

We want to thank the reviewers for their support of the paper and their helpful comments and suggestions which have helped to improve our manuscript. We provide a point by point response to review.

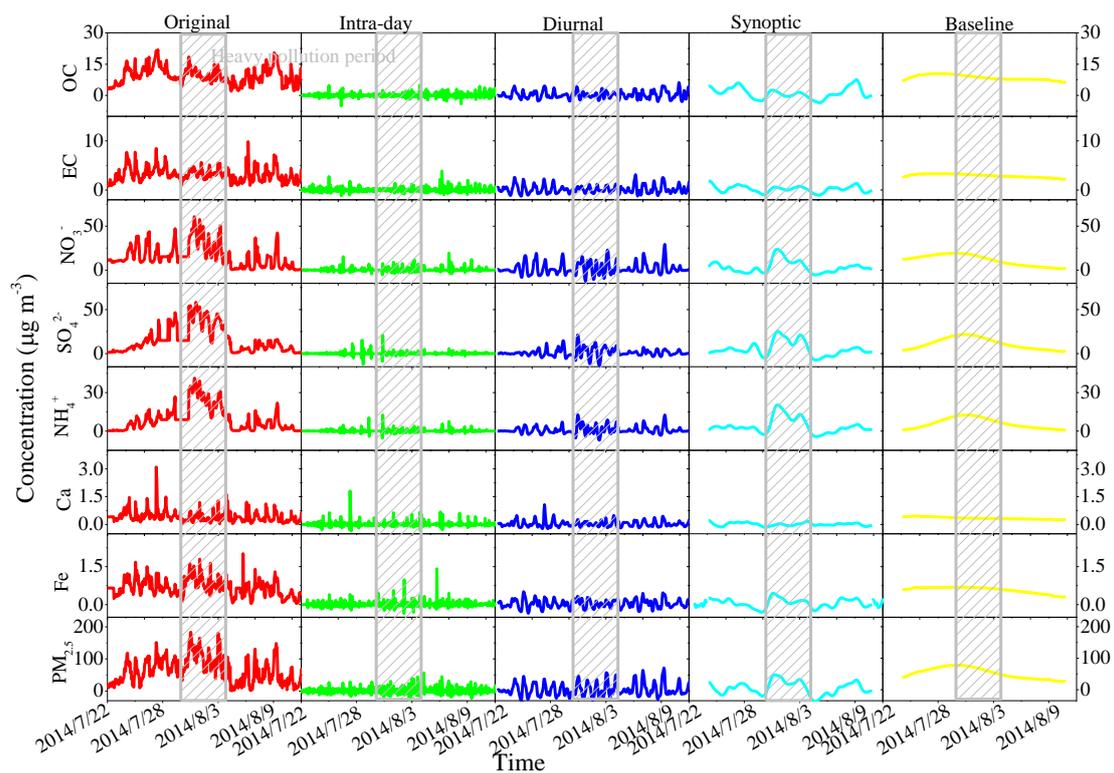
**Response to Anonymous Referee #3**

The authors have proposed to improve the identification of major sources of ambient PM<sub>2.5</sub> by conducting source apportionment upon the spectral decomposition of hourly measurements of ambient PM<sub>2.5</sub> and its major ionic and trace element components. I think the manuscript falls short of fulfilling the above purposes. Several issues arise after reviewing the manuscript:

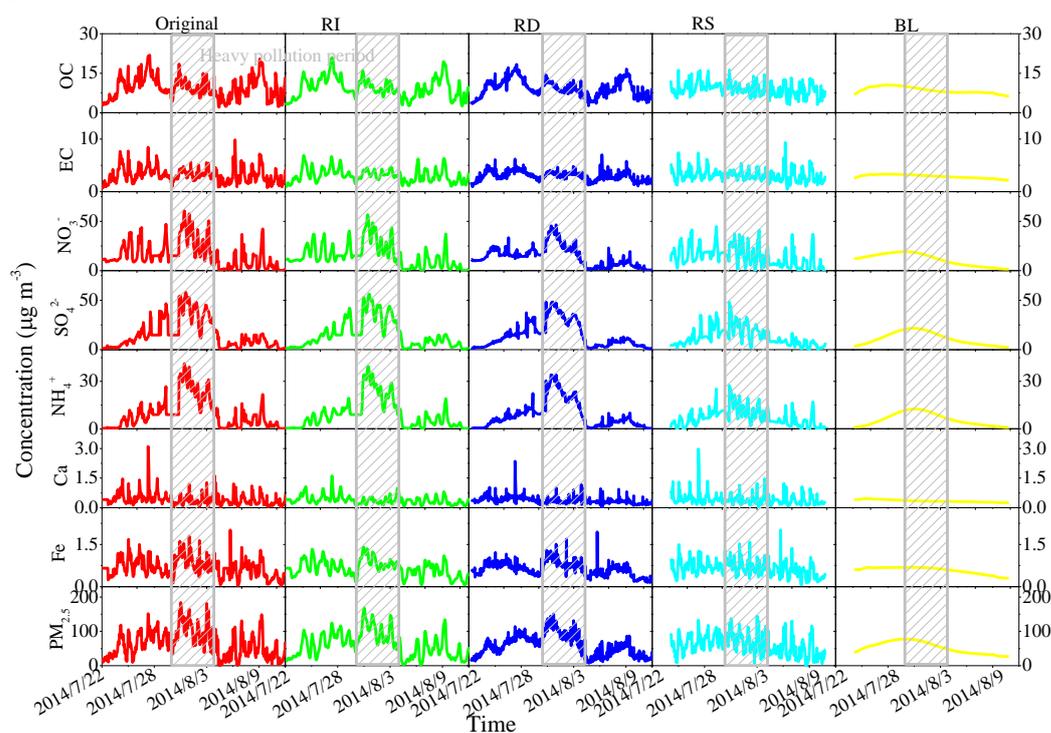
a) The process of decomposing the original data in time series of different temporal variations does not ensure that each of them is strictly positive, as they should. This is problematic for the subsequent application of ME2 (or PMF).

Response: We thank the reviewer for the comments and identifying some points that need to be addressed to strengthen the paper.

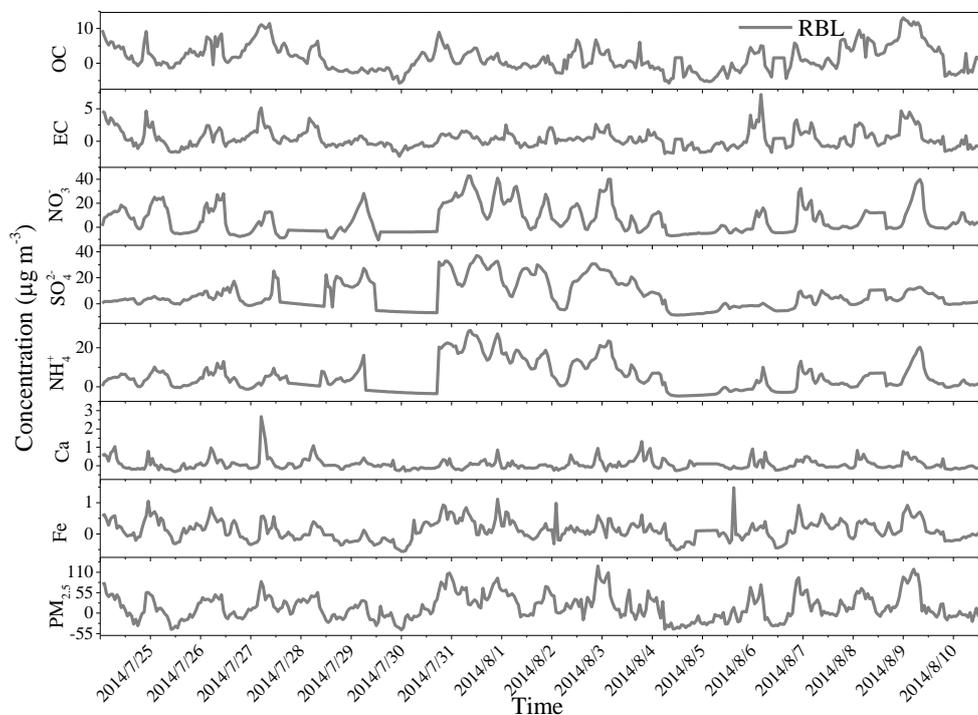
The temporal-scale components might show a positive or negative impact on pollutant concentrations resulting in some negative variations of them (Figures 1 and S6). So we designed an approach that can help solve more negative values in model input data than simply inputting time series of different temporal. We singly removed one a specific temporal-scale component (TS component) from the original dataset to ensure the input data are positive as much as possible, as shown in Figure. S5.



**Figure 1.** The variance of PM<sub>2.5</sub> and chemical species concentrations influenced by intra-day (period less than 12 h), diurnal (12-24 h), synoptic (2-21 days), and baseline (greater than 21 days) temporal-scale (TS) components, for the period of 22 July 2014 to 13 Aug 2014 at Beijing, China. The variation of species that originated from primary sources mainly was influenced by diurnal TS components. The variation of ions and OC (partly from the secondary formation) that originated from secondary formation primarily were affected by synoptic TS component. The vertical gray lines demarcate the heavy pollution period.



**Figure S5.** The variance of PM<sub>2.5</sub> and chemical species concentrations from original, RI, RD, RS and BL datasets, for the period of 2 July 22, 2014, to Aug. 13, 2014, in Beijing, China. (RI: intra-day removed dataset, RD: diurnal removed dataset, RS: synoptic removed dataset, BL: baseline dataset). The vertical gray lines demarcate the heavy pollution period.



**Figure S6.** The variance of PM<sub>2.5</sub> and chemical species concentrations from RBL datasets, for the period of July 22, 2014, to Aug. 13, 2014, in Beijing, China.

b) There is no quantification of uncertainties associated to each spectral decomposition.

This is problematic for a correct application of ME2/PMF. Besides, the subtractions that appear in equations (9) – (16) increase the uncertainties of individual spectral components. This degrades input data quality, potentially hampering source identification.

Response: We have to admit that it is difficult to quantify the uncertainties of each spectral decomposition. Because the average concentrations of species for the four created datasets are similar to the original dataset, we assume they have the same uncertainty for the four datasets when we run ME2.

c) From equations (12) and (16) it follows that the RBL dataset is the sum of three spectral components. Nonetheless, the authors state that the above set has “many negative values” (line 233). This is a strange result and casts doubt on the suitability of the above decomposition for generating suitable input data for ME2/PMF.

Response: As mentioned above that the temporal-scale components have a positive or negative impact on pollutant concentrations. The intra-day, diurnal, and synoptic TS components showed negative variations sometime resulting in negative values of the sum of the three TS components. Negative values accounted for 38% of the total values in the RBL dataset, higher than in RI (0), RD (0), and RS (0.1%) datasets. Part of the RBL dataset was shown in Figures S5 and S6. To reduce the uncertainty of replacing the negative values, we used PCA rather than ME2 to estimate the sources qualitatively.

d) The dataset is too limited to validate the proposed methodology. Only two months of hourly data have been used. Monthly seasonality is absent in the analyses, and this is a serious flaw. Another limitation of this small datasets is the poor fitting of As, Cr, and Se after applying ME2 to the original dataset (lines 338-340). These trace elements may come from intermittent (point) sources that arrive to the receptor site only when specific meteorological conditions are met. The data set is too small to resolve those sources.

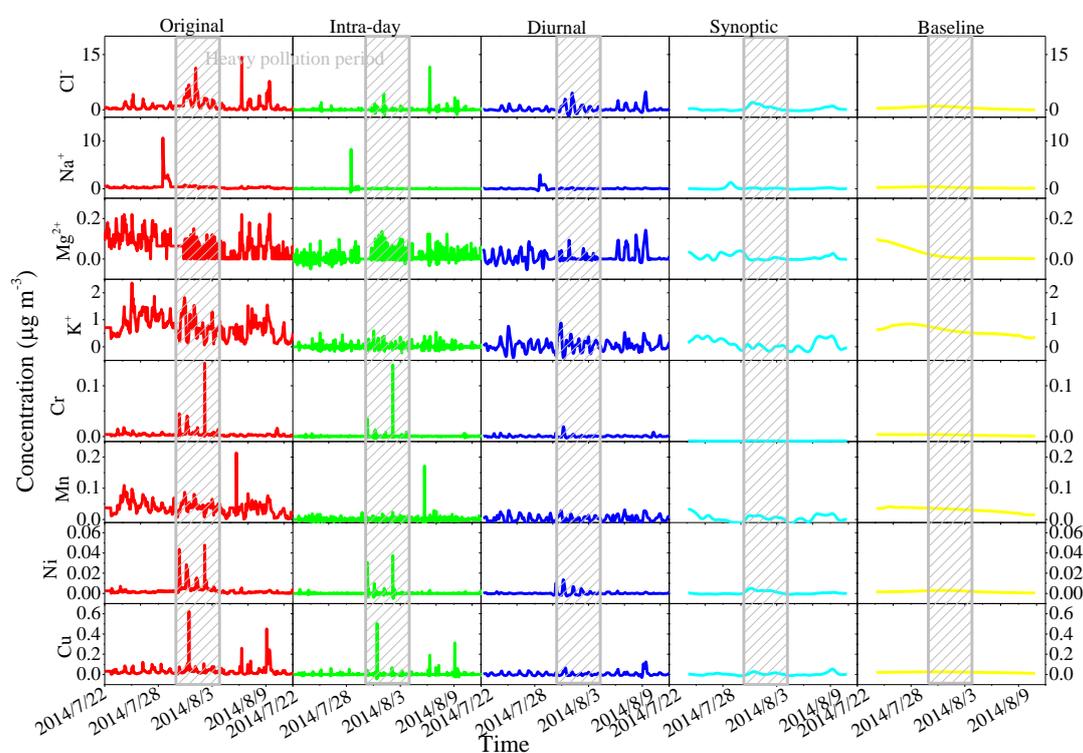
Response: We tried our best to resolve the problem about the seasonal variations and added another analysis about winter dataset. As limitations of the experimental condition, it is difficult to obtain long-term, continuous, and high-quality dataset. We

collected dataset (about five months) during winter in Tianjin, China and analyzed the dataset following the same way of Beijing dataset. Results from Tianjin datasets shown that diurnal and synoptic TS components had higher relative contributions to the total variance of OC, EC, PM<sub>2.5</sub>, and most ions and elements than the intra-day and baseline TS components. Six source categories were identified using ME-2 from both original and RD datasets in Tianjin, including crustal dust, vehicle emissions, coal combustion, secondary formation, biomass burning & sea salt, and industrial source. While industrial source has not been identified from both RD and RS dataset that mixed with crustal dust. The TPS impacts calculated from the original, RI, RD, and RS datasets were similar (ranged from 33.8 to 38.2  $\mu\text{g m}^{-3}$ ), the TSS impacts derived from the original and RI datasets exhibited similar source impacts (about 33  $\mu\text{g m}^{-3}$ ), which was higher than the solution from the RD (27.3  $\mu\text{g m}^{-3}$ ) and RS dataset (23.0  $\mu\text{g m}^{-3}$ ). Both Beijing and Tianjin results suggested TPS impact levels were mainly influenced by baseline TS component, and TSS impact levels were mainly influenced by synoptic and baseline TS components. Seasonal variations of source impacts were investigated in Tianjin. A more significant decrease in secondary source impact during winter suggested that the synoptic TS component had a more significant impact on the secondary source during winter than during the fall in this work.

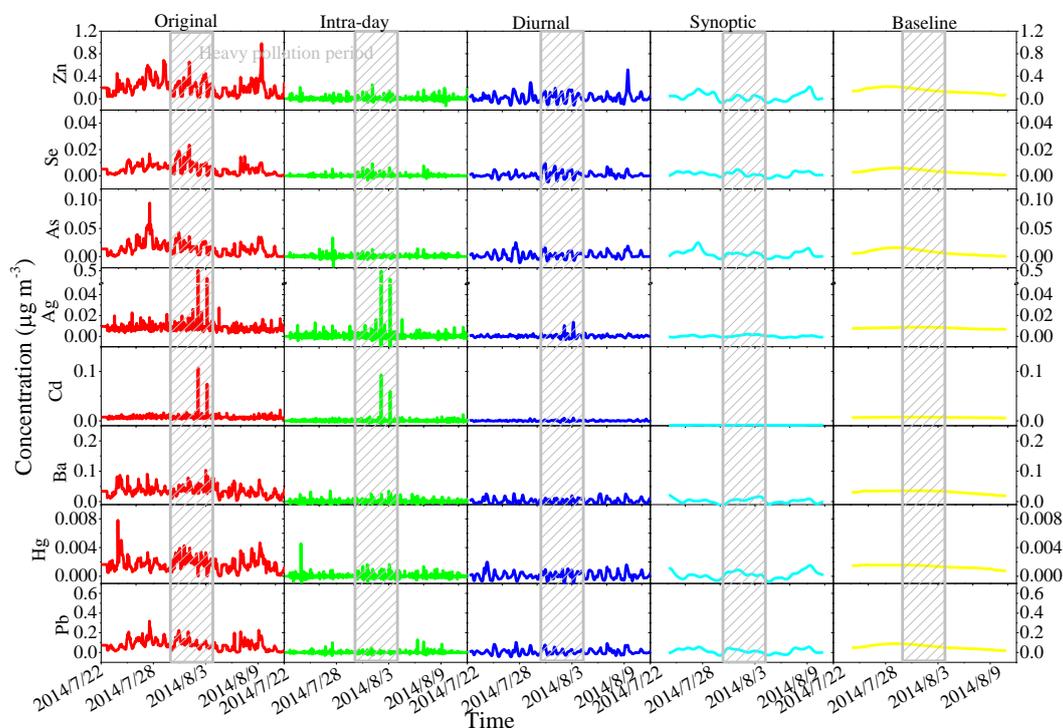
In this work, As and Se obtained moderate fitting, with slop values of 1.2 and 1.2 and r of 0.84 and 0.79, respectively. Poor fitting of Na<sup>+</sup>, Cr, Ag, and Cd mainly linked to poor fitting of some high values. ME2 basically analyzed the typical or average behavior and might show bad fitted of the highest values (Paatero et al., 2003). These high values

might be noisy because they do not continue. High values existed in Intra-day TS components (Figure S2 and S3) and could be handled by filtering intra-day TS components. Comparison of fitting results between original and RI dataset showed that  $r$  values for  $\text{Na}^+$ , Cr, Ag, and Cd were improved from 0.23, 0.23, 0.008, and 0.17 to 0.37, 0.68, 0.56, and 0.58, respectively (Figure S7). The slopes of the four species have also improved that closer to one (Figure S7).

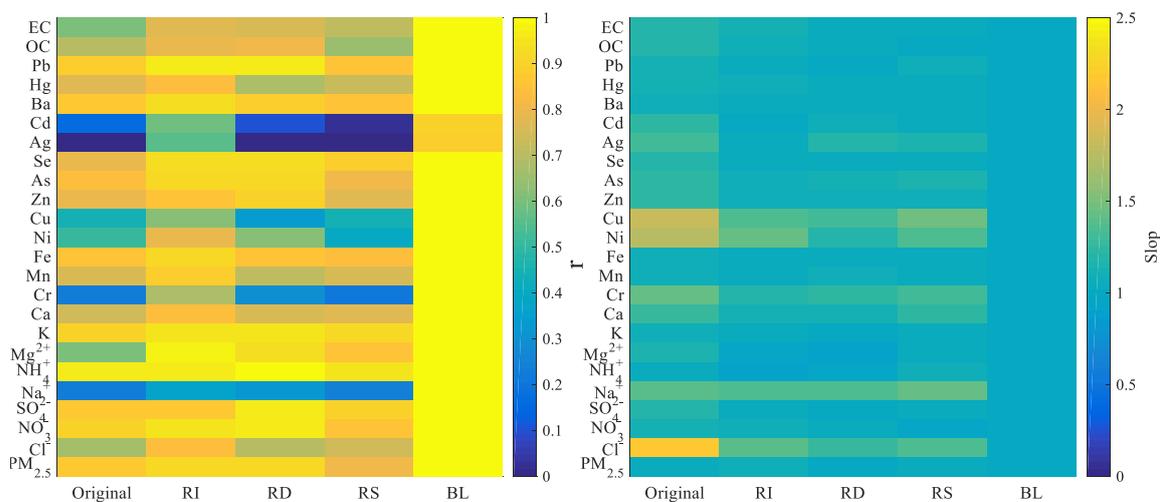
We agree with that it is also difficult to identify point sources even when the point sources existed. On the one hand, small dataset is hard to capture the variation of point source. On the other side, ME2 identifies sources based on temporal variation of sources, and one factor might include multiple sources (Canepari et al., 2009; Lee et al., 2009). It is possible that point source could not be identified by ME2 if the variation of the point source is similar to other sources.



**Figure S2.** Variance of chemical species ( $\text{Cl}^-$ ,  $\text{Na}^+$ ,  $\text{Mg}^{2+}$ ,  $\text{K}^+$ ,  $\text{Cr}$ ,  $\text{Mn}$ ,  $\text{Ni}$ , and  $\text{Cu}$ ) concentrations influenced by intra-day (period less than 12 h), diurnal (12-24 h), synoptic (2-21 days), and baseline (greater than 21 days) temporal-scale (TS) components, for the period of 22 July 2014 to 13 Aug 2014 in Beijing, China. The vertical gray lines demarcate the heavy pollution period.



**Figure S3** Variance of chemical species ( $\text{Zn}$ ,  $\text{Se}$ ,  $\text{As}$ ,  $\text{Ag}$ ,  $\text{Cd}$ ,  $\text{Ba}$ ,  $\text{Hg}$ , and  $\text{Pb}$ ) concentrations influenced by intra-day (period less than 12 h), diurnal (12-24 h), synoptic (2-21 days), and baseline (greater than 21 days) temporal-scale (TS) components, for the period of 22 July 2014 to 13 Aug 2014 in Beijing, China. The vertical gray lines demarcate the heavy pollution period.



**Figure S7.** The performance of ME-2 from five datasets (Beijing site). (Left): the slops between model

and measured concentrations of chemical species and PM<sub>2.5</sub>. (RI: intra-day removed dataset, RD: diurnal removed dataset, RS: synoptic removed dataset, BL: baseline dataset) (Right)The correlation coefficients between modeled and measured concentrations of chemical species and PM<sub>2.5</sub>.

e) The authors acknowledge that the crustal dust source was not resolved in several data sets (RD, RS, BL) (lines 492-495). This contradicts their statement (on lines 506-509) that the proposed methodology improves source identification. See also c) above.

Response: We conclude that filtering intra-day TS components by KZ filter approach can improve the performance of the model. While filtering diurnal and synoptic TS components might lead to an unsuccessful identification of crustal dust because the important information might lose. The performance of the model means the modeling accuracy of species and PM<sub>2.5</sub> in this work. We clarified this point in the revised manuscript.

Revisions made in the manuscript:

“Summarily, receptor data filtering intra-day TS components by KZ filter approach can improve the modeling accuracy of species and PM<sub>2.5</sub> obtained by ME-2.”.

“Receptor data filtering intra-day TS components by KZ filter approach can improve the performance of the model (modeling accuracy of species and PM<sub>2.5</sub>) and produce reasonable source impact results”.

f) Actually, the point in receptor modeling analyses is not removing the noise, but properly quantifying it, so noisy data are downweighted.

Response: Noise included in the significant components can cause a harmful effect and can be handled by downweighting or rejecting components (Escrig. et al., 2009). Noise level varied with variables and might obscure the weakest factors if it been

overweighted (Paatero and Hopke, 2003). Therefore, it is necessary and difficult to specially downweight each data value. In this work, we tried the other way to solve the noise problem that removes the noise and give the same weight to a column data. Receptor data filtering noise (intra-day TS components) by KZ filter approach can improve the modeling accuracy of species and  $PM_{2.5}$  and produce reasonable source impact results, suggesting that filtering noise from the instrument is useful to data analysis.

g) The best input data for applying receptor models is one in which data variability is captured as much as possible, to sample all potential sources impacting a site (including intermittent sources); this also implies sampling for at least a year to include seasonality. The proposed methodology goes in the opposite direction: first, decomposing original data variability into several spectral components (each with lower variability) and, second, analyze each of them separately by receptor models.

Therefore, my recommendation is to reject the manuscript.

Response: We tried firstly the approach that the reviewer proposed. However, many negative values occurred on intra-day, diurnal TS components, which are improperly inputted into the ME2/PMF. We finally used the approach mentioned in the manuscript.

This point has been clarified in the revised manuscript.

“To exam the impact of the four TS components on the source impacts, we tried firstly decomposing original dataset variability into several TS components and then analyzing each of them separately by receptor models. However, too many negative values occurred on intra-day, diurnal TS components, which are not suitable input data

for ME2. We finally designed an approach that removing each TS component from the original dataset respectively and then analyzing them separately by receptor models.”.

As limitations of the experimental condition, it is difficult to obtain one year, continuous, and high-quality dataset. Long-Term observations were conducted in Tianjin, China, from October 03, 2017 to March 16, 2018. PM<sub>2.5</sub>, inorganic ions, OC/EC, and heavy metals were measured by  $\beta$ -ray monitor, an ambient ion monitor (AIM, URG 9000D, URG Corporation, USA) (Shi et al., 2017), OC/EC analyzer (OCEC-100, Focused Photonics Inc, China) and a continuous atmospheric heavy metals monitoring system (AMMS-100, Focused Photonics Inc, China) (Ye et al., 2012), respectively. Twenty chemical species at 1 h time resolution were selected for analysis, including NH<sub>4</sub><sup>+</sup>, Na<sup>+</sup>, Mg<sup>2+</sup>, K<sup>+</sup>, Cl<sup>-</sup>, NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, As, Ca, Tl, Br, Cs, Pb, Se, Cr, Zn, Fe, Mn, OC, and EC. Other species were excluded since they contained more than 40% values below the detection limit.

Each TS component contribution to the total variance of PM<sub>2.5</sub> and the chemical species concentrations in Tianjin was listed in Table S7. For PM<sub>2.5</sub>, OC, EC, and inorganic ions (except Mg<sup>2+</sup> and Na<sup>+</sup>), the diurnal and synoptic TS components had higher relative contributions to the total variance of these species than the intra-day and baseline TS components. Baseline TS component had largest relative contributions to the total variance of Ca, followed by the intra-day, intra-day, and diurnal TS components. For other elements (except Tl and Br), diurnal and synoptic/intra-day TS components had the larger amplitudes and were the larger contributors to the total variance of the concentrations. Dataset observed in Tianjin was processed following the 2.4 section to create RI, RD, RS, RBL datasets. For PM<sub>2.5</sub> and all chemical species, the

largest average concentrations decline occurred when removed baseline TS component from original dataset (Table S8), suggesting baseline TS components dominating the average concentrations of PM<sub>2.5</sub> and chemical species.

The original, RI, RD, and RS datasets were respectively introduced into ME-2 to identify the sources of PM<sub>2.5</sub> (Figures 5 and 6). The regression analysis for the modeled and measured species mass concentrations shown that the slopes and r values of PM<sub>2.5</sub> were ranged from 0.65 to 0.87 and 0.80 to 0.94, respectively (Figure S13). Performance of solutions from RI and BL datasets are better than the solution from the original dataset, due to slopes and r values were closer to 1 than the corresponding results from the original dataset. Comparable performance was obtained from RD, RS, and Original datasets. Six source categories were identified using ME-2 from both original and RD datasets, including crustal dust, vehicle emissions, coal combustion, secondary formation, biomass burning & sea salt, and industrial source. Biomass burning & sea salt was characterized by K<sup>+</sup> and Cl<sup>-</sup> (Tian et al., 2018; Zhu et al., 2018). The industrial source has high loadings of Zn (Monsalve et al., 2018; Ojekunle et al., 2018). While five sources were obtained from both RD and RS dataset except industrial source that mixed with crustal dust. Crustal dust & industrial source was identified by Ca, Fe, and Zn. We tried to separate the crustal dust and industrial source by adding factor number. However, the adding factor did not has a noticeable characteristic of factor profile and cannot be explained by crustal dust or industrial source. For Tianjin dataset, crustal dust was identified and mixed with the industrial source for the RD and RS datasets, suggesting removing diurnal or synoptic TS component affects source identification.

Nitrate and sulfate did not separate from each other after removing the synoptic TS component, because they have similar variation trend. The correlation was 0.70 for the RS dataset, which is close to the results of other datasets (0.80, 0.81, and 0.84 in the original, RI, and RD datasets, respectively).

The correlation coefficient of the time series of source impacts between the original and the RI, RD, and RS datasets was listed in Table S9. Synoptic TS component mainly influences secondary formation impact variation due to a relative low correlation of temporal trend between original and RS datasets. Other four sources, including crustal dust, vehicle emissions, coal combustion, and biomass burning & sea salt, are mainly affected by diurnal and synoptic scale influences. The average impacts of individual source categories on  $PM_{2.5}$  from the datasets with removed TS components (Table 4). Vehicle emissions, crustal dust, coal combustion, biomass burning & sea salt, and industrial source were combined for the TPS. For the entire sampling period, the impacts of TPS obtained from the original, RI, RD, and RS datasets were similar to each other, ranging from 33.8 to 38.2  $\mu g m^{-3}$ . The TSS (secondary formation) solutions from the original and RI datasets exhibited similar source impacts, accounting for about 33  $\mu g m^{-3}$ , which was higher than the solution from the RD (27.3  $\mu g m^{-3}$ ) and RS dataset (23.0  $\mu g m^{-3}$ ). The RBL dataset, including about 45% negative values, was analyzed by PCA (Table S10). Four factors, including crustal dust (44.9%), secondary formation (8.7%), industrial source & coal combustion (6.4%), and vehicle (5.4%), were extracted and accounted for 65.4% of the total variance. In addition, ME-2 was applied to the baseline dataset and identified crustal

dust, vehicle emissions, coal combustion, and secondary formation (Figure S14 and Table S11). The average TPS and TSS impacts on PM<sub>2.5</sub> mass concentrations were 29.6 μg m<sup>-3</sup> (58%) and 21.3 μg m<sup>-3</sup> (42%) respectively.

Seasonal variations of source impacts were investigated in Tianjin, as shown in Table 4. Winter period in this work included 15 days in March 2018 because 15 days is too short to present spring. The TPS impacts derived from the original, RI, RD, and RS datasets were relatively stable, ranging from 31.9 to 35.8 μg m<sup>-3</sup> during fall and 33.6 to 37.1 μg m<sup>-3</sup> during winter (Table 4). The TSS impacts decreased from 27.6 (original dataset) to 20.5 μg m<sup>-3</sup> (RS dataset) during the fall and from 39.0 (original dataset) to 26.6 μg m<sup>-3</sup> (RS dataset) during winter. A more significant decrease in secondary source impact during winter suggested that the synoptic TS component had a more significant impact on the secondary source during winter than during the fall in this work.

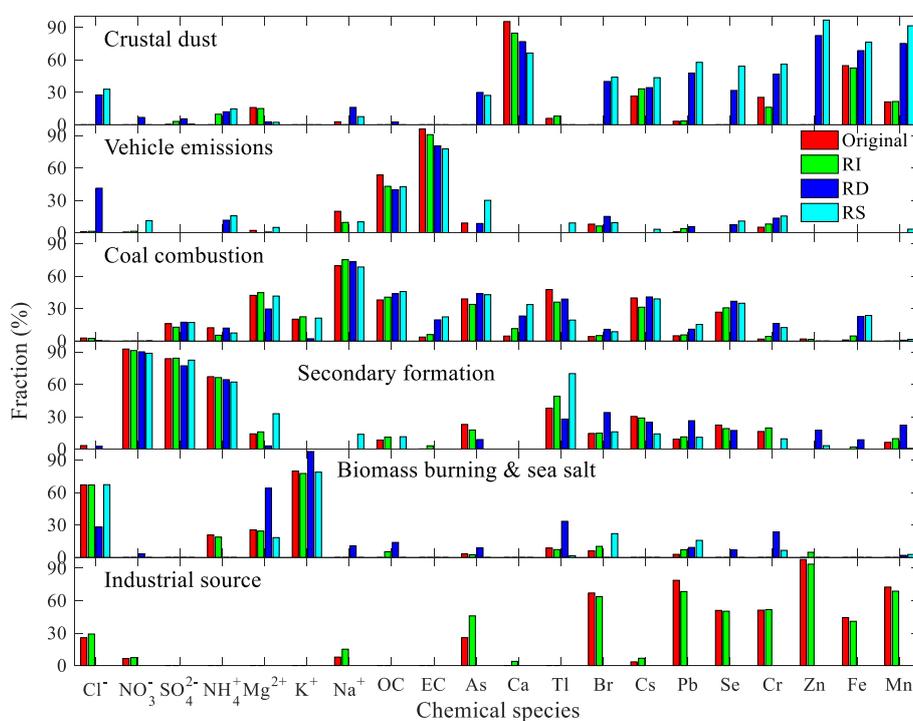


Figure 5. The influence of different TS components on source determination (Tianjin site). The industrial source was not identified from the RD and RS datasets. RBL dataset was investigated by PCA analysis instead of ME-2 due to the dataset has some negative values.

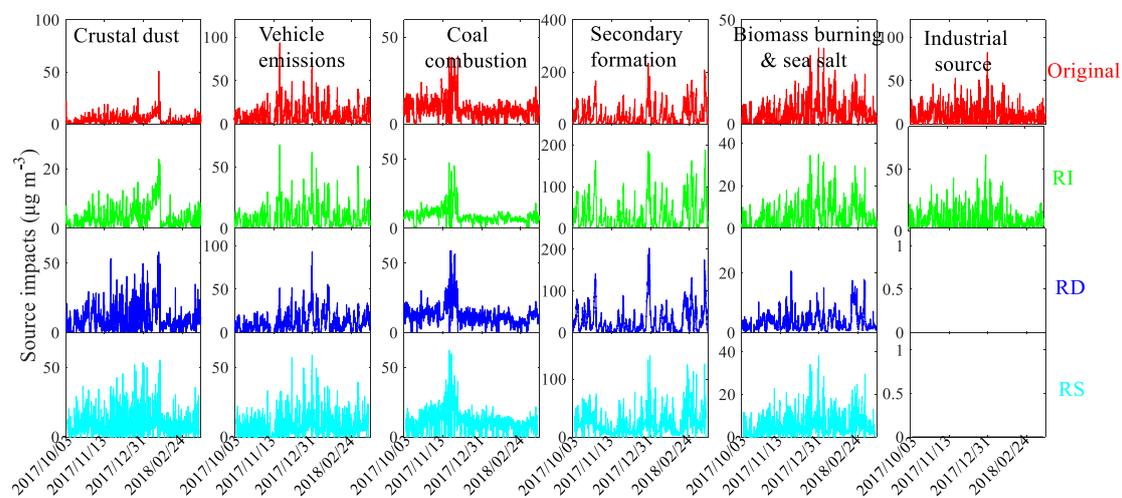


Figure 6. Source contributions to PM<sub>2.5</sub> for each source (vertical columns) and each dataset (horizontal rows) (Tianjin site). The blanks mean that the source has not been identified.

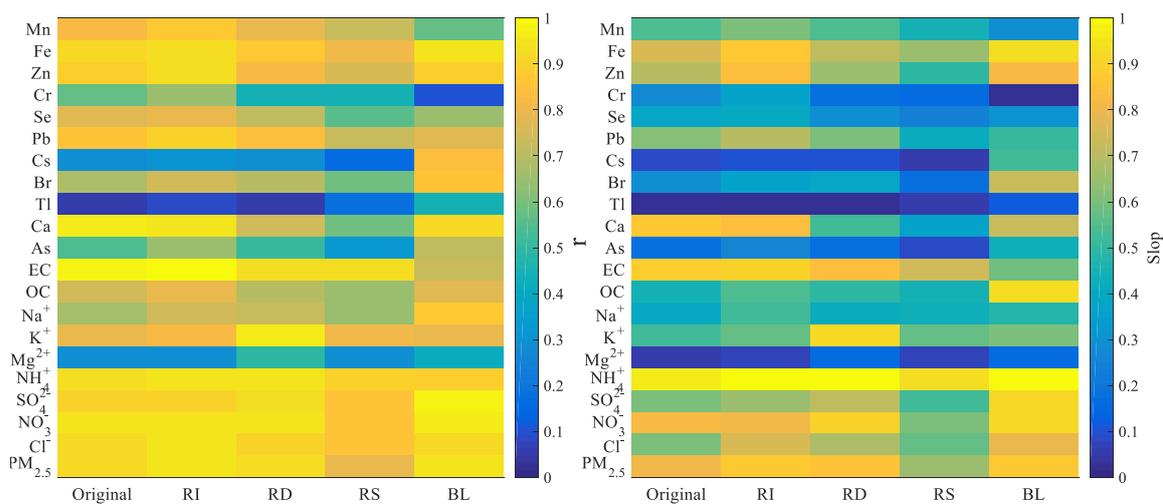
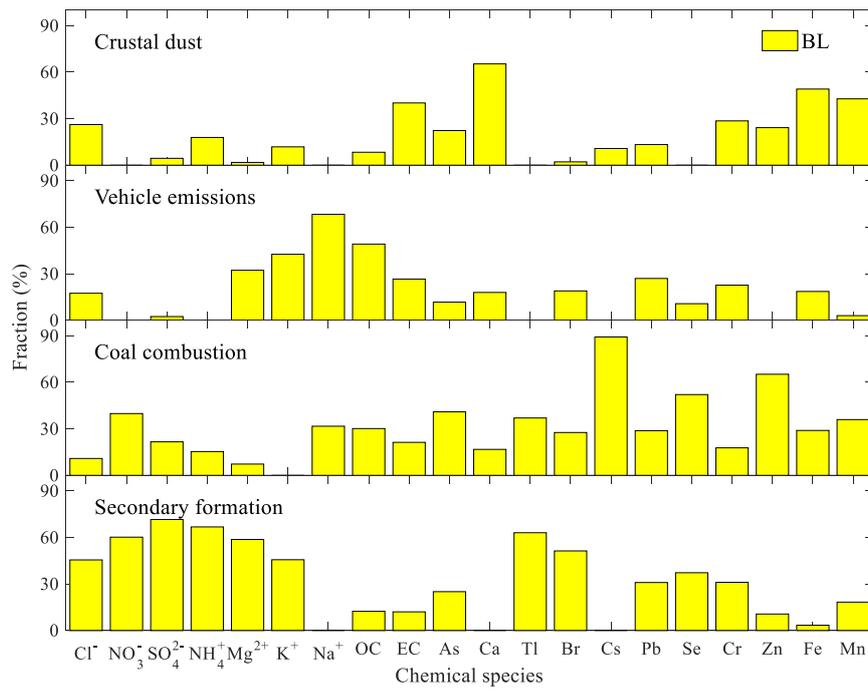


Figure S13. The performance of ME-2 from five datasets (Tianjin site). (Left): the slops between model and measured concentrations of chemical species and PM<sub>2.5</sub>. (RI: intra-day removed dataset, RD: diurnal removed dataset, RS: synoptic removed dataset, BL: baseline dataset) (Right)The correlation coefficients between modeled and measured concentrations of chemical species and PM<sub>2.5</sub>.



**Figure S14.** The factor profiles obtained from ME-2 from baseline dataset (Tianjin site).

**Table 4.** Average source contributions to PM<sub>2.5</sub> (μg m<sup>-3</sup>) estimated by ME-2 from Tianjin for the original, RI, RD, and RS datasets during the entire sampling period.

		Crustal dust	Vehicle emission	Coal combustion	Biomass burning & sea salt	Industrial source	TPS <sup>a</sup>	Secondary formation (TSS <sup>b</sup> )
During the entire sampling period	Original	4.3 <sup>c</sup> (6%)	10.2 (14%)	8.5 (12%)	5.0 (7%)	10.2 (14%)	<b>38.2</b> <b>(54%)</b>	<b>32.5</b> <b>(46%)</b>
	RI	3.8 (6%)	9.2 (13%)	8.9 (13%)	5.9 (9%)	8.6 (12%)	<b>36.5</b> <b>(53%)</b>	<b>32.6</b> <b>(47%)</b>
	RD	9 (14%)	10.9 (17%)	12.8 (20%)	3.5 (6%)		<b>36.2</b> <b>(57%)</b>	<b>27.3</b> <b>(43%)</b>
	RS	9.2 (16%)	8.0 (14%)	11.5 (20%)	5.1 (9%)		<b>33.8</b> <b>(59%)</b>	<b>23.0</b> <b>(41%)</b>
Fall <sup>d</sup>	Original	3.7 (6%)	8.7 (14%)	10.4 (16%)	3.0 (5%)	10.0 (16%)	35.8 (56%)	27.6 (44%)
	RI	2.8 (5%)	7.4 (12%)	11.2 (18%)	3.2 (5%)	8.9 (14%)	33.5 (55%)	27.9 (45%)
	RD	7.9 (14%)	6.4 (11%)	15.2 (27%)	2.9 (5%)		32.4 (58%)	23.7 (42%)
	RS	8.0 (15%)	5.6 (11%)	14.6 (28%)	3.7 (7%)		31.9 (61%)	20.5 (39%)
Winter	Original	4.1 (5%)	10.7 (14%)	7.6 (10%)	5.2 (7%)	9.5 (12%)	37.1 (49%)	39.0 (51%)
	RI	3.9 (5%)	10.2 (14%)	7.6 (10%)	6.3 (8%)	8.1 (11%)	36.0 (48%)	39.4 (52%)
	RD	8.9 (13%)	13.0 (19%)	11.6 (17%)	3.7 (5%)		37.1 (53%)	32.7 (47%)
	RS	9.7 (16%)	9.4 (16%)	9.2 (15%)	5.3 (9%)		33.6 (56%)	26.6 (44%)

<sup>a</sup>TPS is the total contributions of crustal dust, vehicle emissions, coal combustion, biomass burning & sea salt, and industrial source. <sup>b</sup>TSS is the total contributions of secondary formation and nitrate source. <sup>c</sup>The data in the parentheses and outside the parentheses are the absolute values of average source contribution (μg m<sup>-3</sup>) and percentages of average source contribution (%), respectively. <sup>d</sup>Fall included October and November, and winter included December, January, February, and March (15 days).

**Table S7.** Relative contributions (%) of the different TS components to the total variance of chemical species concentrations (Tianjin site).

	Intra-day (%)	Diurnal (%)	Synoptic (%)	Baseline (%)
PM <sub>2.5</sub>	6	<b>25</b>	<b>60</b>	9
Cl <sup>-</sup>	12	<b>44</b>	<b>38</b>	6
NO <sub>3</sub> <sup>-</sup>	4	<b>23</b>	<b>68</b>	6
SO <sub>4</sub> <sup>2-</sup>	4	<b>25</b>	<b>59</b>	12
NH <sub>4</sub> <sup>+</sup>	3	<b>18</b>	<b>62</b>	17
Mg <sup>2+</sup>	9	24	<b>40</b>	<b>27</b>
K <sup>+</sup>	14	<b>32</b>	<b>44</b>	10
Na <sup>+</sup>	6	9	<b>17</b>	<b>69</b>
OC	14	<b>34</b>	<b>48</b>	4
EC	10	<b>33</b>	<b>49</b>	8
As	<b>34</b>	<b>43</b>	21	2
Ca	<b>24</b>	20	<b>24</b>	<b>33</b>
Tl	<b>32</b>	7	2	<b>59</b>
Br	<b>26</b>	<b>53</b>	19	2
Cs	16	13	<b>18</b>	<b>53</b>
Pb	23	<b>39</b>	<b>33</b>	6
Se	28	<b>34</b>	<b>30</b>	8
Cr	<b>48</b>	<b>29</b>	14	9
Zn	<b>26</b>	<b>44</b>	23	6
Fe	27	<b>31</b>	<b>29</b>	13
Mn	<b>29</b>	<b>33</b>	<b>29</b>	9

**Table S8.** Average concentrations of PM<sub>2.5</sub> and chemical species for five datasets (Tianjin site).

	Original	RI	RD	RS	RBL
PM <sub>2.5</sub> ( $\mu\text{g m}^{-3}$ )	69.7	68.1	62.8	57.4	21.1
Cl <sup>-</sup> ( $\mu\text{g m}^{-3}$ )	3.8	3.6	3.1	3.1	1.7
NO <sub>3</sub> <sup>-</sup> ( $\mu\text{g m}^{-3}$ )	14.3	13.9	12.1	9.5	7.5
SO <sub>4</sub> <sup>2-</sup> ( $\mu\text{g m}^{-3}$ )	7.4	7.3	6.7	5.8	2.6
NH <sub>4</sub> <sup>+</sup> ( $\mu\text{g m}^{-3}$ )	13.9	13.7	12.9	11.6	3.6
Mg <sup>2+</sup> ( $\mu\text{g m}^{-3}$ )	0.05	0.05	0.04	0.04	0.01
K <sup>+</sup> ( $\mu\text{g m}^{-3}$ )	0.9	0.9	0.8	0.8	0.3
Na <sup>+</sup> ( $\mu\text{g m}^{-3}$ )	0.9	0.9	0.9	0.9	0.05
OC ( $\mu\text{g m}^{-3}$ )	7.3	7.1	6.7	6.6	1.4
EC ( $\mu\text{g m}^{-3}$ )	3.9	3.7	3.4	3.2	1.5
As (ng m <sup>-3</sup> )	5.7	4.5	4.4	4.5	3.8
Ca (ng m <sup>-3</sup> )	312.8	289.0	283.4	281.8	84.5
Tl (ng m <sup>-3</sup> )	2.8	2.1	2.6	2.7	0.9
Br (ng m <sup>-3</sup> )	33.0	28.5	26.0	27.0	17.9
Cs (ng m <sup>-3</sup> )	9.4	8.3	8.6	8.0	3.4
Pb (ng m <sup>-3</sup> )	63.4	57.1	53.4	52.7	27.4
Se (ng m <sup>-3</sup> )	6.1	5.0	4.8	4.8	3.8
Cr (ng m <sup>-3</sup> )	6.1	4.7	4.7	5.2	3.8
Zn (ng m <sup>-3</sup> )	270.7	235.4	211.5	222.9	145.1
Fe (ng m <sup>-3</sup> )	600.8	545.5	528.6	538.4	191.0
Mn (ng m <sup>-3</sup> )	42.9	36.3	33.6	33.2	26.1

**Table S9.** Correlation coefficients (3378 samples) between original and intra-day removed dataset, diurnal removed dataset, and synoptic removed dataset for source contributions (Tianjin site).

	RI	RD	RS
Crustal dust <sup>a</sup>	0.89**	0.69**	0.52**
Vehicle emission	0.96**	0.82**	0.76**
Coal combustion	0.88**	0.68**	0.59**
Secondary formation	0.99**	0.93**	0.75**
Biomass burning & sea salt	0.97**	0.74**	0.81**
Industrial source <sup>b</sup>	0.90**		

<sup>a</sup>Crustal dust from RD and RS datasets mixed with industrial source. <sup>b</sup>Only ME-2 from original and RI datasets identified the Industrial source. \*\*Significant correlation at 0.01 level.

**Table S10.** The results obtained from PCA from RBL dataset (Tianjin site).

Components	Factor 1	Factor 2	Factor 3	Factor 4
Cl <sup>-</sup>	0.33	0.20	<b>0.78</b>	0.23
NO <sub>3</sub> <sup>-</sup>	0.26	<b>0.82</b>	0.10	0.27
SO <sub>4</sub> <sup>2-</sup>	0.17	<b>0.82</b>	0.22	0.31
NH <sub>4</sub> <sup>+</sup>	0.26	<b>0.73</b>	0.34	0.28
Mg <sup>2+</sup>	0.08	0.16	-0.10	<b>0.83</b>
K <sup>+</sup>	0.21	0.26	0.32	0.77
Na <sup>+</sup>	0.28	0.23	0.21	0.17
OC	0.13	0.24	0.40	<b>0.65</b>
EC	0.24	0.20	<b>0.61</b>	<b>0.51</b>
As	0.10	<b>0.62</b>	0.41	0.07
Ca	<b>0.75</b>	0.06	0.21	0.04
Tl	0.02	-0.17	0.04	0.04
Br	0.18	0.37	<b>0.70</b>	0.01
Cs	0.09	0.34	0.07	-0.10
Pb	0.39	0.43	<b>0.60</b>	0.21
Se	0.47	<b>0.57</b>	0.30	0.11
Cr	<b>0.73</b>	0.20	0.07	0.23
Zn	0.47	0.18	<b>0.71</b>	0.07
Fe	<b>0.85</b>	0.21	0.34	0.08
Mn	<b>0.80</b>	0.29	0.28	0.12
Variance contribution (%)	44.9	8.7	6.4	5.4

**Table S11.** Average source contributions to PM<sub>2.5</sub> (μg m<sup>-3</sup>) estimated by ME-2 from the BL datasets (Tianjin site).

	Crustal dust	Vehicle emission	Coal combustion	TPS <sup>a</sup>	Secondary formation(TSS <sup>b</sup> )
During the entire sampling period	7.3 <sup>c</sup> (14%)	12.0 (23%)	10.4 (20%)	<b>29.6</b> <b>(58%)</b>	<b>21.3</b> <b>(42%)</b>
Fall	3.4 (7%)	12.8 (28%)	16.5 (36%)	<b>32.7</b> <b>(71%)</b>	<b>13.3</b> <b>(29%)</b>
Winter <sup>d</sup>	8 (15%)	9.4 (17%)	10.5 (19%)	<b>28</b> <b>(51%)</b>	<b>26.4</b> <b>(49%)</b>

<sup>a</sup>TPS is the total contributions of crustal dust, vehicle emissions, and coal combustion. <sup>b</sup>TSS is the contributions of secondary formation. <sup>c</sup>The data in the parentheses and outside the parentheses are the absolute values of average source contribution (μg m<sup>-3</sup>) and percentages of average source contribution (%), respectively. <sup>d</sup>The winter included 15 days in March 2018.

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