	15.3	8.6	6.4	5.7	51.4%	**26% ~ 71%**	Offline	Zhao et al., 2015
	2012 Summer	51	9.7	5.6	3.6	3.7	44.3%		Offline	Zhao et al., 2015
	2012 Fall	82	17.9	20.2	7.8	7.7	65.4%		Offline	Zhao et al., 2015
	2012 Winter	70	11.6	13.2	5.6	8.5	55.6%		Offline	Zhao et al., 2015
	2012 Spring	64	12.0	10.8	4.3	4.9	50.0%		Offline	Huang et al., 2014a
	2011- 2013 Spring	49	11.0	11.0	6.9	5.9	71.0%		Offline	Wang et al., 2016a
	2011- 2013 Summer	31	8.1	5.2	4.2	4.7	67.3%		Offline	Wang et al., 2016a
	2011- 2013 Fall	41	8.8	7.4	4.8	5.2	63.9%		Offline	Wang et al., 2016a
	2011- 2013 Winter	65	13.0	13.2	8.3	6.7	63.4%		Offline	Wang et al., 2016a
	2012 - 2013	68	13.6	11.9	5.8	8.6	58.7%		Offline	Liu et al., 2018b
	Oct - Nov 2013	75	12.9	15.0	6.6	4.2	51.6%		Offline	Ming et al., 2017
	Dec 2013 – Jan 2014	138	19.5	29.1	12.6	10.3	51.8%		Offline	Ming et al., 2017
	Mar 2014 – Apr 2014	96	12.3	10.4	5.5	4.5	34.1%		Offline	Ming et al., 2017
	Jun 2014 – Jul 2014	56	6.7	2.8	2.1	2.9	25.9%		Offline	Ming et al., 2017
	2013 Winter	91	10.8	12.4	7.5	21.8[b]	57.7%		AMS	Huang et al., 2014b
	Dec 2014 – Jan 2015	103	18.3	25.4	14.4	14.1[b]	70.1%		Offline	Du et al., 2017
	Mar 2015 – Apr 2015	74	8.7	11.2	5.7	9.2[b]	47.0%		Offline	Du et al., 2017
	MTEA estimation						**67%**			This study
Nanjing	Apr – May 2013	110	23.1	11.7	6.4	17.7[a]	53.5%	**52% ~ 79%**	Offline	Li et al., 2016

	Aug 2013	86	18.4	8.1	5.4	14.2[a]	53.6%		Offline	Li et al., 2016
	Oct 2013	77	12.6	7.3	3.8	36.8[a]	78.6%		Offline	Li et al., 2016
	Dec 2014 – Jan 2015	100	11.7	16.4	12.3	11.8[b]	52.2%		Offline	Du et al., 2017
	Mar 2015 – Apr 2015	83	21.4	16.1	7.9	9.1[b]	65.6%		Offline	
	MTEA estimation							**53%**		This study
Hangzhou	Oct 2013	36	9.7	5.3	6.0	6.5	76.4%	**76%**	Offline	Wu et al., 2016
	MTEA estimation							**53%**		This study
Guangzhou	Dec 2012 – Jan 2013	75	10.6	5.8	5.1	6.5	37.3%	**37% ~ 58%**	Offline	Liu et al., 2014
	2012 - 2013	75	13.1	7.2	4.8	8.4	44.6%		Offline	Liu et al., 2018b
	Nov 2012 – Dec 2013	61	9.3	5.7	4.2	11.9[b]	51.0%		Offline	Chen et al., 2016
	2013 Summer	51	8.9	4.9	4.0	6.6	47.8%		Offline	Cui et al., 2015
	2013 Fall/Winter	68	9.8	7.3	4.5	9.4	45.6%		Offline	Cui et al., 2015
	2013 Winter	69	12.7	8.9	6.9	11.4[b]	57.8%		AMS	Huang et al., 2014b
	2014 Spring	44	8.2	2.4	3.6	4.5	42.5%		Offline	Tao et al., 2017
	2014 Summer	37	7.6	0.3	2.6	3.7	38.4%		Offline	Tao et al., 2017
	2014 Fall	48	11.4	1.0	4.4	4.7	44.8%		Offline	Tao et al., 2017
	2014 Winter	63	9.8	5.5	4.8	7.0	43.0%		Offline	Tao et al., 2017
	MTEA estimation							**66%**		This study
Xi'an	Dec 2012	137	13.5	9.8	6.6	21.6[b]	37.6%	**33% ~ 55%**	Offline	Zhang et al., 2015
	2012 Spring	164	17.8	15.2	6.5	13.9	32.6%		Offline	Niu et al., 2016
	2012 Summer	109	25.0	10.1	6.6	8.8	46.3%		Offline	Niu et al., 2016
	2012 Fall	155	18.7	16.5	8.2	18.4	39.9%		Offline	Niu et al., 2016
	Nov 2012 – Feb 2013	244	32.1	29.3	16.8	39.7	48.3%		Offline	Niu et al., 2016

	Dec 2014 – Nov 2015	113	15.2	16.6	8.4	21.3	54.7%		Offline	Dai et al., 2018
	MTEA estimation							**55%**		This study
Chengdu	Oct – Nov 2014	62	10.5	9.3	6.9	8.3[b]	56.5%		Offline	Wang et al., 2018
	Jan – Feb 2015	114	16.4	17.5	12.7	15.8[b]	54.7%		Offline	Wang et al., 2018
	Apr 2015	48	8.3	5.9	5.1	5.0[b]	50.6%	**44% ~ 57%**	Offline	Wang et al., 2018
	Jul 2015	45	9.7	3.9	4.2	5.9[b]	52.6%		Offline	Wang et al., 2018
	Jan 2015	48	6.1	3.7	2.4	8.7	43.5%		Offline	Li et al., 2017a
	MTEA estimation							**46%**		This study
Chongqing	2012 - 2013	74	19.7	6.5	6.1	8.6	55.3%		Offline	Liu et al., 2018b
	Oct – Nov 2014	56	9.9	7.8	5.7	7.8[b]	55.7%		Offline	Wang et al., 2018
	Jan – Feb 2015	115	17.5	15.8	11.3	19.4[b]	55.7%	**44% ~ 56%**	Offline	Wang et al., 2018
	Apr 2015	58	10.4	5.9	5.2	8.0[b]	50.1%		Offline	Wang et al., 2018
	Jul 2015	54	11.1	1.6	4.0	6.8[b]	43.5%		Offline	Wang et al., 2018
	MTEA estimation							**61%**		This study
Lanzhou	Dec 2012	120	11.8	7.2	6.7	21.2	39.1%		Offline	Tan et al., 2016
	Jun – Jul 2013	34	4.3	1.9	1.9	5.8	40.9%		Offline	Tan et al., 2016
	Apr – May 2014	83	4.0	1.7	0.8	8.0	17.5%	**18% ~ 41%**	Offline	Wang et al., 2016b
	Aug 2014	38	4.8	2.0	1.3	3.5	30.5%		Offline	Wang et al., 2016b
	Oct 2014	93	5.8	7.1	3.6	12.7	31.4%		Offline	Wang et al., 2016b
	Jan, Dec 2014	141	7.6	10.1	6.0	18.2	29.7%		Offline	Wang et al., 2016b
	MTEA estimation							**63%**		This study
Changsha	Sep – Oct 2013	102	19.4	2.6	8.7	12.5[b]	42.4%	**41% ~ 44%**	Offline	Tang et al., 2017
	Dec 2013 – Jan 2014	145	19.3	9.7	14.3	20.5[b]	44.0%		Offline	Tang et al., 2017

	Apr – May 2014	97	17.0	1.4	7.5	14.0[b]	41.1%		Offline	Tang et al., 2017
	Jul – Aug 2014	78	13.9	2.9	7.4	9.4[b]	43.1%		Offline	Tang et al., 2017
	MTEA estimation							**67%**		This study
Haikou	Jan 2015	17	3.1	0.5	1.0	2.3	40.1%	**32% ~ 40%**	Offline	Liu et al., 2017
	Mar 2015	9	1.6	0.2	0.5	1.2	38.8%		Offline	Liu et al., 2017
	Jul 2015	23	3.8	0.3	0.8	2.4	31.7%		Offline	Liu et al., 2017
	Sep 2015	47	7.9	3.1	3.0	2.8	35.7%		Offline	Liu et al., 2017
	MTEA estimation							**61%**		This study
Zhengzhou	Oct 2014	143	19.6	17.9	9.2	12.0	41.0%	**41% ~ 54%**	Offline	Jiang et al., 2017
	Dec 2014 – Jan 2015	191	23.5	26.5	19.8	22.6	48.4%		Offline	Jiang et al., 2017
	Apr 2015	138	19.7	20.3	14.4	11.3	47.6%		Offline	Jiang et al., 2017
	Jul 2015	110	24.2	14.3	13.9	7.3	54.3%		Offline	Jiang et al., 2017
	MTEA estimation							**60%**		This study
Shenyang	2013 – 2014	82	13.2	4.6	4.5	11.7	41.5%	**42%**	Offline	Liu et al., 2018b
	MTEA estimation							**51%**		This study
Lhasa	2013 – 2014	36	0.8	0.5	0.4	7.6	25.8%	**26%**	Offline	Liu et al., 2018b
	MTEA estimation							**64%**		This study

[1] SOA = 0.5*OM, OM = f * OC. Default f is 1.2. In case of a, b and c, the f is 1.8, 1.6 and 1.4 respectively.

[2] For period of 2014-2018.

Revision in 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. R1 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 $\mu g\ m^{-3}$ and 1.9 $\mu g\ m^{-3}$) 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. R1 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.

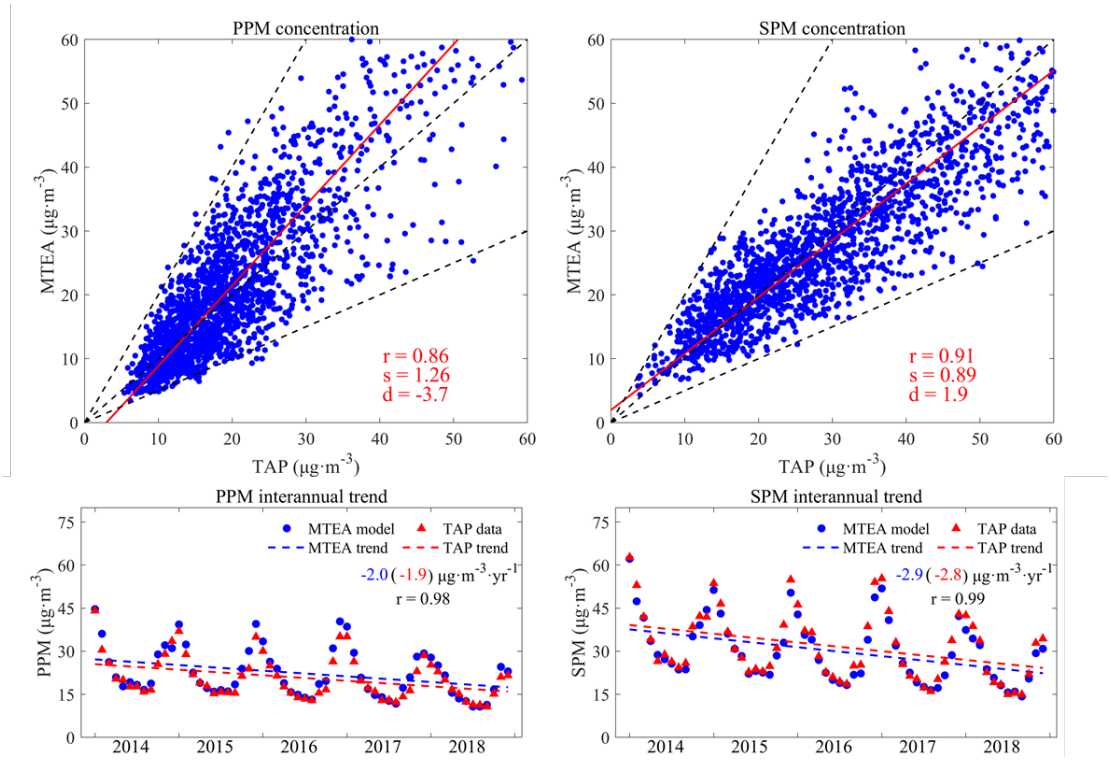

Figure R1. Comparisons between MTEA and TAP in terms of PPM, SPM concentrations and their annual trends from 2014 to 2018 in 31 populous cities of China. In panel (a) and (b), each blue solid dot stands for a monthly mean concentration of PPM or SPM in one of 31 populous cities. The

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.

3. It is true as the authors state that those other methods are labor-intensive and expensive, but they are also scientifically tried and tested and therefore more convincing, so it would make sense to develop the performance of the MTEA against such methods more than has been done in this manuscript.

Response: Thank the reviewer for pointing this out. There is no doubt that the measurements via offline or online methods are absolutely crucial to scientifically understanding the compositions of $PM_{2.5}$. To some extent, the identification of $PM_{2.5}$ based on these methods offers a conclusive insight for model developers, and the MTEA model we developed should be in line with the observational results. We heartfeltly acknowledged the efforts that the highly scientific observations made. We compared the MTEA results with a series of observational studies as shown in Table S4, and revised the related text in Section 2.2.

In addition, this study mainly devotes to illustrating the general pattern of primary and secondary $PM_{2.5}$ pollution over a wide spatio-temporal coverage with the aid of a convenient proxy tool, and has no intention to replace the crucial observational methods with MTEA. Thank you for the review's comment again and we have revised the related texts in Section 1 to clarify the roles and relationships between observational method and MTEA.

Revision in Section 1:

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).

4. Also, the manuscript states that the numerical calculations were done on a supercomputing system. It can be argued that if the approach requires a supercomputing facility, then it is no less costly or inaccessible than the existing source apportionment methods, but the cost has been shifted from scientific equipment to IT services.

Response: Thank you for the comment and the careful reminding. We indeed agree that traditional numerical models such as WRF-Chem, CMAQ and CAMx does cost considerable computational sources. However, our model is based on the statistical principle. Actually, it is capable of running on the personal computer (PC) platform with basic equipment requirement. In the future, we look forward to simplifying the model for a more lightweight version so that it can be easily utilized for application anywhere.

5. The manuscript does touch on a discussion that has scientific interest, and that is contained in the sections 4.1 and 4.2 on spatial and temporal variation. The discussion on spatial variation has some merit. There is potentially a better motivation for

developing the MTEA approach in order to inform a discussion on the spatial and temporal variation where only proxy parameters are available, by leveraging national monitoring networks to learn more about geographical distribution of secondary aerosols and feed into a discussion on variations in atmospheric processes.

Response: Thank you for your comments and rigorous attitude to scientific research. We have rephrased our statement of the motivation for this study in Section 5.

Revision in Section 5:

In this study, we developed a new approach MTEA to distinguish the primary and secondary compositions of $PM_{2.5}$ efficiently from routine observation of $PM_{2.5}$ concentration with much less computation cost than traditional CTMs.

Meanwhile, our model posed a great agreement with the reanalysis dataset from one of the most advanced CTMs in China as well.

The methods to quantify different $PM_{2.5}$ components are often based on either lab analysis of offline filter samplings or online observation instruments such as AMS. However, these methods are often labor-intensive, strict technical and high economic cost. CTM is another useful tool to reveal the composition characteristics of $PM_{2.5}$. But traditional CTMs are short in high requirement of hardware as well. Our study develops an efficient approach based on statistical principle to explore PPM and SPM with lower cost, and applying this approach to large-scale observation networks, such as the MEE network, can offer an unprecedented opportunity to quantify the $PM_{2.5}$ components on a large space and time scale.

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