Responses to Comments by Reviewer #1, Reviewer #2 and Reviewer #3

Ms. Ref. No.: acp-2018-1234

Title: High Time Resolution Source Apportionment of $PM_{2.5}$ in Beijing with Multiple Models

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We are grateful for the helpful comments by all the reviewers. Based on these comments, we have carefully revised the manuscript. The response to each comment is listed below. The original comments from reviewers are in *blue and italic*, our replies are in normal font and the tracked changes in the revised manuscript are in *red and italic*. The page and line number refers to the revised manuscript without marks.

Reviewer #1 Comment No.1: *I suggest the authors to reconstruct the hourly PM2.5 mass based on the online chemical components (e.g. ions, OC/EC, and elements). The convert factor of OC to OM should be related to the sources rather than 1.8 which obtained in the regional parks in USA. Thus, the relationship between OC and EC should be discussed firstly.*

Response to reviewer comment NO.1: We agree with the reviewer that the convert factor of OC to OM (R_{oc}) should be related to the sources. According to previous studies, R_{oc} would be relatively higher for samples influenced by biomass burning event (Malm et al., 2007; Poirot and Husar., 2004; Turpin and Lim., 2001). For example, Turpin and Lim (2001) have reported R_{oc} values ranging from 2.2 to 2.6 for samples with impacts from biomass burning. Poirot and Husar (2004) found good agreement between measured and reconstructed fine mass by applying a R_{oc} factor of 1.8 during a biomass burning event. Besides, R_{oc} of urban aerosol tends to be lower than that of nonurban aerosol because nonurban areas are likely to have higher contributions of both biogenic and secondary anthropogenic sources than observed in urban areas (Turpin and Lim., 2001). Turpin and Lim (2001) recommend a factor of 1.6±0.2 for urban organic aerosols, a factor of 2.1±0.2 for nonurban organic aerosols. The R_{oc} of 1.6 has also been used in studies of urban areas in China (Cao et al., 2007; Huang et al., 2017). According to the source apportionment results in our study, the contribution of biomass burning source was not very significant during the sampling period (9% on average, see Figure 3 in the manuscript), and the sampling site is more influenced by the urban sources of Beijing. Therefore, we change the R_{oc} from 1.8 to 1.6 which is more fit for our case. The related description has been revised as follows (see Page 6, Line 151):

Organic matter (OM) was calculated as OM= 1.6 ×OC (Turpin and Lim, 2001).

After the revision of R_{oc}, we reconstructed the hourly $PM_{2.5}$ mass based on the online chemical components (see Figure 1). The correlation between the measured and reconstructed $PM_{2.5}$ is good with the slope close to 1.0 and the R^2 over than 0.9 ($p< 0.05$, $n=1099$). Figure 1 has also been added in the supplementary materials as **Figure S6**.

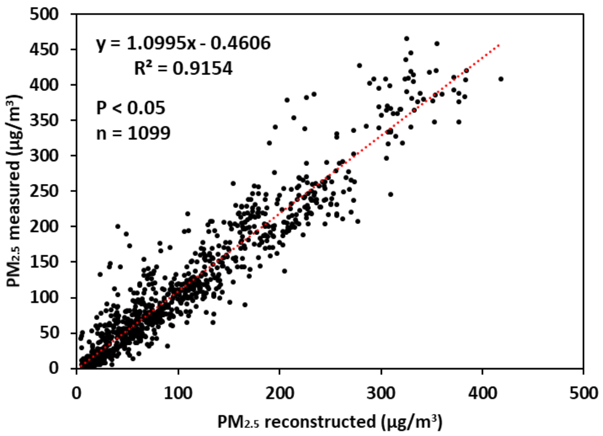

Figure 1 Correlation of measured and reconstructed $PM_{2.5}$ mass

Reference

[1] Cao, J. J., Lee, S. C., Ho, K. F., Zou, S. C., Fung, K., Li, Y., Watson, J. G., and Chow, J. C.: Spatial and seasonal variations of atmospheric organic carbon and elemental carbon in Pearl River Delta Region, China, Atmos. Environ., 38, 4447-4456, 2004.

[2] Huang, X., Liu, Z., Liu, J., Bo, H., and Wang, Y.: Chemical characterization and source identification of $PM_{2.5}$ at multiple sites in the Beijing-Tianjin-Hebei region, China, Atmos. Chem. Phys., 17, 12941-12962, 2017.

[3] Lim, H.J., and Turpin, B. J.: Origins of primary and secondary organic aerosol in Atlanta: Results of time-resolved measurements during the Atlanta supersite experiment, Environ. Sci. Technol., 36, 4489-4496, 2002.

[4] Malm, W. C., and Hand, J. L.: An examination of the physical and optical properties of aerosols collected in the IMPROVE program, Atmos. Environ., 41, 3407-3427, 2007.

[5] Poirot, R.L., and Husar, R.B.: Chemical and physical characteristics of wood smoke in the northeastern US during July

2002: impacts from Quebec forest fires. A&WMA Specialty Conference: Regional and Global Perspectives on Haze: Causes, Consequences and Controversies, Asheville, NC, 2004.

Reviewer #1 Comment No.2: *I suggest the authors to present the data quality assurance and control in more detail in section 2.2. For example, will the reconstructed PM2.5 match the measured PM2.5? What are the relationships between the elements (such as S, K, Cl and Ca) measured by Xact 625 and their water soluble form (SO42-, K+, Cl- and Ca2+) measured by IGAC. Moreover, the EC concentrations obtained by Sunset are also suggested to compare with the BC concentrations measured by MAAP or AE-31 etc. There are quite important for the source apportionment.*

Response to reviewer comment NO.2: According to *Response to reviewer comment NO.1*, the reconstructed and the measured $PM_{2.5}$ mass agree well with the slope of 1.0 and the R^2 over than 0.9 (see Figure 1). About quality control, we have added more detailed information in the supplementary materials including the concentration of the internal standard (Pd) of Xact and the internal standard (LiBr) of IGAC. As shown in Figure 2 (**Figure S1** in the supplementary materials), for IGAC the internal standard (LiBr) was added continuously to each sample and analyzed by the IC system during the analysis to check the stability of the IGAC instrument. During the sampling period, the mean concentrations of Li^+ and Br^- were within the range of three standard deviations, suggesting a stable condition of the IGAC. As shown in Figure 3 (**Figure S3** in the supplementary materials), for the Xact a Pd rod was used as automatic internal quality control to check the performance of the instrument on a daily basis and the mean concentration of Pb was within the range of three standard deviations during the sampling period.

We have collected offline samples simultaneously with online measurement at the same site and compared the online concentration of ion species measured by IGAC with those measured by offline ion chromatography (Zhang et al., under review). The results are shown in Figure 4. The correlation of SO_4^{2-}, NO_3^-, Cl^- and Na^+ between online and offline measurement is relatively good (R^2>0.8) while the correlation of Ca^{2+}, K^+ and Mg^{2+} is relatively poor. We also compared the online concentration of elemental species measured by Xact with those measured by offline ICP-MS and found better correlation (R^2>0.9) for most species such as K, Cr, Mn, Fe, Cu, Ni, Zn, As and Pb (Zhang et al., under review). Therefore, in our study, after quality check, we use 12 species measured by Xact while only use the SO_4^{2-}, NO_3^-, Cl^- and Na^+ concentration measured by IGAC for further discussion. Based on the comparison between online and offline measurements, K and Ca show better correlation than K^+ and Ca^{2+}, therefore, only K and Ca data by Xact are used in the analysis in this study.

During the sampling period, we also measured BC by AE33 at the same site but we did not use MAAP. However, we used a Sunset OC/EC analyzer at the same time. The correlation of BC measured by AE33 and EC measured by Sunset OC/EC analyzer is shown in Figure 5. Relatively high correlation could be found between the two instruments with the slope of 1.0 and the R^2 over than 0.9.

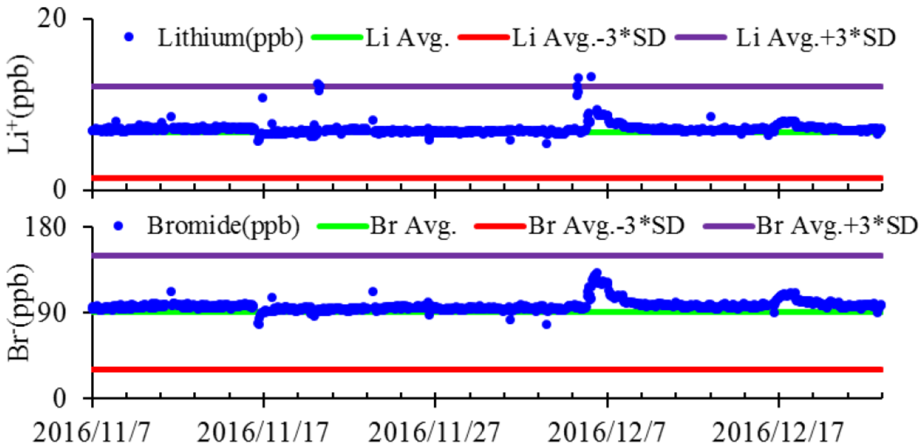

Figure 2. Concentration of the internal standard (LiBr) of IGAC

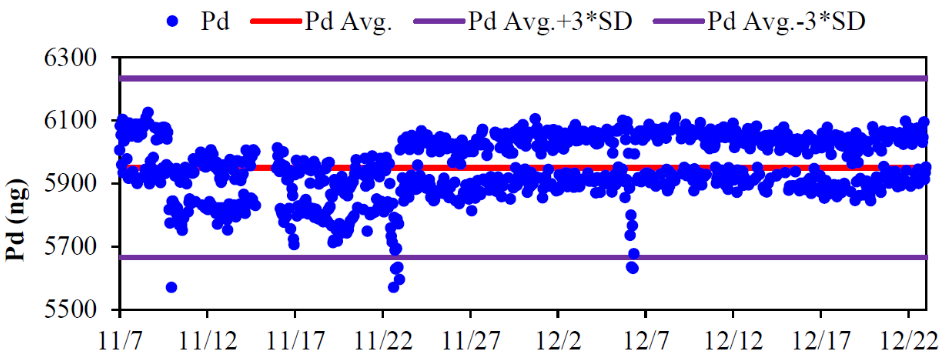

Figure 3. Concentration of the internal standard (Pd) of Xact

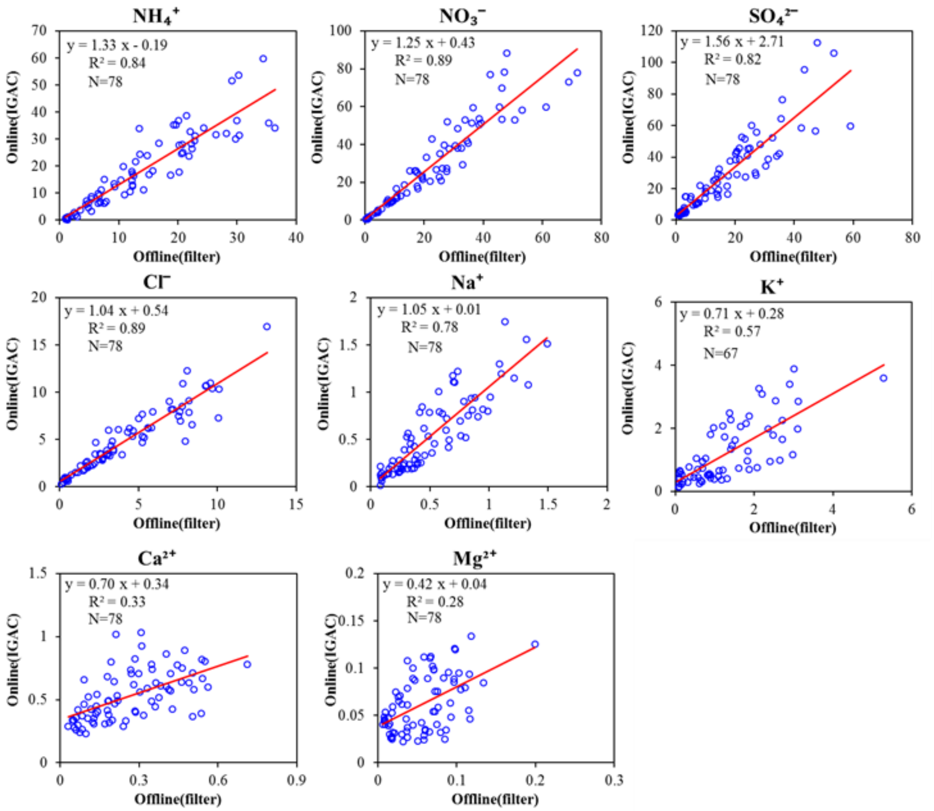

Figure 4. Correlation between online IGAC and offline method for ion species (Zhang et al., under review)

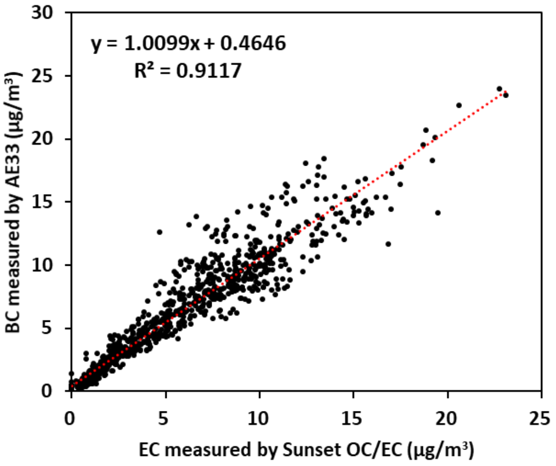

Figure 5. Correlation of BC measured by AE33 and EC measured by Sunset OC/EC analyzer

Reference

[1] Zhang, B. Y., Tian, Z., Yan, C. Q., Li, X. Y., Yu, J. T., Wang, S. X., Liu, B. X., Zheng, M.: Comparison of water-soluble inorganic ions and trace metals in $PM_{2.5}$ between online and offline measurements in Beijing during winter, under review.

Reviewer #1 Comment NO.3: *I suggest the authors to summarize the mass concentrations of chemical components in Table rather than in Figure for different episodes. It is important to judge the online data quality when compared to the previous studies in Beijing.*

Response to reviewer comment NO.3: According to the reviewer's suggestions, the mass concentration of chemical components has been summarized in Table 1 and Table 2 as follows, and also added in the supplementary material as Table S1 and Table S4:

Table 1 Statistical summary of identified species of $PM_{2.5}$ in the entire sampling period

n=1099

	Mean	**Std.**	**Max**	**Min**	**Detection limit**	**BDL%**
OC/EC	**µg/m³**					**%**
OC	20.8	17.0	89.9	1.1	0.4	-
EC	5.6	4.4	23.1	0.2	0.1	4.3
SIA	**µg/m³**					**%**
SO_4^{2-}	23.5	20.8	95.8	0.04	0.04	0.21
NO_3^-	22.0	23.3	104.7	0.03	0.03	0.1
NH_4^+	14.0	14.7	66.6	0.04	0.05	1.4
Na^+	0.39	0.32	1.89	0.02	0.04	8.5
Cl^-	4.89	4.19	27.6	0.05	0.05	0.1
Metal	**µg/m³**				**ng/m³**	**%**
K	1.49	1.17	5.28	0.10	2.366	-
Fe	0.769	0.541	2.22	0.015	0.759	-
Ca	0.384	0.277	2.08	0.001	0.902	-
Zn	0.286	0.261	1.85	0.005	0.231	-
Pb	0.107	0.091	0.469	0.004	0.218	-

Mn	0.058	0.046	0.210	0.001	0.283	-
Ba	0.035	0.023	0.160	0.002	0.945	-
Cu	0.027	0.024	0.171	0.002	0.267	-
As	0.022	0.021	0.084	0.000	0.114	17
Cr	0.010	0.010	0.110	0.000	0.288	11
Se	0.008	0.008	0.046	0.000	0.141	5.2
Ni	0.002	0.002	0.044	0.000	0.226	3.4

* BDL% refers to the percentage of data below the detection limit

* The unit of the detection limit of each metal is ng/m^3

* ‘-’ means that all data are above the detection limit

Table 2 Average concentration of $PM_{2.5}$ and identified species in different haze and non-haze periods

Unit: μg m^{-3}

Average Conc.	**EP1**	**EP2**	**EP3**	**EP4**	**NH1**
	n=102	n=95	n=117	n=131	n=78
$PM_{2.5}$	97.7±70.7	143.8±119.1	115.3±108.6	241.8±115.5	18.8±20.7
OC	19.1±10.7	24.7±18.7	23.1±21.1	40.3±14.5	3.33±2.85
EC	4.1±2.7	7.1±5.1	5.6±4.3	11.0±3.7	0.9±0.6
SO_4^{2-}	18.6±10.9	25.1±20.4	23.4±19.7	53.3±19.2	4.43±3.89
NO_3^-	19.9±14.6	18.9±16.0	23.3±23.3	56.8±24.8	2.81±3.69
NH_4^+	13.3±8.36	12.6±11.6	13.3±13.2	36.4±16.7	2.42±3.80
Na^+	0.37±0.23	0.35±0.28	0.49±0.41	0.69±0.23	0.10±0.14
Cl^-	5.07±3.05	5.17±3.71	5.66±5.87	8.15±3.41	0.97±1.10
K	2.55±1.33	1.21±0.90	1.23±1.08	2.53±0.78	0.410±0.410
Fe	1.26±0.58	0.724±0.568	0.641±0.480	1.06±0.25	0.144±0.133
Ca	0.530±0.250	0.479±0.447	0.356±0.249	0.360±0.148	0.054±0.033
Zn	0.442±0.242	0.242±0.197	0.252±0.289	0.409±0.170	0.060±0.076
Pb	0.182±0.105	0.087±0.071	0.090±0.084	0.185±0.063	0.024±0.028
Mn	0.093±0.050	0.049±0.038	0.049±0.045	0.082±0.021	0.011±0.012
Ba	0.047±0.016	0.038±0.032	0.032±0.022	0.048±0.012	0.006±0.005

Cu	0.028±0.015	0.023±0.021	0.028±0.026	0.042±0.017	0.007±0.009
As	0.031±0.019	0.019±0.019	0.021±0.023	0.040±0.013	0.003±0.006
Cr	0.019±0.022	0.007±0.009	0.008±0.011	0.014±0.010	0.001±0.001
Se	0.012±0.007	0.006±0.005	0.007±0.007	0.018±0.007	0.001±0.001
Ni	0.003±0.002	0.002±0.001	0.002±0.002	0.004±0.004	0.0006±0.0004

Reviewer #1 Comment NO.4: *I don't think the explanation for the six factors in section 3.2 is reasonable, and it is not suitable for PMF to resolve PM sources with too many episode cases. The uncertainties of source contributions would be huge. For example, the contributions of the dominant sources during the episode cases would be overestimated during the non- episode period.*

Response to reviewer comment NO.4: This is a very helpful comment. We think that the reviewer may concern that the major source types might change from haze episodes to non-haze periods and we agree that the uncertainties of PMF would be big if the main source types change during the sampling period. However, the sampling period of our study was within the heating period in winter. In Beijing, source types would change with season (e.g., more dust in spring), but within the same season, it does not show a significant change (Lv et al., 2016).

We agree that this is an important concern. To test this, we separate the data in haze episodes and non-haze periods into two different input files and apply PMF for source apportionment separately. The result based on haze episode data is represented by EP-PMF, the result based on non-haze period data is represented by NH-PMF and the previous PMF result based on all the input data is showed as PRE-PMF. The average source contributions in EP1, EP4 and NH1, NH2 calculated by EP-PMF, NH-PMF and PRE-PMF are shown in Figure 6. It could be seen that the source contributions in different haze episodes and non-haze periods did not change significantly based on different input of PMF.

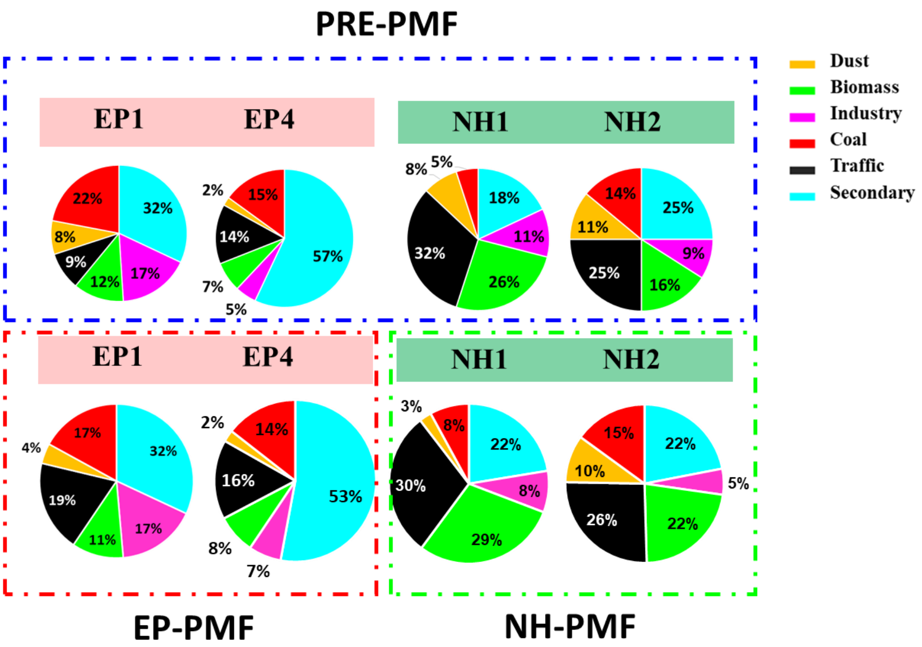

Figure 6. The average source contributions in EP1, EP4 and NH1, NH2 calculated by EP-PMF, NH-PMF and PRE-PMF

Reference

[1] Lv, B., Zhang, B., and Bai, Y.: A systematic analysis of $PM_{2.5}$ in Beijing and its sources from 2000 to 2012, Atmos. Environ., 124, 98-108, 2016.

Reviewer #1 Comment NO.5: *I also don't think the identified source profiles are reasonable. Firstly, K can be also originated from dust. Why not use K+? More fraction of K was showed in the industrial source rather than biomass burning. Secondary, industrial sources should be explained in detail. Why only little fractions of OC and EC in this source profile? Thirdly, why large fraction of Cl- was found in traffic emission? Lastly, why large fraction of Ni was found in biomass burning source?*

Response to reviewer comment NO.5: According to *Response to reviewer comment NO.2*, only the concentration of SO_4^{2-}, NO_3^-, Cl^- and Na^+ measured by IGAC were used for further discussion in the manuscript after performing comparison between online and offline measurement data. We agree with the reviewer that K^+ is a better indicator for biomass burning than K. However, besides poor agreement between online and offline data for K^+, a certain fraction (about 25%) of K^+ measured by IGAC was below or close to its detection limit (0.05 μg/m^3) during the sampling period. Therefore, K^+ was not included in the input of PMF and K was used instead. There have been studies in which K was used as a tracer for biomass burning source when K^+ was not available (Song et al., 2006; Yu et al., 2013; Zheng et al., 2014; Gao et al., 2016; Zíková et al., 2016). The correlation of K^+ concentration with the biomass burning source identified by PMF model are shown in Figure 7. It could be seen that K^+ exhibited a relatively good correlation with the contribution of biomass burning source from PMF with K as input data, with R^2 over 0.6.

In our PMF results, industrial source and biomass burning source were the two major sources of K in Beijing, China. This conclusion could be supported by the local source profiles in China shown in Figure 8 (Dai et al., 2015; Cao et al., 2015; Ma et al., 2015; Wang et al., 2009; Wen et al., 2009; Wu et al., 2016; Zhang et al., 2012; Zheng et al., 2013). As shown in Figure 8(c), the average fraction of K in industrial source profiles (including coal-fired power plant and boiler, steel and iron plant in Beijing, Shanghai and Jilin) was similar with that in biomass burning source profiles (including wheat, corn and rice), which is about 8%. Moreover, in some industrial source profiles the fraction of K exceeded that in biomass burning source profiles. The potential reasons of K in industrial source including that coal was used in some power plants and the use of potassium feldspar in glass and fertilizer industry (Ma et al., 2010). Therefore, this might be the reason that K is found in industrial source identified by the PMF analysis. As shown in Figure 8(a) and (b), the fraction of OC and EC in industrial profiles were much lower than those in coal combustion source and biomass burning source, which might be attributed to the sufficient pretreatment process and higher combustion efficiency in industries (Wang et al., 2009; Zheng et al., 2013). Therefore, lower OC and EC are seen in industry factor of our PMF results. In secondary source profile, EC is low because it is mostly emitted from primary sources including traffic source. OC is relatively higher than EC due to the formation of secondary organic carbon.

For the third question, we agree with the reviewer that Cl^- has not been used as a specific tracer for traffic source in previous studies. In our study, large fraction of Cl^- was observed in the profiles of coal combustion and biomass burning source which corresponded well with previous studies (Frigge et al., 2016; Lobert et al., 1999). However, as shown in Figure 9, we also have found relatively high fraction of Cl^- in traffic source profiles in some previous studies. Except for the predominant species OC and EC, the fraction of Cl^- is relatively high compared to other species, which might be attributed to the emission of rubber accessory (neoprene) of vehicles and the use of chlorine in the catalytic reforming of gasoline (Chen et al., 2015; Chow et al., 2004; Cui et al., 2016; Watson et al., 2001; Schauer et al., 1999; Veksha et al., 2018). Similar with Cl^- in traffic source, Ni in biomass burning source has not been regarded as a specific tracer. However, during the process of biomass gasfication, Ni is commonly used as a catalyst (Corella et al., 1999; Sutton et al., 2001).

In conclusion, our PMF results are based on the commonly used tracers and are supported by previous source profiles. It is not uncommon to find that the factor resolved by PMF could not be exactly the same as the source profile from emission source testing. In the future, with more samples and more tracers such as organic markers, it will help to achieve a quite distinct factor and very close to specific emission source profile.

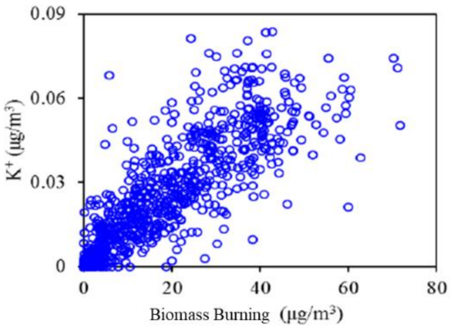

Figure 7. Time series and correlation of K^+ with the biomass burning source identified by PMF

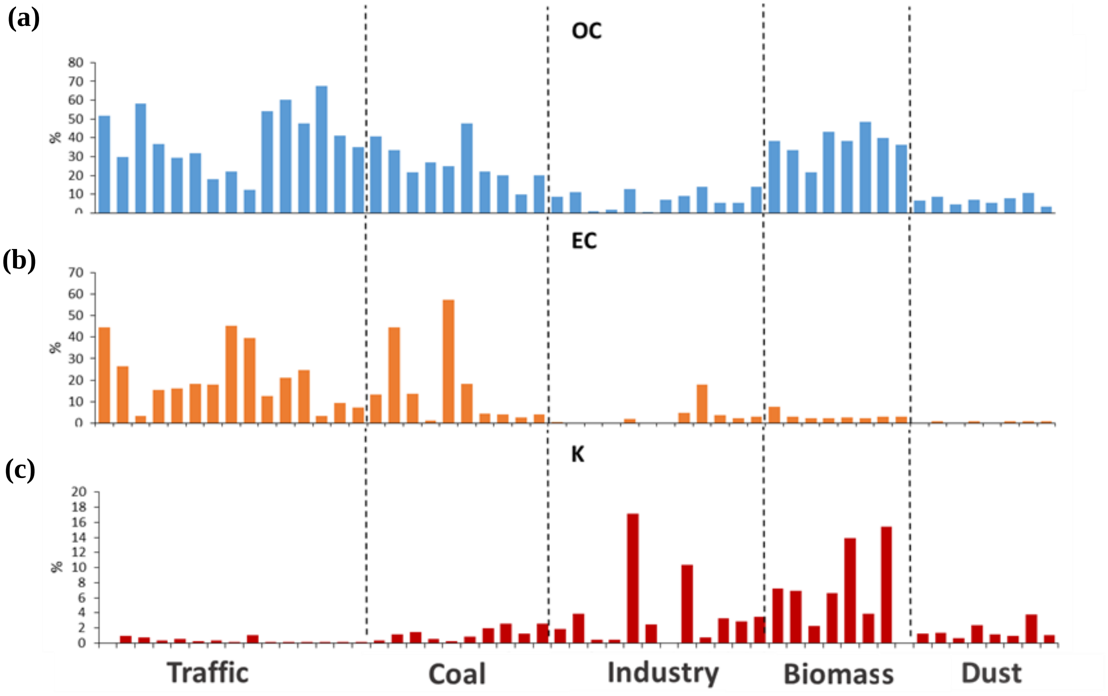

Figure 8. Fraction of (a) OC, (b) EC and (c) K in different source profiles (Dai et al., 2015; Cao et al., 2015; Ma et al., 2015; Wang et al., 2009; Wang et al., 2016; Wen et al., 2009; Wu et al., 2016; Zhang et al., 2012; Zheng et al., 2013)

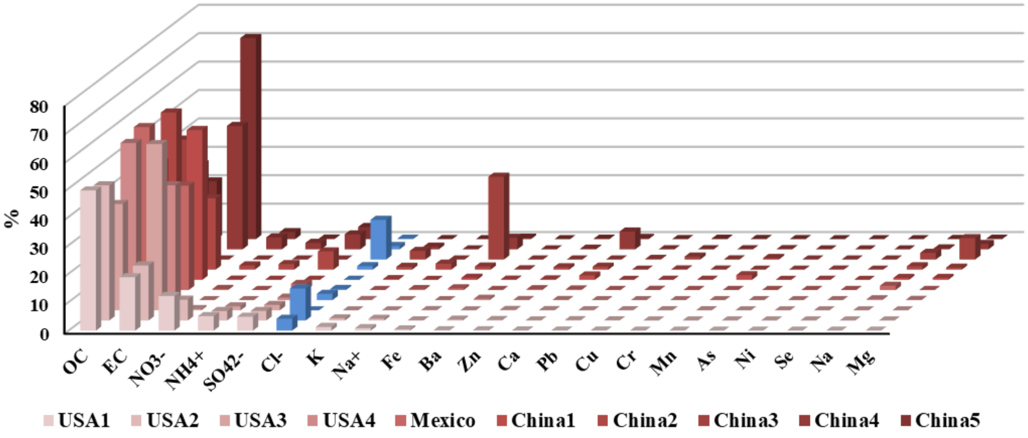

Figure 9. Source profiles of traffic sources in different country (USA1: Watson et al., 2001; USA2:

California, Schauer et al., 1999; USA3: Colorado, Watson et al., 2001; USA4: Texas, Chow et al., 2004; Mexico: Watson et al., 2001; China1: Nanjing, Chen et al., 2015; China2: Pearl River Delta, Feng et al., 2013; China3: Shandong-gasoline, Cui et al., 2016; China4: Shandong-diesel, Cui et al., 2016; China5: Shandong-truck, Cui et al., 2016)

Reference

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[2] Chen, P., Wang, T., Hu, X., Xie, M., Zhuang, B., and Li, S.: A study of chemical mass balance source apportionment of fine particulate matter in Nanjing, Journal of Nanjing University (Natural Science), 51, 524-534, 2015.

[3] Chow, J. C., Watson, J. G., Kuhns, H., Etyemezian, V., Lowenthal, D. H., Crow, D., Kohl, S. D., Engelbrecht, J. P., and Green, M. C.: Source profiles for industrial, mobile, and area sources in the Big Bend Regional Aerosol Visibility and Observational study, Chemosphere, 54, 185-208, 2004.

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[5] Cui, M., Chen, Y., Tian, C., Zhang, F., Yan, C., and Zheng, M.: Chemical composition of PM$_{2.5}$ from two tunnels with different vehicular fleet characteristics, Sci. Total. Environ., 550, 123-132, 2016.

[6] Dai, S. H.: Real-time single-particle volatility of urban aerosol and characteristics of particles emitted from vehicles in the PRD region, Doctoral dissertation, Chinese Academy of Sciences, 2015 (in Chinese).

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[8] Gao, J., Peng, X., Chen, G., Xu, J., Shi, G.L., Zhang, Y.C., and Feng, Y.C.: Insights into the chemical characterization and sources of PM$_{2.5}$ in Beijing at a 1-h time resolution, Sci. Total. Environ., 542, 162-171, 2016.

[9] Lobert, J. M., Keene, W. C., Logan, J. A., and Yevich, R.: Global chlorine emissions from biomass burning: Reactive Chlorine Emissions Inventory, J. Geophys. Res-Atmos., 104, 8373-8389, 1999.

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[22] Zheng, M., Zhao, X., Cheng, Y., Yan, C., Shi, W., Zhang, X., Weber, R. J., Schauer, J. J., Wang, X., and Edgerton, E. S.: Sources of primary and secondary organic aerosol and their diurnal variations, J. Hazard. Mater., 264, 536-544, 2014.

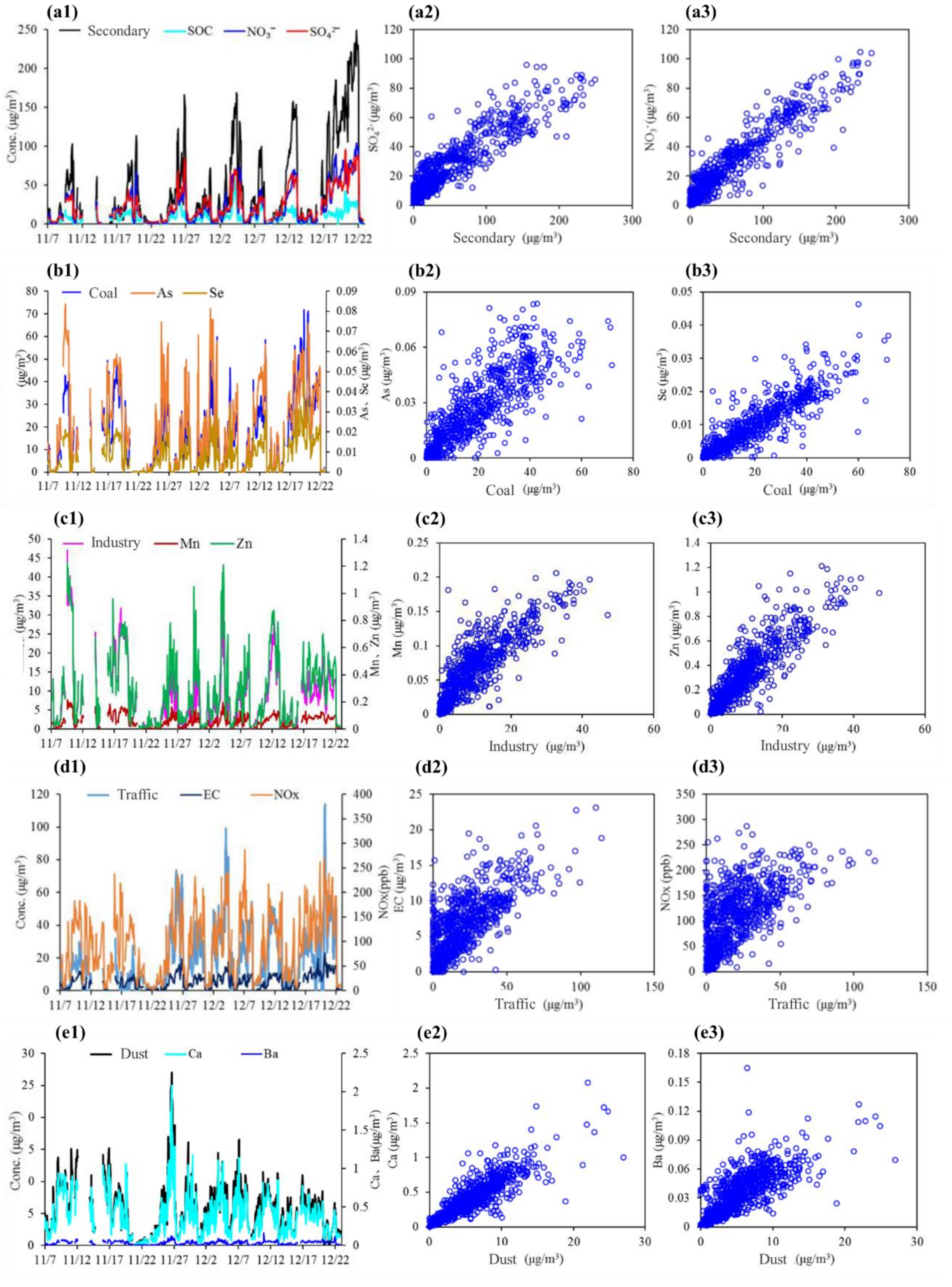
[23] Zheng, M., Zhang, Y. J., Yan, C. Q., Fu, H. Y., Niu, H. Y., Huang, K., Hu, M., Zeng, L. M., Liu, Q. Z., Pei, B., Fu, Q. Y.: Establishing $PM_{2.5}$ industrial source profiles in Shanghai, China Environ. Sci., 33(8), 1354~1359, 2013 (in Chinese).

[24] Zíková, N., Wang, Y., Yang, F., Li, X., Mi, T., and Hopke, P. K.: On the source contribution to Beijing $PM_{2.5}$ concentrations, Atmos. Environ., 134, 84-95, 2016.

Reviewer #1 Comment NO.6: *What are the relationships between the tracers of identified sources and sources mass concentrations?*

Response to reviewer comment NO.6: According to the reviewer's suggestions, we have added the relationships between the tracers of identified sources and sources mass concentrations in the supplementary materials as **Figure S8**. SO_4^{2-} and NO_3^- were typical of the secondary source profiles (Gao et al., 2016; Peng et al., 2016). Mn and Zn were used as indicators for industrial source (Hu et al., 2015; Li et al., 2017). Ca and Ba were mainly emitted from dust source (Amato et al., 2013; Shen et al., 2016). The correlation of EC and NOx was analyzed with traffic source and As and Se were typical tracers for coal combustion (Vejahati et al., 2010).

Figure 10(a1), (b1), (c1), (d1) and (e1) shows the time series of source concentration and source tracers of secondary source, coal combustion source, industrial source, traffic source and dust source. Figure 10(a2), (a3) shows the scatterplot of secondary source with SO_4^{2-} and NO_3^-. Figure 10(b2), (b3) shows the scatterplot of coal combustion source with As and Se. Figure 10(c2), (c3) shows the scatterplot of industrial source with Mn and Zn. Figure 10(d2), (d3) shows the scatterplot of traffic source with EC and NOx. Figure 10(e2), (e3) shows the scatterplot of dust source with Ca and Ba. It could be seen that secondary source concentration showed high correlation with SO_4^{2-} and NO_3^- concentration, with both R^2 higher than 0.8. Coal combustion source concentration correlated well with As and Se concentration, with the R^2 of 0.86 and 0.98, respectively. Industrial source concentration correlated well with Mn and Zn, with R^2 over 0.85. As mentioned before, the correlation between biomass burning source and K^+ was good with R^2 over 0.6. For traffic source, we investigated its correlation with EC and NOx, and found relatively high correlation with R^2 of 0.70 and 0.51, respectively. Dust source concentration correlated well with the concentration of Ca and Ba, with R^2 of 0.96 and 0.72, respectively. In summary, the tracers of identified sources and sources mass concentrations show good correlation in general, which help to verify the PMF results. We appreciate this helpful suggestion by the reviewer.



(a1)
(a2)
(a3)
Secondary
SOC
NO_3^-
SO_4^{2-}
Conc. (μg/m³)
SO_4^{2-} (μg/m³)
NO_3^- (μg/m³)
Secondary (μg/m³)
(b1)
(b2)
(b3)
Coal
As
Se
(μg/m³)
As、Se (μg/m³)
As (μg/m³)
Se (μg/m³)
Coal (μg/m³)
(c1)
(c2)
(c3)
Industry
Mn
Zn
(μg/m³)
Mn、Zn (μg/m³)
Mn (μg/m³)
Zn (μg/m³)
Industry (μg/m³)
(d1)
(d2)
(d3)
Traffic
EC
NOx
Conc. (μg/m³)
NOx(ppb)
EC (μg/m³)
NOx (ppb)
Traffic (μg/m³)
(e1)
(e2)
(e3)
Dust
Ca
Ba
Conc. (μg/m³)
Ca、Ba(μg/m³)
Ca (μg/m³)
Ba (μg/m³)
Dust (μg/m³)
11/7
11/12
11/17
11/22
11/27
12/2
12/7
12/12
12/17
12/22

Figure 10. Relationships between the tracers of identified sources and sources mass concentrations (secondary for secondary source; coal for coal combustion source; industry for industrial source; traffic for traffic source; dust for dust source). (a1)-(e1) shows the time series of source concentration with source tracers. (a2)-(e2) and (a3)-(e3) shows the scatterplot of source concentration with source tracers.

Reference

[1] Amato, F., Schaap, M., van der Gon, H. A. D., Pandolfi, M., Alastuey, A., Keuken, M., and Querol, X.: Short-term variability of mineral dust, metals and carbon emission from road dust resuspension, Atmos. Environ., 74, 134-140, 2013.

[2] Gao, J., Peng, X., Chen, G., Xu, J., Shi, G.L., Zhang, Y.C., and Feng, Y.C.: Insights into the chemical characterization and sources of $PM_{2.5}$ in Beijing at a 1-h time resolution, Sci. Total Environ., 542, 162-171, 2016.

[3] Hu, Y., Lin, J., Zhang, S., Kong, L., Fu, H., and Chen, J.: Identification of the typical metal particles among haze, fog, and clear episodes in the Beijing atmosphere, Sci. Total Environ., 511, 369-380, 2015.

[4] Li, M., Zhang, Q., Kurokawa, J., Woo, J.H., He, K., Lu, Z., Ohara, T., Song, Y., Streets, D. G., and Carmichael, G. R.: MIX: a mosaic Asian anthropogenic emission inventory under the international collaboration framework of the MICS-Asia and HTAP, Atmos. Chem. Phys., 17, 2017.

[5] Peng, X., Shi, G.L., Gao, J., Liu, J.Y., HuangFu, Y.Q., Ma, T., Wang, H.T., Zhang, Y.C., Wang, H., and Li, H.: Characteristics and sensitivity analysis of multiple-time-resolved source patterns of $PM_{2.5}$ with real time data using Multilinear Engine 2, Atmos. Environ., 139, 113-121, 2016.

[6] Shen, Z., Sun, J., Cao, J., Zhang, L., Zhang, Q., Lei, Y., Gao, J., Huang, R.J., Liu, S., and Huang, Y.: Chemical profiles of urban fugitive dust $PM_{2.5}$ samples in Northern Chinese cities, Sci. Total Environ., 569, 619-626, 2016.

[7] Vejahati, F., Xu, Z., and Gupta, R.: Trace elements in coal: Associations with coal and minerals and their behavior during coal utilization–A review, Fuel, 89, 904-911, 2010.

Reviewer #1 Comment NO.7: *I don't think the discussions in 3.3 and 3.4 are necessary if the authors can't reply the above comments.*

Response to reviewer comment NO.7: We have tried our best to reply the above comments and the detailed reply to each comment is listed above. Therefore, we would like to keep discussions in 3.3 and 3.4.

Reviewer #1 Comment NO.8: *I suggest the authors to add more information about the spatial mass concentrations of PM2.5, PM10, SO2 and NO2 in Figure 7. Moreover, sources inventories used in this study would be suggested to add in the supplementary materials. The results resolved by the footprint and*

NAQPMS models should be discussed based on above information.

Response to reviewer comment NO.8: Based on the reviewer's suggestion, the spatial mass concentrations of $PM_{2.5}$, wind speed and wind direction during EP1 in Figure 7 are added to the supplementary materials as **Figure S9** for better understanding of the evolution of the haze episode. The source inventory used in NAQPMS is the MIX (http://www.meicmodel.org/dataset-mix.html) anthropogenic emission inventory with the original resolution of 0.25 °(about 25 km at middle latitudes) and the year of 2010. It could be applied and downloaded on the website. The results of NAQPMS model are based on the MIX inventory while the footprint model do not include an inventory as its input.

Reviewer #2 General Comment: *While PMF was the key model to apportion PM2.5 sources, further details about the optimum solution of PMF need to be discussed systematically, such as; the examination of the optimum factor solution, factor analysis, and the uncertainties associated with the estimation of each factor.*

Response to reviewer general comment: Thanks for the reviewer's suggestion. We added the detailed information as to how to find the optimum factor solution and the uncertainty estimation by bootstrapping (BS), displacement (DISP), and bootstrapping with displacement (BS-DISP) in the manuscript and the supplementary materials.

To determine the optimal number of source factors, a string of effective test, in which factors number was from four to nine, was carried out. The resulting Q parameters were shown in Figure 11 (**Figure S4** in the supplementary materials). Obviously, there was a lowest Q_{Robust} value (13087) at six factors in moving from four to nine factors. Although $Q_{expected}$ has been decreasing in the process, $Q/Q_{expected}$ shared similar variation with Q_{Robust} showing the lowest value at six factors (1.3).

Uncertainty of PMF model is usually estimated by bootstrapping (BS), displacement (DISP), and bootstrapping with displacement (BS-DISP). Here, characteristics of factors nearby six, where Q_{Robust} was relative lower, were explored. With five factors, three factors were mapped 100% of BS, while industry source and traffic source were mapped 92% and 94%, respectively of runs. There were no swaps with DISP, and 100% of the BS-DISP runs were successfully. At six factors, results were more stable with all factors mapped in BS in 100% (Table 3 below, also **Table S2** in the supplementary materials), no swaps occurred with DISP and all BS-DISP runs were successfully. However, the solution became less stable in moving from six to seven factors. The new sea salt factor was only mapped in BS in 87% and coal combustion factor was mapped in BS in 89%, traffic source factor was mapped in 93%, other factors were mapped in 100% of runs. No swaps were found in DISP. Therefore, based on the above analysis, six factors were found to be the optimal solution in this study.

Table 3 Percentage of BS factors assigned to each base case factor with a correlation threshold of 0.6.

Boot Factor	Secondary	Industrial	Dust	Traffic	Coal	Biomass
1	100	0	0	0	0	0
2	0	100	0	0	0	0
3	0	0	100	0	0	0
4	0	0	0	100	0	0
5	0	0	0	0	100	0
6	0	0	0	0	0	100

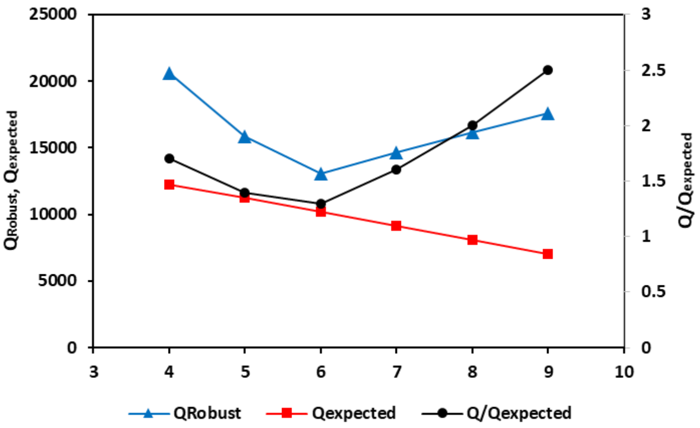


Figure 11. The variation of Q parameters from four factors to ten factors

<u>Reviewer #2 General Comment</u>: *Also, many of your comparisons with the previous study need to include more details, such as size fraction of PM, type of receptor model used and weather organic tracers were used or not, time resolution, which month, year, etc.*

Response to reviewer general comment: According to the reviewer's suggestion, the detailed information including size fractions of previous studies has been added in the supplementary materials as follows:

Table 4. Previous studies about source apportionment of Beijing

	Sampling period and time resolution	Size fraction	Receptor model	Tracers
Gao et al., 2016	July to August, 2014; 1 hour	2.5 μm	PCA; PMF; ME2	Inorganic tracers
Peng et al., 2016	July to August, 2014; 1 hour	2.5 μm	ME2	Inorganic tracers
Zhang et al., 2013	2009-2010; daily	2.5 μm	PMF	Inorganic tracers

Reference

[1] Gao, J., Peng, X., Chen, G., Xu, J., Shi, G.L., Zhang, Y.C., and Feng, Y.C.: Insights into the chemical characterization and sources of $PM_{2.5}$ in Beijing at a 1-h time resolution, Sci. Total Environ., 542, 162-171, 2016.

[2] Peng, X., Shi, G.L., Gao, J., Liu, J.Y., HuangFu, Y.Q., Ma, T., Wang, H.T., Zhang, Y.C., Wang, H., and Li, H.: Characteristics and sensitivity analysis of multiple-time-resolved source patterns of $PM_{2.5}$ with real time data using Multilinear Engine 2, Atmos. Environ., 139, 113-121, 2016.

[3] Zhang, R., Jing, J., Tao, J., Hsu, S.-C., Wang, G., Cao, J., Lee, C. S. L., Zhu, L., Chen, Z., and Zhao, Y.: Chemical characterization and source apportionment of $PM_{2.5}$ in Beijing: seasonal perspective, Atmos. Chem. Phys., 13, 7053-7074, 2013.

Reviewer #2 Comment NO.1: *Page 1, line 25: Subscript PM2.5 to $PM_{2.5}$.*

Response to reviewer comment NO.1: Based on the reviewer's comment, the word has been revised to "$PM_{2.5}$".

Reviewer #2 Comment NO.2: *Page 1, line 26: Define "receptor model" and replace the name with the new sentence in the same line*

Response to reviewer comment NO.2: The sentence has been revised as follows (Page 1, Line 26): *This study developed a combined method to investigate source types of* $PM_{2.5}$ *and its source regions during winter 2016 in Beijing, which include the receptor model (Positive Matrix Factorization, PMF), footprint, and an air quality model. The PMF model was performed with high-time resolution measurements of trace elements, water soluble ions, organic carbon, and elemental carbon using online instruments during the wintertime campaign of the Air Pollution and Human Health-Beijing (APHH-Beijing) program in 2016.*

Reviewer #2 Comment NO.3: *Why you have chosen to report 11% of the dust contribution only? I recommend adding the percent contribution for the major sources or each analyzed footprint, systematically.*

Response to reviewer comment NO.3: Yes, we agree that it is better to add the percent contribution of each major source. The sentence has been revised as follows (see Page 2, line 37):

The combination of PMF with footprint results revealed that the secondary source was mainly associated with southern footprints (53%). The northern footprint was characterized by a high dust source contribution (11%) while industrial source increased with the eastern footprint (10%).

Reviewer #2 Comment NO.4: *Page 2, line 48: Insert a citation after "…life and human health".*

Response to reviewer comment NO.4: A citation has been inserted in this sentence (see Page 2, line 48):

Presently, haze in China has the characteristics of high frequency and long duration on a regional scale, and has influenced public life and human health (Xie et al., 2016).

Reference

[1] Xie, Y., Dai, H., Dong, H., Hanaoka, T., and Masui, T.: Economic impacts from $PM_{2.5}$ pollution-related health effects in China: a provincial-level analysis, Environ. Sci. Technol., 50, 4836-4843, 2016.

Reviewer #2 Comment NO.5: *Page 2, line 51, Coal combustion can also considered as industrial source. Please be more specific here.*

Response to reviewer comment NO.5: Thanks for the helpful suggestion. The coal combustion mentioned in this sentence is the residential coal combustion, which is not included in the industrial source. To be more specific and clear, the sentence has been revised as follows (see Page 2, line 51):

Previous studies have found that $PM_{2.5}$ can be emitted from various sources, including residential coal combustion, biomass burning, traffic-related sources, industrial sources and dust (Watson et al., 2001; Gao et al., 2016).

Reviewer #2 Comment NO.6: *Page 2, line 51: It sounds like there are many sources related to traffic. I suggest replace "traffic sources" with "traffic-related sources", after visiting the two papers you have cited here.*

Response to reviewer comment NO.6: The sentence has been revised as follows (see Page 2, line 51):

Previous studies have found that $PM_{2.5}$ can be emitted from various sources, including residential coal combustion, biomass burning, traffic-related sources, industrial sources and dust (Watson et al., 2001; Gao et al., 2016).

Reviewer #2 Comment NO.7: *Page 3, line 62: "The model can: : :". Which model you are referring to? Please specify.*

Response to reviewer comment NO.7: Sorry for the ambiguity. The sentence has been revised as follows (see Page 3, line 63):

The receptor model can identify and quantify the contribution of multiple source types based on in-situ measurements and specific source tracers.

Reviewer #2 Comment NO.8: *Page 3, line 77: Insert citation after ": : :in previous studies".*

Response to reviewer comment NO.8: Three previous papers have been cited after "… in previous studies" as follows (see Page 3, line 72):

By conducting receptor models based on high-time resolution online measurement, the source types and source contributions in Beijing have been analyzed in previous studies (Gao et al., 2016; Peng et al., 2016; Song et al., 2006).

Reference

[1] Gao, J., Peng, X., Chen, G., Xu, J., Shi, G.L., Zhang, Y.C., and Feng, Y.C.: Insights into the chemical characterization and sources of $PM_{2.5}$ in Beijing at a 1-h time resolution, Sci. Total Environ., 542, 162-171, 2016.

[2] Peng, X., Shi, G.L., Gao, J., Liu, J.Y., HuangFu, Y.Q., Ma, T., Wang, H.T., Zhang, Y.C., Wang, H., and Li, H.: Characteristics and sensitivity analysis of multiple-time-resolved source patterns of $PM_{2.5}$ with real time data using Multilinear Engine 2, Atmos. Environ., 139, 113-121, 2016.

[3] Song, Y., Zhang, Y., Xie, S., Zeng, L., Zheng, M., Salmon, L. G., Shao, M., and Slanina, S.: Source apportionment of $PM_{2.5}$ in Beijing by Positive Matrix Factorization, Atmos. Environ., 40, 1526-1537, 2006.

Reviewer #2 Comment NO.9: *Page 5, line 106: "The room". Which room you are referring to?*

Response to reviewer comment NO.9: "The room" refers to the sampling site and it has been revised as follows (see Page 5, line 108):

The sampling site is located on the sixth floor of a teaching building within PKU.

Reviewer #2 Comment NO.10: *Page 6, line 130: Here you report that XRF was used to quantify metals. I see that you need to add an excel sheet or a table to the supplement that shows: measured concentrations, uncertainties of the measurements, and the detection limit. Also, it should include PM2.5, EC, OC, SIA. These data are important for the science community to replicate the PMF result. Also, in many places later you report the averages of a certain species without the standard deviation and/or the range of that average, which can also be extracted from the suggested table.*

Response to reviewer comment NO.10: We agree with the reviewer about providing detailed information. Table 4 has been added in the supplementary materials as **Table S1** which shows the average concentration, standard deviation, detection limit and BLD% (the percentage of data below the detection limit) of each species, including OC, EC, SIA and metals. The table was presented as follows:

Table 4 Statistical summary of identified species of PM$_{2.5}$ in the entire sampling period

	Mean	Std.	Max	Min	Detection limit	BDL%
OC/EC			**µg/m^3**			**%**
OC	20.8	17.0	89.9	1.1	0.4	-
EC	5.6	4.4	23.1	0.2	0.1	4.3
SIA			**µg/m^3**			**%**
SO_4^{2-}	23.5	20.8	95.8	0.04	0.04	0.21
NO_3^-	22.0	23.3	104.7	0.03	0.03	0.1
NH_4^+	14.0	14.7	66.6	0.04	0.05	1.4
Na^+	0.39	0.32	1.89	0.02	0.04	8.5
Cl^-	4.89	4.19	27.6	0.05	0.05	0.1
Metal		**µg/m^3**			**ng/m^3**	**%**
K	1.49	1.17	5.28	0.10	2.366	-
Fe	0.769	0.541	2.22	0.015	0.759	-
Ca	0.384	0.277	2.08	0.001	0.902	-
Zn	0.286	0.261	1.85	0.005	0.231	-
Pb	0.107	0.091	0.469	0.004	0.218	-
Mn	0.058	0.046	0.210	0.001	0.283	-
Ba	0.035	0.023	0.160	0.002	0.945	-
Cu	0.027	0.024	0.171	0.002	0.267	-
As	0.022	0.021	0.084	0.000	0.114	17
Cr	0.010	0.010	0.110	0.000	0.288	11
Se	0.008	0.008	0.046	0.000	0.141	5.2
Ni	0.002	0.002	0.044	0.000	0.226	3.4

* BDL% refers to the percentage of data below the detection limit

* The unit of the detection limit of each metal is ng/m^3

* '-' means that all data are above the detection limit

Reviewer #2 Comment NO.11: *Page 8, line 181: Add a comma after "5 km".*

Response to reviewer comment NO.11: A comma has been added after "5 km" (see Page 8, line 199).

Reviewer #2 Comment NO.12: *Page 8, line 181: Add a space after "x", and before "2.5"..*

Response to reviewer comment NO.12: A space has been added after "x" and before "2.5" (see Page 9, line 202).

Reviewer #2 Comment NO.13: *Page 10, line 229-232: you have calculated the concentration of mineral species (Al, Si, Fe) based on Ca concentration, and the composition of urban soil. Dose this typical urban soil was affected by regional and local pollution? During summer or winter? During hazy or non-hazy effect? And what is the estimated uncertainty in this calculation (estimation).*

Response to reviewer comment NO.13: The composition of urban soil is investigated by An et al (2016). In this study, 2692 topsoil samples were collected in the urban area of Beijing during 2011. The composition of soil was an annual average result and most of the urban areas of Beijing including our sampling site (PKU) was covered. Based on large amount of samples, the soil composition in this study could be considered as typical urban soil composition of Beijing. Thus we choose it to calculate mineral species in our study. An et al., (2016) identified that local soil in Beijing were influenced by both local and regional sources including industrial, traffic, agricultural and biomass burning source.

Due to the spatial and temporal variability in soil dust sources, it is very difficult to characterize an appropriate aerosol soil composition for a specific site and the uncertainty might be large. The concentrations of Al, Si, Fe and Mg were calculated by the concentration of Ca and the composition of urban soils of Beijing: Al= 1.7Ca, Si= 7.3Ca, Fe=0.7Ca, Mg= 0.3Ca (An et al., 2016). Therefore, the uncertainty in this calculation could be roughly estimated as follows:

$$\left(\frac{U_{Al}}{C_{Al}}\right)^2 = \left(\frac{U_{\mathrm{Ca}}}{C_{\mathrm{Ca}}}\right)^2 + \left(\frac{U_{\mathrm{Al/Ca}}}{R_{\mathrm{Al/Ca}}}\right)^2 \tag{1}$$

$$\left(\frac{U_{Si}}{C_{Si}}\right)^2 = \left(\frac{U_{\mathrm{Ca}}}{C_{\mathrm{Ca}}}\right)^2 + \left(\frac{U_{\mathrm{Si/Ca}}}{R_{\mathrm{Si/Ca}}}\right)^2 \tag{2}$$

$$\left(\frac{U_{Fe}}{C_{Fe}}\right)^2 = \left(\frac{U_{\mathrm{Ca}}}{C_{\mathrm{Ca}}}\right)^2 + \left(\frac{U_{\mathrm{Fe/Ca}}}{R_{\mathrm{Fe/Ca}}}\right)^2 \tag{3}$$

$$\left(\frac{U_{Mg}}{C_{Mg}}\right)^2 = \left(\frac{U_{\mathrm{Ca}}}{C_{\mathrm{Ca}}}\right)^2 + \left(\frac{U_{\mathrm{Mg/Ca}}}{R_{\mathrm{Mg/Ca}}}\right)^2 \tag{4}$$

$$(U_{\mathrm{mineral}})^2 = (U_{\mathrm{Ca}})^2 + (U_{\mathrm{Al}})^2 + (U_{\mathrm{Si}})^2 + (U_{\mathrm{Fe}})^2 + (U_{\mathrm{Mg}})^2 \tag{5}$$

where U refers to uncertainty, C refers to concentration and R refers to ratio. U_{Ca} is the MDL of Ca measured by Xact (0.9 ng/m^3) and C_{Ca} is the average concentration of Ca. U_{Al} / U_{Ca} and R_{Al} / R_{Ca} (same with Si, Fe and Mg) are all calculated from An et al., 2016. Based on the above equations, U_{mineral} is 2.46 µg/m^3 and the average mineral concentration during the sampling period is 3.28±2.46 µg/m^3.

Reference

[1] An, Y. L., Huang, Y., Liu, Q. J., Sun, C., Deng, K. W., Li, D., and Huang, D.: The distribution of surface soil elements and the pollution assessment of heavy metal elements in Beijing. Geological Bulletin of China, 35(12):2111-2120, 2016.

Reviewer #2 Comment NO.14: *Page 10, line 230-232: Belong to the method section. Please move them.*

Response to reviewer comment NO.14: The sentence has been moved to Page 6, line 150 as follows:

Chemical closure has been done between the measured and reconstructed $PM_{2.5}$*. Organic matter (OM) was calculated as OM= 1.6 ×OC (Turpin and Lim, 2001). Mineral species was calculated as Mineral= 1.89 Al +2.14 Si + 1.4 Ca + 1.43 Fe + 1.66 Mg (Zhang et al., 2003). The concentrations of Al, Si, Fe and Mg were calculated by the concentration of Ca and the composition of urban soils of Beijing: Al= 1.7Ca, Si= 7.3Ca, Fe=0.7Ca, Mg= 0.3Ca (An et al., 2016). Since the concentration of Al and Si were not directly measured by Xact, the calculated mineral component might be underestimated. "Others" were calculated by subtracting OM, EC, Mineral and secondary inorganic aerosol (SIA, including* SO_4^{2-}*,*

NO_3^-, NH_4^+) concentration from total $PM_{2.5}$ concentration. The correlation of measured and reconstructed $PM_{2.5}$ mass could be seen in Fig. S6 with R^2=0.892.

Reviewer #2 Comment NO.15: *Page 10, line 234: You have stated that Al and Si might be underestimated. Why? And by how much? Please provide supporting details.*

Response to reviewer comment NO.15: As Al and Si are not be directly measured by Xact, the mineral species (including Al, Si, Ca, Fe, Mg) might be underestimated with only Ca and Fe measured. However, we have already calculated the concentrations of Al, Si, Fe and Mg based on the concentration of Ca and the composition of urban soils of Beijing to compensate for the underestimation. Therefore, the sentence "*Since the concentration of Al and Si were not directly measured by Xact, the calculated mineral component might be underestimated*" is no longer correct here and has been deleted in the revised manuscript.

Reviewer #2 Comment NO.16: *Page 10, line 241: Here you compare the average OC/EC ratio with Yan et al., 2015. Can you be more specific about the time resolution, duration, months, and/or any special pollution events.*

Response to reviewer comment NO.16: Based on the reviewer's suggestion, we add some detailed information about the study of Yan et al., 2015. The time resolution of this study is 1 day (23.5 hours). The sampling period in winter is from January 11th to 18th, 2013. There was a haze episode from January 11th to 14th, with the highest $PM_{2.5}$ concentration of 500 μg/m^3. The $PM_{2.5}$ concentration decreased during January 15th to 18th, ranging from 50 ~ 200 μg/m^3. Generally, the air pollution during the whole sampling period in winter was severe, with the average $PM_{2.5}$ concentration of 209 ± 145 μg/m^3. Therefore, the OC/EC ratio was much higher than that in our study.

Reference

[1] Yan, C., Zheng, M., Sullivan, A. P., Bosch, C., Desyaterik, Y., Andersson, A., Li, X., Guo, X., Zhou, T., and Gustafsson, Ö.: Chemical characteristics and light-absorbing property of water-soluble organic carbon in Beijing: Biomass burning contributions, Atmos. Environ., 121, 4-12, 2015.

Reviewer #2 Comment NO.17: *Page 10, line 242-243: Is the concentrations of SO42- (23±20) ug/m3 significantly higher than that for NO3- (22±23)ug/m3? For me they look the same, taking the high variation of the concentrations, as they are reported. Please check these comparisons here and in other places along the manuscript. Also, discuss what is the potential reason for this observation based on previous PM2.5 studies conducted in Beijing during winter.*

Response to reviewer comment NO.17: We agree with the reviewer that the concentration level of SO_4^{2-} and NO_3^- is not significantly different. The sentence has been revised as follows (see Page 11, line 255):

SO_4^{2-} is the predominant ion in SIA with an average concentration of 23.5 ± 20.8 μg m^{-3}, which was similar with that of NO_3^- (22.0 ± 23.3 μg m^{-3}).

In previous studies, SIA has also been found to be predominant composition of $PM_{2.5}$ in winter in Beijing, which could be attributed to large emission of gas precursors (SO_2 and NOx), rapid aqueous-phase production of SO_4^{2-} and regional transport (Sun et al., 2014; Sun et al., 2016; Yang et al., 2015).

Reference

[1] Sun, Y., Chen, C., Zhang, Y., Xu, W., Zhou, L., Cheng, X., Zheng, H., Ji, D., Li, J., and Tang, X.: Rapid formation and evolution of an extreme haze episode in Northern China during winter 2015, Sci. Rep., 6, 27151, 2016.

[2] Sun, Y., Qi, J., Wang, Z., Fu, P., Jie, L., Yang, T., and Yan, Y.: Investigation of the sources and evolution processes of severe haze pollution in Beijing in January 2013, J. Geophys. Res-Atmos., 119, 4380-4398, 2014.

[3] Yang, Y., Liu, X., Yu, Q., Wang, J., An, J., Zhang, Y., and Fang, Z.: Formation mechanism of continuous extreme haze episodes in the megacity Beijing, China, in January 2013, Atmos. Res., 155, 192-203, 2015.

Reviewer #2 Comment NO.18: *Page 10, line 246: Insert a citation(s) after "…in winter in Beijing"*

Response to reviewer comment NO.18: A citation has been added after "…in winter in Beijing" as follows (see Page 11, line 258):

In general, the large contribution of SIA, OM as well as the high OC/EC ratio indicated the importance *of secondary formation in winter in Beijing (Sun et al., 2016b), while the high concentration of species like SO_4^{2-} and K suggested a significant contribution of combustion sources including coal combustion and biomass burning to $PM_{2.5}$ (Achad et al., 2018; Chen et al., 2017; Li et al., 2017).*

Reference

[2] Sun, Y., Chen, C., Zhang, Y., Xu, W., Zhou, L., Cheng, X., Zheng, H., Ji, D., Li, J., and Tang, X.: Rapid formation and evolution of an extreme haze episode in Northern China during winter 2015, Sci. Rep., 6, 27151, 2016b.

Reviewer #2 Comment NO.19: *Page 10, line 246-247: What type of combustion source are usually attributed to K and SO4, please be more specific and include a citation.*

Response to reviewer comment NO.19: According to previous studies, coal combustion and biomass burning source are usually attributed to K and SO_4^{2-}, and the citations has been added as follows (see Page 11, line 260):

In general, the large contribution of SIA, OM as well as the high OC/EC ratio indicated the importance of secondary formation in winter in Beijing (Sun et al., 2016), while the high concentration of species like SO_4^{2-} and K suggested a significant contribution of combustion sources including coal combustion and biomass burning to $PM_{2.5}$ (Achad et al., 2018; Chen et al., 2017; Li et al., 2017).

Reference

[1] Achad, M., Caumo, S., de Castro Vasconcellos, P., Bajano, H., Gómez, D., and Smichowski, P.: Chemical markers of biomass burning: Determination of levoglucosan, and potassium in size-classified atmospheric aerosols collected in Buenos Aires, Argentina by different analytical techniques, Microchem. J., 139, 181-187, 2018.

[2] Chen, S., Guo, Z., Guo, Z., Guo, Q., Zhang, Y., Zhu, B., and Zhang, H.: Sulfur isotopic fractionation and its implication: Sulfate formation in $PM_{2.5}$ and coal combustion under different conditions, Atmos. Res., 194, 142-149, 2017.

[3] Li, H., Zhang, Q., Zhang, Q., Chen, C., Wang, L., Wei, Z., Zhou, S., Parworth, C., Zheng, B., and Canonaco, F.: Wintertime aerosol chemistry and haze evolution in an extremely polluted city of the North China Plain: significant contribution from coal and biomass combustion, Atmos. Chem. Phys., 17, 4751-4768, 2017.

Reviewer #2 Comment NO.20: *Page 10, line 249-250: “ : : :the contribution of sulfate increased significantly (up to 24%), compared to what? Please be more specific here. I think you want to say that* *sulfate is the major component when PM2.5< 50ug/m3.*

Response to reviewer comment NO.20: The sentence has been revised as follows (see Page 11, line 265):

Under low $PM_{2.5}$ *concentration (< 50 μg* m^{-3}*),* SO_4^{2-} *was one of the major components of* $PM_{2.5}$ *with the contribution of around 24%.*

Reviewer #2 Comment NO.21: *Page 10, line 249-250: You are using SO42- and sulfate back and forth. Choose one term and stick with it.*

Response to reviewer comment NO.21: We agree that it is very important to be consistent. We choose to use SO_4^{2-} throughout the manuscript and have replaced the term “sulfate” with “SO_4^{2-}”.

Reviewer #2 Comment NO.22: *Page 11, line 254-255: Insert citations after “: : :tracers of biomass burning and coal combustion”, and after “tracers of dust sources”.*

Response to reviewer comment NO.22: According to the reviewer’s suggestion, several citations have been added in this sentence as follows (see Page 11, line 270):

The proportion of K, Pb, As and Se, which were tracers of biomass burning and coal combustion (Achad et al., 2018; Chen et al., 2017; Vejahati et al., 2010), increased with $PM_{2.5}$ *concentration. While the contribution of Ca, Ba, Fe, tracers of dust source (Amato et al., 2013; Shen et al., 2016), decreased with* $PM_{2.5}$ *concentration.*

Reference

[1] Achad, M., Caumo, S., de Castro Vasconcellos, P., Bajano, H., Gómez, D., and Smichowski, P.: Chemical markers of biomass burning: Determination of levoglucosan, and potassium in size-classified atmospheric aerosols collected in Buenos Aires, Argentina by different analytical techniques, Microchem. J., 139, 181-187, 2018.

[2] Amato, F., Schaap, M., van der Gon, H. A. D., Pandolfi, M., Alastuey, A., Keuken, M., and Querol, X.: Short-term variability of mineral dust, metals and carbon emission from road dust resuspension, Atmos. Environ., 74, 134-140, 2013.

[3] Chen, S., Guo, Z., Guo, Z., Guo, Q., Zhang, Y., Zhu, B., and Zhang, H.: Sulfur isotopic fractionation and its implication: Sulfate formation in $PM_{2.5}$ and coal combustion under different conditions, Atmos. Res., 194, 142-149, 2017.

[4] Shen, Z., Sun, J., Cao, J., Zhang, L., Zhang, Q., Lei, Y., Gao, J., Huang, R.J., Liu, S., and Huang, Y.: Chemical profiles of urban fugitive dust $PM_{2.5}$ samples in Northern Chinese cities, Sci. Total Environ., 569, 619-626, 2016.

[5] Vejahati, F., Xu, Z., and Gupta, R.: Trace elements in coal: Associations with coal and minerals and their behavior during coal utilization-A review, Fuel, 89, 904-911, 2010.

Reviewer #2 Comment NO.23: *Page 11, line 270: Add a "comma" after Ba.*

Response to reviewer comment NO.23: We have added a "comma" after Ba.

Reviewer #2 Comment NO.24: *Page 12, line 277-278: Add an estimation for the of cooking sources to PM2.5 in Beijing during winter, based on studies utilized organic tracers during winter. And discuss weather it is a minor or major source.*

Response to reviewer comment NO.24: In China, cooking source is believed to be an important source to organic carbon of $PM_{2.5}$ (Sun et al., 2013; Zheng et al., 2005). In the study of Sun et al. (2013) the contribution of cooking source to organic aerosol during winter in Beijing was investigated with an Aerodyne Aerosol Chemical Speciation Monitor (ACSM). It was found that cooking source was a major source to OA, contributing to 19% on average. The contribution of cooking source to ambient $PM_{2.5}$ is also important. Chafe et al. (2014) investigated the contribution of household cooking with solid fuels to ambient $PM_{2.5}$, and found the average contribution of 10% in East Asia. Based on previous studies, cooking source might be an important source to $PM_{2.5}$ in China, but not a predominant source. The estimation by Chafe et al. (2014) has been added in the manuscript as follows (see Page 12, line 293):

In previous studies, cooking source could be one of the important sources of $PM_{2.5}$, contributing to about 10% on average in East Asia (Chafe et al., 2014; Sun et al., 2013), but in this study cooking source was not identified by PMF due to the lack of organic tracers.

Reference

[1] Chafe, Z. A., Michael, B., Zbigniew, K., Rita, V. D., Sumi, M., Shilpa, R., Keywan, R., Frank, D., and Smith, K. R.: Household cooking with solid fuels contributes to ambient $PM_{2.5}$ air pollution and the burden of disease, Environ. Health Perspect., 122, 1314-1320, 2014.

[2] Sun, Y., Wang, Z., Pingqing, F. U., Jiang, Q. I., Yang, T., Jie, L. I., and Xinlei, G. E.: The impact of relative humidity on

aerosol composition and evolution processes during wintertime in Beijing, China, Atmos. Environ., 77, 927-934, 2013.

[3] Zheng, M., Salmon, L. G., Schauer, J. J., Zeng, L., Kiang, C., Zhang, Y., and Cass, G. R.: Seasonal trends in $PM_{2.5}$ source contributions in Beijing, China, Atmos. Environ., 39, 3967-3976, 2005.

Reviewer #2 Comment NO.25: *Page 12, line 287: Add a SD for the average value of 75 ug/m3. Or you can report the average PM2.5 during the four haze episodes > 98ug/m3 to be consistent with the table.*

Response to reviewer comment NO.25: We took the suggestion by the reviewer. In the revised manuscript, we reported that the average $PM_{2.5}$ during the four haze episodes were all above 97 μg m^{-3} to ensure the consistency with the table. The revised sentence is shown below:

The average $PM_{2.5}$ concentrations in four haze episodes were all above 97 μg m^{-3} (see Table S4).

Reviewer #2 Comment NO.26: *Page 14, line 348-349: Here you almost restated the previous paragraph (line341-345). I think it is not necessary.*

Response to reviewer comment NO.26: Yes, we agree that this sentence is not necessary and has deleted it from the revised manuscript.

Reviewer #2 Comment NO.27: *Page 15, line 271: We can control precursors of secondary sources, but not the secondary sources. Please modify accordingly.*

Response to reviewer comment NO.27: We have modified the sentence as follows (Page 16, line 388):

In the meantime, more control of biomass burning and precursors of secondary source in surrounding areas are also needed to mitigate air pollution in Beijing.

Reviewer #2 Comment NO.28: *Page 20, line 523: Fix (PM2. 5). Extra space.*

Response to reviewer comment NO.28: We have changed PM2.5 to $PM_{2.5}$ in the revised manuscript.

Reviewer #2 Comment NO.29: *Page 20, line 532: Capitalize the first word of the title only.*

Response to reviewer comment NO.29: We have revised the citation as follows:

Reference

[1] Yang, Y., Liu, X., Qu, Y., An, J., Jiang, R., Zhang, Y., Sun, Y., Wu, Z., Zhang, F., and Xu, W.: Characteristics and formation mechanism of continuous hazes in China: a case study in autumn of 2014 in the North China Plain, Atmos. Chem. Phys, 15, 8165-8178, 2015.

Reviewer #2 Comment NO.30: *Page 22, line 569: Sulfate and nitrate (check technical comment #21).*

Response to reviewer comment NO.30: We have replaced the term “sulfate” with “SO_4^{2-}” and the term “nitrate” with “NO_3^-”.

Reviewer #2 Comment NO.31: *Page 24, Figure two: I suggest naming them a and b. Also, please explain the what the white bars represent?.*

Response to reviewer comment NO.31: The white bars represent the frequency of $PM_{2.5}$ concentration. Figure 2 has been revised as follows:

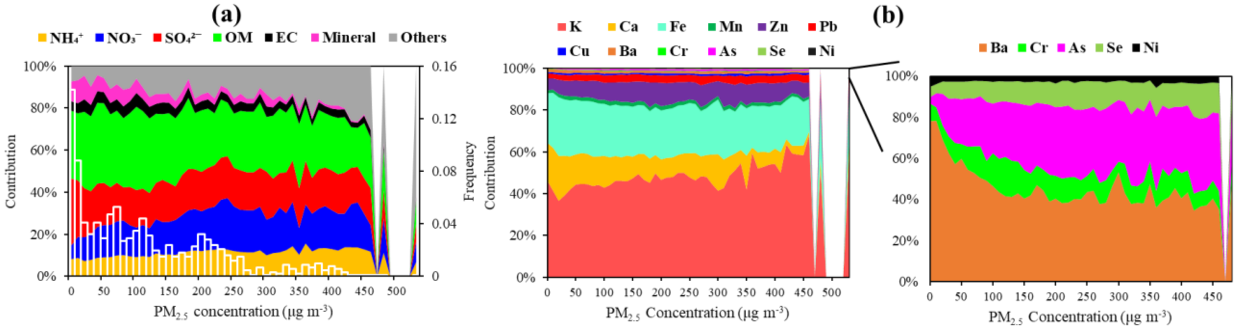


Figure 2. Variation of (a) chemical composition and (b) elemental species with $PM_{2.5}$ concentration (the white bars represent the frequency of $PM_{2.5}$ concentration).

Reviewer #2 Comment NO.32: *Page 26, Figure 4: The right side of the Y-axis shows more than 100%. These are % of what?*

Response to reviewer comment NO.32: The right side of the Y-axis shows the concentration of different sources with the unit of $\mu g/m^3$, while the left side of the Y-axis shows the measured concentration of $PM_{2.5}$ with the unit of $\mu g/m^3$. The unit of Y-axis has been revised in Figure 4, Figure 5, Figure 6 and Figure 7.

Reviewer #2 Comment NO.33: *Page 27, Figure 5: Move the boxes of PMF source identifiers to the left side of the figure and locate them under source apportionment results only. Also, it would be better if you rename these figures as a, and b.*

Response to reviewer comment NO.33: Figure 5 has been revised according to the reviewer's suggestion.

Reviewer #2 Comment NO.34: *Page 28, Figure 6: same comment as for (technical comment #32).*

Response to reviewer comment NO.34: Figure 6 has been revised as mentioned in Reviewer #2 Comment NO.32.

Reviewer #2 Comment NO.35: *Page 29, Figure 7: Check technical comment # 33.*

Response to reviewer comment NO.35: Figure 7 has been revised according to the reviewer's suggestion.

Reviewer #2 Comment NO.36: *Page 30, Figure 30: Add r and p value for the correlations. And discuss in the text.*

Response to reviewer comment NO.36: The r and p value for the correlations have been added in **Figure**

8 as below. The p for correlations between secondary source and local contribution is 0.022 and the p for correlations between coal combustion and local contribution is 0.036. The p for correlations between biomass burning and local contribution exhibited a possible trend toward significance (p=0.052). The r and p values have also been added in the manuscript as follows (see Page 15, Line 376):

The results showed that for PM$_{2.5}$ in Beijing, secondary source contribution decreased when local emission was more significant (p<0.05, r=0.4) while coal combustion, as a primary combustion source, showed an increasing trend along with local contribution estimated by NAQPMS (p<0.05, r=0.3).

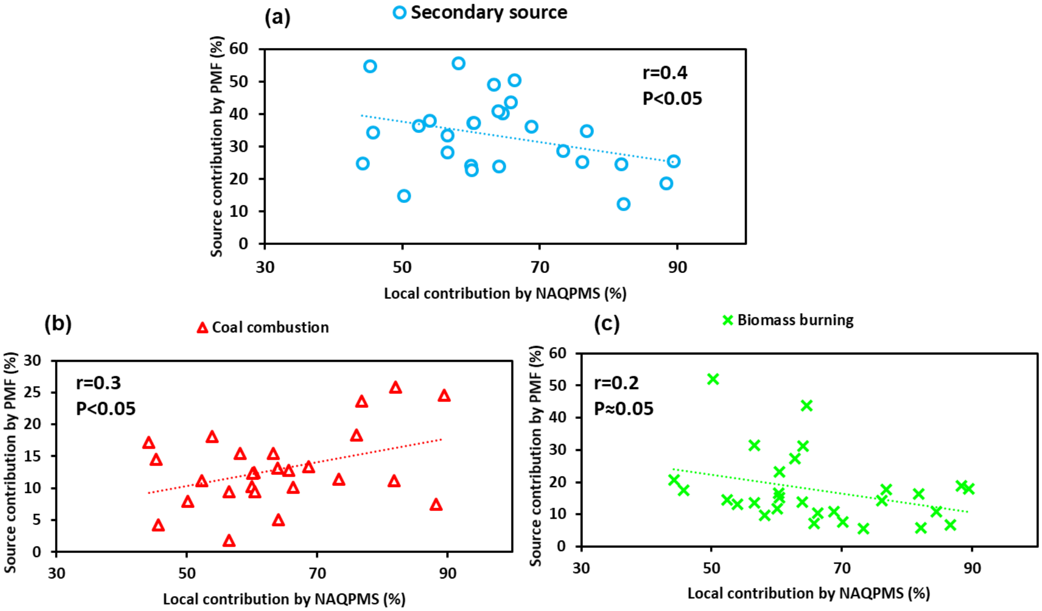


Figure 8. Correlations of local contribution by NAQPMS with the relative contribution by PMF of (a) secondary source, (b) coal combustion source and (c) biomass burning source.

Reviewer #3 Comment NO.1: *Line 52- please add two or three more references for the PM2.5 source apportionment studies. For example you might add the following papers: Kotchenruther, R. a., 2016. Source apportionment of PM2.5 at multiple Northwest U.S.*

sites: Assessing regional winter wood smoke impacts from residential wood combustion.

Atmos. Environ. 142, 210–219.

Taghvaee, S., Sowlat, M.H., Mousavi, A., Hassanvand, M.S., Masud, Y., Naddafi, K., *Sioutas, C., 2018. Source apportionment of ambient PM 2.5 in two locations in central Tehran using the Positive Matrix Factorization (PMF) model. Sci. Total Environ. 629, Zong, Z., Wang, X., Tian, C., Chen, Y., Qu, L., Ji, L., Zhi, G., Li, J., Zhang, G., 2016. Source apportionment of PM2.5 at a regional background site in North China using PMF linked with radiocarbon analysis: Insight into the contribution of biomass burning.Atmos. Chem. Phys. 16, 11249–11265.*

Response to reviewer comment NO.1: Based on the reviewer's comment, the citations have been added in the sentence as follows (Page 1, line 52-53):

Previous studies have found that $PM_{2.5}$ can be emitted from various sources, including residential coal combustion, biomass burning, traffic-related sources, industrial sources and dust (Gao et al., 2016; Kotchenruther et al., 2016; Taghvaee et al., 2018; Watson et al., 2001; *Zong et al., 2016).*

Reference

[1] Kotchenruther, R. A.: Source apportionment of $PM_{2.5}$ at multiple Northwest U.S. sites: Assessing regional winter wood smoke impacts from residential wood combustion, Atmos. Environ., 142, 210-219, 2016.

[2] Taghvaee, S., Sowlat, M. H., Mousavi, A., Hassanvand, M. S., Yunesian, M., Naddafi, K., and Sioutas, C.: Source apportionment of ambient $PM_{2.5}$ in two locations in central Tehran using the Positive Matrix Factorization (PMF) model, Sci. Total Environ., 628-629, 672, 2018.

[3] Zong, Z., Wang, X., Tian, C., Chen, Y., Qu, L., Ji, L., Zhi, G., Li, J., and Zhang, G.: Source apportionment of $PM_{2.5}$ at a regional background site in North China using PMF linked with radiocarbon analysis: Insight into the contribution of biomass burning. Atmos. Chem. Phys. 16, 11249–11265, 2016.

Reviewer #3 Comment NO.2: *Line 106- I suggest you to add references for your claim that "the sampling site is representative of the Beijing urban area" (if applicable).*

Response to reviewer comment NO.2: According to the reviewer's suggestion, we have added several references as follows (Page 5, line 108):

Situated in a mixed district of teaching, residential, and commercial areas, the sampling site is representative of the Beijing urban area (Liu et al., 2018; Yan et al., 2015).

Reference

[1] Liu, Y., Yan, C., and Zheng, M.: Source apportionment of black carbon during winter in Beijing, Sci. Total Environ., 618, 531-541, 2018.

[2] Yan, C., Zheng, M., Sullivan, A. P., Bosch, C., Desyaterik, Y., Andersson, A., Li, X., Guo, X., Zhou, T., and Gustafsson, Ö.: Chemical characteristics and light-absorbing property of water-soluble organic carbon in Beijing: Biomass burning contributions, Atmos. Environ., 121, 4-12, 2015.

Reviewer #3 Comment NO.3: *Line 111- You need to mention more details regarding the usage of Semi-continuous OC/EC Carbon Aerosol Analyzer (Sunset Laboratory Inc.) (e.g., thermal protocols used). Please also add references for the instrument.*

Response to reviewer comment NO.3: More details about the Sunset OC/EC analyzer have been added as follows (Page 5, line 113):

The Sunset OCEC analyzer uses a modified NIOSH 5040 thermal-optical protocol as its default protocol, which produces a relatively reliable determination of OC, EC, and the OCEC split (Bauer et al., 2009). More detailed information could be found in Bauer et al., 2009.

Reference

[1] Bauer, J. J., Xiao-Ying, Y., Robert, C., Nels, L., and Carl, B.: Characterization of the Sunset Semi-continuous Carbon Aerosol Analyzer, Air Repair, 59, 826-833, 2009.

Reviewer #3 Comment NO.4: *Line 142- You definitely need to present the average concentration of PM2.5 chemical components in a table for different episodes of your study. This table should also include the min, max, signal/ noise (S/N) ratio for your data as the important parameters in PMF analysis.*

Response to reviewer comment NO.4: Detailed parameters of $PM_{2.5}$ chemical components for different episodes have been summarized in Table 4 and Table 5 (**Table S1 and Table S4** in the supplementary materials) as follows:

Table 4 Statistical summary of identified species of PM$_{2.5}$ in the entire sampling period

	Mean	Std.	Max	Min	Detection limit	BDL%
OC/EC			**µg/m^3**			**%**
OC	20.8	17.0	89.9	1.1	0.4	-
EC	5.6	4.4	23.1	0.2	0.1	4.3
SIA			**µg/m^3**			**%**
SO_4^{2-}	23.5	20.8	95.8	0.04	0.04	0.21
NO_3^-	22.0	23.3	104.7	0.03	0.03	0.1
NH_4^+	14.0	14.7	66.6	0.04	0.05	1.4
Na^+	0.39	0.32	1.89	0.02	0.04	8.5
Cl^-	4.89	4.19	27.6	0.05	0.05	0.1
Metal		**µg/m^3**			**ng/m^3**	**%**
K	1.49	1.17	5.28	0.10	2.366	-
Fe	0.769	0.541	2.22	0.015	0.759	-
Ca	0.384	0.277	2.08	0.001	0.902	-
Zn	0.286	0.261	1.85	0.005	0.231	-
Pb	0.107	0.091	0.469	0.004	0.218	-
Mn	0.058	0.046	0.210	0.001	0.283	-
Ba	0.035	0.023	0.160	0.002	0.945	-
Cu	0.027	0.024	0.171	0.002	0.267	-
As	0.022	0.021	0.084	0.000	0.114	17
Cr	0.010	0.010	0.110	0.000	0.288	11
Se	0.008	0.008	0.046	0.000	0.141	5.2
Ni	0.002	0.002	0.044	0.000	0.226	3.4

* BDL% refers to the percentage of data below the detection limit

* The unit of the detection limit of each metal is ng/m^3

* ‘-’ means that all data are above the detection limit

Table 5 Average concentration of PM$_{2.5}$ and identified species in different haze and non-haze periods

Unit: µg m^{-3}

Average	**EP1**	**EP2**	**EP3**	**EP4**	**NH1**
Conc.	n=102	n=95	n=117	n=131	n=78
PM$_{2.5}$	97.7±70.7	143.8±119.1	115.3±108.6	241.8±115.5	18.8±20.7
OC	19.1±10.7	24.7±18.7	23.1±21.1	40.3±14.5	3.33±2.85
EC	4.1±2.7	7.1±5.1	5.6±4.3	11.0±3.7	0.9±0.6
SO_4^{2-}	18.6±10.9	25.1±20.4	23.4±19.7	53.3±19.2	4.43±3.89
NO_3^-	19.9±14.6	18.9±16.0	23.3±23.3	56.8±24.8	2.81±3.69
NH_4^+	13.3±8.36	12.6±11.6	13.3±13.2	36.4±16.7	2.42±3.80
Na^+	0.37±0.23	0.35±0.28	0.49±0.41	0.69±0.23	0.10±0.14
Cl^-	5.07±3.05	5.17±3.71	5.66±5.87	8.15±3.41	0.97±1.10
K	2.55±1.33	1.21±0.90	1.23±1.08	2.53±0.78	0.410±0.410
Fe	1.26±0.58	0.724±0.568	0.641±0.480	1.06±0.25	0.144±0.133
Ca	0.530±0.250	0.479±0.447	0.356±0.249	0.360±0.148	0.054±0.033
Zn	0.442±0.242	0.242±0.197	0.252±0.289	0.409±0.170	0.060±0.076
Pb	0.182±0.105	0.087±0.071	0.090±0.084	0.185±0.063	0.024±0.028
Mn	0.093±0.050	0.049±0.038	0.049±0.045	0.082±0.021	0.011±0.012
Ba	0.047±0.016	0.038±0.032	0.032±0.022	0.048±0.012	0.006±0.005
Cu	0.028±0.015	0.023±0.021	0.028±0.026	0.042±0.017	0.007±0.009
As	0.031±0.019	0.019±0.019	0.021±0.023	0.040±0.013	0.003±0.006
Cr	0.019±0.022	0.007±0.009	0.008±0.011	0.014±0.010	0.001±0.001
Se	0.012±0.007	0.006±0.005	0.007±0.007	0.018±0.007	0.001±0.001
Ni	0.003±0.002	0.002±0.001	0.002±0.002	0.004±0.004	0.0006±0.0004

Reviewer #3 Comment NO.5: *Line 149- please add the (Norris et al., 2014; Paatero and Tapper, 1994; Paatero et al., 2014;Paatero, 1997) as the main references for PMF model:*

Paatero, P., 1997. Least Squares Formulation of Robust Non-negative Factor Analysis.pp. 23–35.

Paatero, P., Tapper, U., 1994. Positive matrix factorization: a non-negative factor model with optimal utilization of error estimates of data values. Environmetrics 5, 111–126.

Paatero, P., Eberly, S., Brown, S.G., Norris, G.a., 2014. Methods for estimating uncertainty in factor analytic solutions. Atmos.Meas. Tech. 7:781–797. https://doi.org/10.5194/amt-7-781-2014.

Norris, G., Duvall, R., Brown, S., Bai, S., 2014. EPA Positive Matrix Factorization (PMF) 5.0 Fundamentals and User Guide.

Response to reviewer comment NO.5: The above references have been added to the introduction of PMF model as follows (Page 7, line 163):

Factor contributions and profiles were derived by minimizing the objective function Q in the PMF model, which was determined as follows (Norris et al., 2014; Paatero and Tapper, 1994; Paatero et al., 2014;Paatero, 1997):

Reference

[1] Norris, G., Duvall, R., Brown, S., and Bai, S.: EPA Positive Matrix Factorization (PMF) 5.0 Fundamentals and User Guide, 2014.

[2] Paatero, P.: Least squares formulation of robust non-negative factor analysis, Chemometr. Intell. Lab., 37, 23-35, 1997.

[3] Paatero, P., and Tapper, U.: Positive matrix factorization: A non-negative factor model with optimal utilization of error estimates of data values, Environmetrics, 5, 111-126, 2010.

[4] Paatero, P., Eberly, S., Brown, S. G., and Norris, G. A.: Methods for estimating uncertainty in factor analytic solutions, Atmos. Meas. Tech.,7,3(2014-03-27), 6, 7593-7631, 2014.

Reviewer #3 Comment NO.6: *Line 163- Please provide the Q robust values for different PMF solutions in an SI figure. This would be really helpful in showing why you picked the 6 factor solution as the optimal PMF resolved solution.*

Response to reviewer comment NO.6: Thanks for the reviewer's suggestion. To determine the optimal number of source factors, a string of effective test, in which factors number was from four to nine, was carried out. The resulting Q parameters were shown in Figure 12 (**Figure S4** in the supplementary materials). Obviously, there was a lowest Q_{Robust} value (13087) at six factors in moving from four to nine factors. Although $Q_{expected}$ has been decreasing in the process, $Q/Q_{expected}$ shared similar variation with Q_{Robust} showing the lowest value at six factors (1.3).

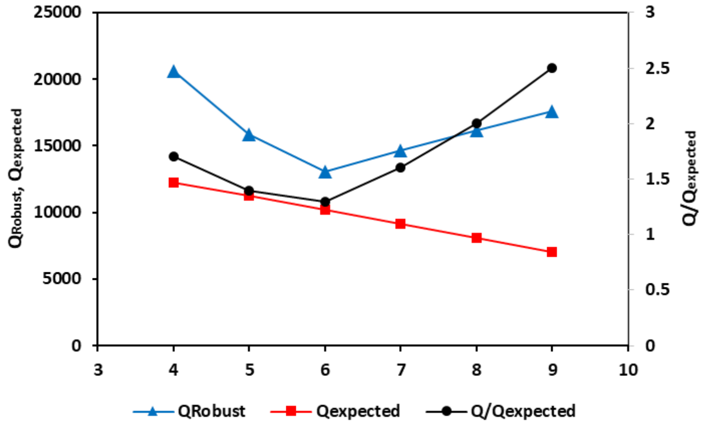


Figure 12. The variation of Q parameters from four factors to ten factors

Reviewer #3 Comment NO.7: *Line 166- In addition to briefly touching the results of your uncertainty analysis, you need to mention the uncertainty analysis results in detail (more discussions can be found in PMF source apportionment papers)*

Response to reviewer comment NO.7: Uncertainty of PMF model is usually estimated by bootstrapping (BS), displacement (DISP), and bootstrapping with displacement (BS-DISP). Here, characteristics of factors nearby six, where Q_{Robust} was relative lower, were explored. With five factors, three factors were mapped 100% of BS, while industry source and traffic source were mapped 92% and 94%, respectively of runs. There were no swaps with DISP, and 100% of the BS-DISP runs were successfully. At six factors, results were more stable with all factors mapped in BS in 100% (Table 6 below, also Table S2 in the supplementary materials), no swaps occurred with DISP and all BS-DISP runs were successfully. However, the solution became less stable in moving from six to seven factors. The new sea salt factor was only mapped in BS in 87% and coal combustion factor was mapped in BS in 89%, traffic source factor was mapped in 93%, other factors were mapped in 100% of runs. No swaps were found in DISP. Therefore, based on the above analysis, six factors were found to be the optimal solution in this study.

Table 6 Percentage of BS factors assigned to each base case factor with a correlation threshold of 0.6.

Boot Factor	Secondary	Industrial	Dust	Traffic	Coal	Biomass

1	100	0	0	0	0	0
2	0	100	0	0	0	0
3	0	0	100	0	0	0
4	0	0	0	100	0	0
5	0	0	0	0	100	0
6	0	0	0	0	0	100

Reviewer #3 Comment NO.8: *—Why the simulation period for footprint model, and NAQPMS model are not the same? For example, the footprint simulation was performed from 1-31 December while the NAQPMS model analysis was performed from 10th of November to 15th of December.*

Response to reviewer comment NO.8: The simulation period for footprint model and NAQPMS model are not the same because each analysis was performed by different research group. Based on the input data availability, the footprint simulation was performed from 1-31 December while the NAQPMS model analysis was carried out from 10th of November to 15th of December. Therefore, in this study, we use the data from December 1st to 15th for the analysis when both NAQPMS and the footprint results are used.

Reviewer #3 Comment NO.9: *Line 247- Please add a couple of references for the following sentence: In general, the large contribution of SIA, OM as well as the high OC/EC ratio indicated the importance of secondary formation in winter in Beijing, while the high concentration of species like SO42-and K suggested a significant contribution of combustion sources to PM2.5.*

Response to reviewer comment NO.9: Several references have been added as follows (Page 11, line 260):

In general, the large contribution of SIA, OM as well as the high OC/EC ratio indicated the importance of secondary formation in winter in Beijing (Sun et al., 2016b), while the high concentration of species like SO_4^{2-} *and K suggested a significant contribution of combustion sources including coal combustion and biomass burning to* $PM_{2.5}$ *(Achad et al., 2018; Chen et al., 2017; Li et al., 2017b).*

Reference

[1] Achad, M., Caumo, S., de Castro Vasconcellos, P., Bajano, H., Gómez, D., and Smichowski, P.: Chemical markers of biomass burning: Determination of levoglucosan, and potassium in size-classified atmospheric aerosols collected in Buenos Aires, Argentina by different analytical techniques, Microchem. J., 139, 181-187, 2018.

[2] Chen, S., Guo, Z., Guo, Z., Guo, Q., Zhang, Y., Zhu, B., and Zhang, H.: Sulfur isotopic fractionation and its implication: Sulfate formation in $PM_{2.5}$ and coal combustion under different conditions, Atmos. Res., 194, 142-149, 2017.

[3] Li, H., Zhang, Q., Zhang, Q., Chen, C., Wang, L., Wei, Z., Zhou, S., Parworth, C., Zheng, B., and Canonaco, F.: Wintertime aerosol chemistry and haze evolution in an extremely polluted city of the North China Plain: significant contribution from coal and biomass combustion, Atmos. Chem. Phys., 17, 4751-4768, 2017b.

[4] Sun, Y., Chen, C., Zhang, Y., Xu, W., Zhou, L., Cheng, X., Zheng, H., Ji, D., Li, J., and Tang, X.: Rapid formation and evolution of an extreme haze episode in Northern China during winter 2015, Sci. Rep., 6, 27151, 2016b.

Reviewer #3 Comment NO.10: *Line 255- As a general comment, you need to add references while mentioning different chemical components as tracers of a specific source. For example, references are required for the fact that K is a tracer of biomass burning..*

Response to reviewer comment NO.10: Several references have been added as follows (Page 11, line 270):

The proportion of K, Pb, As and Se, which were tracers of biomass burning and coal combustion (Achad et al., 2018; Chen et al., 2017; Vejahati et al., 2010), increased with $PM_{2.5}$ concentration. While the contribution of Ca, Ba, Fe, tracers of dust source (Amato et al., 2013; Shen et al., 2016), decreased with $PM_{2.5}$ concentration.

Reference

[1] Achad, M., Caumo, S., de Castro Vasconcellos, P., Bajano, H., Gómez, D., and Smichowski, P.: Chemical markers of biomass burning: Determination of levoglucosan, and potassium in size-classified atmospheric aerosols collected in Buenos Aires, Argentina by different analytical techniques, Microchem. J., 139, 181-187, 2018.

[2] Amato, F., Schaap, M., van der Gon, H. A. D., Pandolfi, M., Alastuey, A., Keuken, M., and Querol, X.: Short-term variability of mineral dust, metals and carbon emission from road dust resuspension, Atmos. Environ., 74, 134-140, 2013.

[3] Chen, S., Guo, Z., Guo, Z., Guo, Q., Zhang, Y., Zhu, B., and Zhang, H.: Sulfur isotopic fractionation and its implication: Sulfate formation in $PM_{2.5}$ and coal combustion under different conditions, Atmos. Res., 194, 142-149, 2017.

[4] Shen, Z., Sun, J., Cao, J., Zhang, L., Zhang, Q., Lei, Y., Gao, J., Huang, R.J., Liu, S., and Huang, Y.: Chemical profiles of urban fugitive dust $PM_{2.5}$ samples in Northern Chinese cities, Sci. Total Environ., 569, 619-626, 2016.

[5] Vejahati, F., Xu, Z., and Gupta, R.: Trace elements in coal: Associations with coal and minerals and their behavior during

coal utilization-A review, Fuel, 89, 904-911, 2010.

Reviewer #3 Comment NO.11: *Line 260-275: Unfortunately, the source apportionment profiles are not distinguished well. For example, K as a tracer of biomass burning has higher percentage of contribution in Industrial sources rather than the biomass burning. In addition, we have significant loadings of Na+ and Ni (which are not tracers of biomass burning) in biomass burning profile. How do you justify your source profiles?*

Response to reviewer comment NO.11: The detailed explanation of high fraction of K in industrial source and Ni in biomass burning source could be found in Response to reviewer comment NO.5 (Page 9-13 in this response file). Also, in Response to reviewer comment NO.5 and Response to reviewer comment NO.6 (Page 13-16 in this response file), we identified factors and justify our results by comparing the source profiles from PMF results in this study with those of specific emission sources reported in previous studies, and by good correlations between the tracers of identified sources and sources mass concentrations.

Reviewer #3 Comment NO.12: *Line 335- How do you compensate the lack of data for regional and local contribution from the NAQPMS model for the EP4?*

Response to reviewer comment NO.12: For better understanding of the evolution of EP4, the potential source contribution function (PSCF) model could be conducted to justify the result of the footprint model and compensate the lack of NAQPMS model results. The PSCF model was established by Malm et al. (1986). Total potential source contribution function (TPSCF) model was then developed based on this method by integrating air trajectories from different endpoint heights (Cheng et al., 1993). With combination of pollutant concentration ($PM_{2.5}$) and air mass transport information, TPSCF model was used for analyzing the dominant transport pathways to a certain receptor site (Liu et al., 2017). More detailed information about TPSCF model could be found in Liu et al. (2017). The TPSCF result during EP4 is shown in Figure 13. With higher TPSCF value (in orange, pink and red), the potential contribution to $PM_{2.5}$ at the sampling site (Beijing) increased. From Figure 13, it could be seen that the high TPSCF value concentrated in the southwestern area to Beijing (mostly in Hebei province), indicating that regional transport contributed significantly to $PM_{2.5}$ in Beijing during EP4.

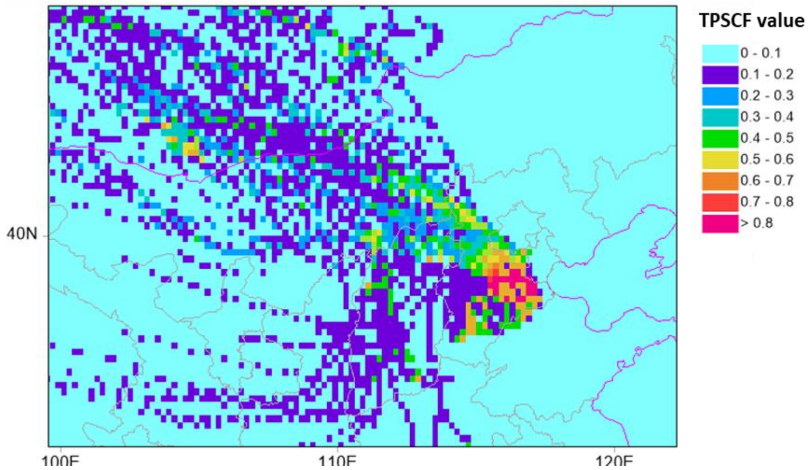


Figure 13. TPSCF results during EP4

Reference

[1] Cheng, M.D., Hopke, P.K., Barrie, L., Rippe, A., Olson, M., and Landsberger, S.: Qualitative determination of source regions of aerosol in Canadian high arctic. Environ. Sci. Technol., 27 (10), 2063-2071, 1993.

[2] Liu, Y., Yan, C., and Zheng, M.: Source apportionment of black carbon during winter in Beijing, Sci. Total Environ., 618, 531-541, 2017.

[3] Malm, W., Johnson, C., and Bresch, J.: Application of principal components analysis for purposes of identifying source-receptor relationships. In: Pace, T.G. (Ed.), Receptor Methods for Source Apportionment. Air Pollution Control Association, Pittsburgh, 127–148, 1986.

Reviewer #3 Comment NO.13: *Line 425- Authors should include the limitations of their research. Please add the limitations as a separate session.*

Response to reviewer comment NO.13: We agree that this comment is very important. A new session has been added to the end of the manuscript (Page 17-18, line 420-433) as follows:

3.5 Future prospect

In this study, the high-time resolution online measurement was conducted by Xact, IGAC and the

Sunset OCEC analyzer, which could measure inorganic species including water-soluble ions, elemental components, OC and EC. As a result, most of the tracers selected for PMF source apportionment were inorganic species. In previous studies based on online measurement, organic tracers are also not commonly used due to current technical difficulty in carrying out online and quantitative measurements of organic species with high-time resolution (Gao et al., 2016; Li et al., 2017e; Peng et al., 2016). However, some organic tracers are believed to be more specific for certain sources, such as levoglucosan for biomass burning, hopane and sterane for traffic source, and cholesterol for cooking source (Fraser et al., 2000; Yin et al., 2010; Zhao et al., 2015). Therefore, future online measurement of organic species could be conducted, which will be very helpful in identifying sources. Besides, vertical measurement of $PM_{2.5}$ *is important for better understanding of sources and regional transport of* $PM_{2.5}$ *in Beijing. Li et al. (2017) found that the height of regional transport ranged from 200 to 700 m above ground level using the NAQPMS model. In the future, the integration of ambient measurement with the air quality model should be considered in a vertical level as well.*

Reference

[1] Fraser, M. P., Lakshmanan, K.: Using Levoglucosan as a molecular marker for the long-range transport of biomass combustion aerosols, Environ. Sci. Technol., 34(21), 4560-4564, 2000.

[2] Gao, J., Peng, X., Chen, G., Xu, J., Shi, G.L., Zhang, Y.C., and Feng, Y.C.: Insights into the chemical characterization and sources of $PM_{2.5}$ in Beijing at a 1-h time resolution, Sci. Total Environ., 542, 162-171, 2016.

[3] Li, Y., Chang, M., Ding, S., Wang, S., Ni, D., and Hu, H.: Monitoring and source apportionment of trace elements in $PM_{2.5}$: Implications for local air quality management, J. Environ. Manage., 196, 16-25, 2017e.

[4] Peng, X., Shi, G.-L., Gao, J., Liu, J.-Y., HuangFu, Y.Q., Ma, T., Wang, H.T., Zhang, Y.C., Wang, H., and Li, H.: Characteristics and sensitivity analysis of multiple-time-resolved source patterns of $PM_{2.5}$ with real time data using Multilinear Engine 2, Atmos. Environ., 139, 113-121, 2016.

[5] Yin, J., Harrison, R. M., Chen, Q., Rutter, A., and Schauer, J. J.: Source apportionment of fine particles at urban background and rural sites in the UK atmosphere, Atmos. Environ., 44, 841-851, 2010.

[6] Zhao, X., Hu, Q., Wang, X., Xiang, D., He, Q., Zhou, Z., Shen, R., Lü, S., Liu, T., and Fu, X.: Composition profiles of organic aerosols from Chinese residential cooking: case study in urban Guangzhou, south China, J. Atmos. Chem., 72, 1-18, 2015.

High-time Resolution Source Apportionment of $PM_{2.5}$ in Beijing with Multiple Models

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Abstract

Beijing has suffered from heavy local emissions as well as regional transport of air pollutants, resulting in severe atmospheric fine particle ($PM_{2.5}$) pollution. This study developed a combined method to investigate source types of $PM_{2.5}$ and its source regions during winter 2016 in Beijing, which include the receptor model (Positive Matrix Factorization, PMF), footprint, and an air quality model. The ~~receptor~~ PMF model was performed with high-time resolution measurements of trace elements, water soluble ions, organic carbon, and elemental carbon using online instruments during the wintertime campaign of the Air

Pollution and Human Health-Beijing (APHH-Beijing) program in 2016. Source types and their contributions estimated by ~~the receptor~~ PMF model ~~(Positive Matrix Factorization, PMF)~~ using online measurement were linked with source regions identified by the footprint model, and the regional transport contribution was estimated by an air quality model (the Nested Air Quality Prediction Model System, NAQPMS) to analyze the specific sources and source regions during haze episodes. Our results show that secondary and biomass burning sources were dominated by regional transport while the coal combustion source showed an increased local contribution, suggesting that strict control strategies for local coal combustion in Beijing and a reduction of biomass burning and gaseous precursor emissions in surrounding areas were essential to improve air quality in Beijing. The combination of PMF with footprint results revealed that the secondary source was mainly associated with southern footprints (53%). The northern footprint was characterized by a high dust source contribution (11%) while industrial source~~s~~ increased with the eastern footprint (10%). The results demonstrated the power of combining receptor model-based source apportionment with other models in understanding the formation of haze episodes and to identify specific sources from different source regions affecting air quality in Beijing.

Keywords: Source apportionment, multiple models, regional transport

1. Introduction

Presently, haze in China has the characteristics of high frequency and long duration on a regional scale, and has influenced public life and human health (Xie et al., 2016). High concentrations of fine particulates, which can significantly reduce atmospheric visibility, are one of the main factors in the formation of haze episodes (Sun et al., 2016b; Watson et al., 2002; Yang et al., 2015). Previous studies have found that $PM_{2.5}$ can be emitted from various sources, including residential coal combustion, biomass burning, traffic-related sources, industrial sources and dust (Gao et al., 2016; Kotchenruther et al., 2016; Taghvaee et al., 2018; Watson et al., 2001; Zong et al., 2016~~; Gao et al., 2016~~). Therefore, it is important to have a better understanding of the major source types and their contribution to $PM_{2.5}$ in order to formulate science-based effective policies and regulations.

As the capital of China, Beijing has suffered from heavy emissions from various sources, resulting in severe $PM_{2.5}$ pollution (Li et al., 2017a; Lv et al., 2016). The source apportionment of $PM_{2.5}$ in Beijing has received great attention in recent years, which is mostly based on receptor models (Gao et al., 2016; Li et al., 2017~~ed~~; Lv et al., 2016; Song et al., 2006; Yang et al., 2016). Receptor models including the Chemical Mass Balance model (CMB) and Positive Matrix Factorization model (PMF) are the most commonly used methods of source apportionment in China, and are implemented by application of mathematical methods to measurements of chemical composition of fine particles at receptor sites (Cooper et al., 1980; Gao et al., 2016; Lv et al., 2016; Zheng et al., 2005). The receptor model can identify and quantify the contribution of multiple source types based on in-situ measurements and specific source tracers. Gao et al. (2016) employed two receptor models, PMF and Multilinear Engine 2 (ME2), to conduct high-time resolution source apportionment of $PM_{2.5}$ in summer in Beijing. The results showed that PMF and ME2 corresponded well with each other, and secondary sources were predominant in Beijing (38-39%). Similar source apportionment results were reported by in Peng et al. (2016) with secondary sources contributing 35-40%. Sun et al. (2016b) used online instruments and PMF to investigate the rapid evolution of a severe haze episode in winter in Beijing and showed the variation of chemical components during four stages of haze. By conducting receptor models based on high-time resolution online measurement, the source types and source contributions in Beijing have been analyzed in previous studies (Gao et al., 2016; Peng et al., 2016; Song et al., 2006). However, these studies still have limitations in that source apportionment based on receptor models are only restricted to one or several receptor sites without information about detailed source regions as well as the local and regional source contributions.

Previous studies have indicated that $PM_{2.5}$ pollution in Beijing has been significantly influenced by regional transport and meteorological conditions (Han et al., 2015; Li et al., 2017a; Zhao et al., 2013).

With the development of the function of source apportionment in air quality models, source regions and relative contributions to the receptor site could be quantitively estimated, based on emission inventory of pollution sources and meteorological fields (Burr et al., 2011; Kwok et al., 2013; Zhang et al., 2015). Li et al. (2016) found that regional transport highly contributed to the rapid increase stage, with the transport height ranged from 200 to 700 m above ground level with application of the Nested Air Quality Prediction Model System (NAQPMS). Han et al. (2018) used a regional air quality modeling system coupling with ISAM (Integrated Source Apportionment Method) and found that air pollutants derived from Hebei and Shandong provinces were major contributors to $PM_{2.5}$ in Beijing, with contributions up to 25% and 10%, respectively. The air quality model has advantages of analyzing spatial distribution and regional transport of pollutants, but it also has large uncertainties due to emission inventory, boundary layer meteorological processes and complex atmospheric chemical processes.

Due to the importance of the regional transport contribution to $PM_{2.5}$ in Beijing, the limitations of receptor models cannot be ignored. The source types and source contribution derived from receptor models can be combined with the contribution and direction of regional transport derived from chemical transport models. In this study, we employed the receptor model (PMF), the air quality model (NAQPMS) and a footprint model simultaneously based on high-time resolution online measurement data to investigate sources and regional transport of $PM_{2.5}$ in Beijing during November to December in 2016, as part of the Air Pollution and Human Health (APHH) campaign. The goal of the study is to link the contribution of different sources by PMF with the source regions by footprint, and the regional transport contribution by NAQPMS. The combination of multiple models gives greater power to identify specific sources and source regions.

2. Materials and methods

2.1 Online measurement of $PM_{2.5}$

Online sampling of $PM_{2.5}$ was conducted from November 2016 to December 2016 in winter, which was within the heating period of Beijing. The sampler was operated at the Peking University monitoring site (PKU, 39°59′21″N, 116°18′25″E) in the northwestern part of Beijing city. There are no obvious emission sources locally, except two major roads (150 m to the east and 200 m to the south). Situated in a mixed district of teaching, residential, and commercial areas, the sampling site is representative of the Beijing urban area (Liu et al., 2018; Yan et al., 2015). The sampling site ~~room~~ is located on the sixth floor of a teaching building within PKU. The inlet of the instrument is about 20 m above the ground.

Online $PM_{2.5}$ mass concentrations were continuously measured using a Tapered Element Oscillating Microbalance (TEOM 1405F, Thermo Fisher Scientific Inc.). Organic carbon (OC) and elemental carbon (EC) were simultaneously monitored by a Semi-continuous OCEC Carbon Aerosol Analyzer (Sunset Laboratory Inc.) with 1-h time resolution. The Sunset OC/EC analyzer uses a modified NIOSH 5040 thermal-optical protocol as its default protocol, which produces a relatively reliable determination of OC, EC, and the OCEC split (Bauer et al., 2009). More detailed information could be found in Bauer et al., 2009.

An in-situ Gas and Aerosol Compositions monitor (IGAC, Model S-611, Fortelice International Co.Ltd.), which could collect both gases and particles simultaneously, was applied to measure water-soluble ions online with 1-h time resolution in this study. A detailed description of IGAC can be found in Young et al. (2016). Briefly, IGAC was composed of three major units, including a Wet Annular Denuder (WAD) to collect gases into aqueous solution, a Scrub and Impact Aerosol Collector (SCI) to collect particles into solution and a sample analysis unit comprised of two ion chromatographs (DionexICS-1000) for analyzing anions and cations (IC). Ambient air was drawn through a PM_{10} inlet followed by a $PM_{2.5}$ cyclone at a flow rate of 16.7 L min^{-1}, and then gases and $PM_{2.5}$ were separately collected by WAD and SCI. Both gaseous and aerosol samples were injected into 10 mL glass syringes which were connected to the IC for analysis (30-min time resolution for each sample). The concentrations of eight water-soluble inorganic ions (e.g., NH_4^+, Na^+, K^+, Ca^{2+}, Mg^{2+}, SO_4^{2-}, NO_3^- and Cl^-) in the fine particles were measured.

Twenty-three trace elements in $PM_{2.5}$ were measured by an Xact 625 Ambient Metal Monitor

(Cooper Environmental Services LLC, USA) with 1-h time resolution. Among them twelve elements (e.g., K, Ca, Ba, Cr, Mn, Fe, Cu, Ni, Zn, As, Se, Pb) were selected for further analysis, while other trace elements (such as V, Co, Tl) were not used here due to the low concentrations (below the method detection limit). The ambient air was sampled on a Teflon filter tape inside the instrument through a $PM_{2.5}$ cyclone inlet at a constant flow rate of 16.7 L min^{-1}, and then the sample was automatically analyzed by nondestructive energy-dispersive X-ray fluorescence (XRF) to determine the mass of the species. This instrument has been documented with Environment Technology Verification (ETV) and certified by the US Environment Protection Agency (EPA, 2012). The detection limit of each species measured by the online instruments could be seen in Table S1.

Strict quality assurance (QA) and quality control (QC) protocols for online instruments were performed during the whole sampling period. For IGAC, the internal standard (LiBr) was added continuously to each sample and analyzed by the IC system during the analysis to check the stability of the IGAC instrument. During the sampling period, the mean concentrations of Li^+ and Br^- were within the range of three standard deviations, suggesting a stable condition of the IGAC (see Fig. S1). As shown in Fig. S2~~1~~, the slope of the linear fitting between the anions and cations was 0.93, and R^2 was 0.96. As for the OC/EC analyzer, external standard calibration using the stock sucrose solution was conducted before operation to calibrate carbon analysis. For the Xact, a Pd rod was used as automatic internal quality control to check the performance of the instrument on a daily basis (see Fig. S3). Additionally, a QA energy calibration test and QA energy level test were performed for a half hour after midnight every day to monitor any possible shift and instability of the XRF. During our field campaign, the Xact remained stable and reliable.

Chemical closure has been done between the measured and reconstructed $PM_{2.5}$. Organic matter (OM) was calculated as OM= 1.6 ×OC (Turpin and Lim, 2001). Mineral species was calculated as Mineral= 1.89 Al +2.14 Si + 1.4 Ca + 1.43 Fe + 1.66 Mg (Zhang et al., 2003). The concentrations of Al, Si, Fe and Mg were calculated by the concentration of Ca and the composition of urban soils of Beijing:

Al= 1.7Ca, Si= 7.3Ca, Fe=0.7Ca, Mg= 0.3Ca (An et al., 2016). “Others” were calculated by subtracting OM, EC, Mineral and secondary inorganic aerosol (SIA, including SO_4^{2-}, NO_3^-, NH_4^+) concentration from total $PM_{2.5}$ concentration. The correlation of measured and reconstructed $PM_{2.5}$ mass could be seen in Fig. S6 with R^2=0.892.

2.2 Methodology

2.2.1 Positive Matrix Factorization (PMF)

To qualitatively and quantitatively identify sources of $PM_{2.5}$ and estimate the associated contributions, the USEPA PMF 5.0 model was adopted in this study. The principle and detailed information on this model may be found in Paterson et al. (1999) and the EPA 5.0 Fundamentals and User Guide. Factor contributions and profiles were derived by minimizing the objective function Q in the PMF model, which was determined as follows (Norris et al., 2014; Paatero and Tapper, 1994; Paatero et al., 2014; Paatero, 1997):

$$Q = \sum_{i=1}^{n}\sum_{j=1}^{m}\left[\frac{x_{ij}-\sum_{k=1}^{p} g_{ik}f_{kj}}{u_{ij}}\right]^2 \qquad (1)$$

Data values below the MDL were substituted with MDL/2. Missing data values were substituted with median concentrations. If the concentration was less than or equal to the MDL, the uncertainty (Unc) was calculated using a fixed fraction of the MDL:

$$\mathrm{Unc} = \frac{5}{6}MDL \qquad (2)$$

If the concentration was greater than the MDL provided, the calculation was based on the following equation:

$$\mathrm{Unc} = \sqrt{(Error\ Fraction \times concentration)^2 + (0.5 \times MDL)^2} \qquad (3)$$

In total, nineteen chemical components were used in the PMF model, including OC, EC, Cl^-, SO_4^{2-}, NO_3^-, Na^+, NH_4^+, K, Ca, Ba, Cr, Mn, Fe, Cu, Ni, Zn, As, Se and Pb. To determine the optimal number of source factors, a string of effective test, in which factors number was from four to nine, was carried out.

The resulting Q parameters were shown in Figure 10. Obviously, there was a lowest Q_{Robust} value (13087) at six factors in moving from four to nine factors. Although $Q_{expected}$ has been decreasing in the process, $Q/Q_{expected}$ shared similar variation with Q_{Robust} showing the lowest value at six factors (1.3) (see Fig. S4).~~To determine the optimal number of source factors, a string of effective tests, in which factor number was from four to eight, was carried out and among which the best performance was found at six factors with the lowest Q_{Robust} and $Q/Q_{expected}$ value.~~ Bootstrapping (BS), displacement (DISP), and bootstrapping with displacement (BS-DISP) were conducted to analyze the uncertainty of the PMF model at six factors. The results were stable with all factors mapped in BS in 100% and no swaps with DISP and all BS-DISP runs, indicating a convincing source apportionment result (see Table S2).

2.2.2 Footprint analysis model

The footprint model developed by Peking University was used to simulate the potential source region of air pollution. The footprint is a transfer function in a diffusion problem linking the source and the measurement result at a point (receptor) (Pasquill and Smith 1983). That is,

$$\mathrm{c}(r) = \int_R Q(r+r')f(r,r')dr' \qquad (4)$$

where c is the measured concentration at spatial location r, Q is the source strength with spatial location $(r+r')$, f is the footprint or the transfer function and R is the integration domain. The footprint links point measurements (receptors) in the atmosphere to upstream forcings, in which turbulent dispersion plays a central role. The Lagrangian stochastic (LS) particle models was used to calculate the footprint function (Cai et al., 2007; Leclerc and Thurtell 1990; Kurbanmuradov and Sabelfeld 2000).

The meteorological data used to drive footprint model was provided by the Weather Research and Forecasting model (WRF-ARWv3.6.1) (http://www.wrf-model.org/), initialized using the Final Analysis (FNL) data from the United States National Centers for Environmental Prediction (NCEP). Two nested domains were used in this study with horizontal resolutions of 15 and 5 km, and 28 vertical levels. The simulation period was from November 1 to December 31, 2016, with a 12 h spin-up time before the start for each 48 h simulation. The domain of the footprint model is the same as the domain 2 in WRF which covers the North China Plain (500×600km), and the horizontal resolution is 2.5 × 2.5 km. Every hour, 5000 particles were released 10 m above the ground at the center of Beijing, and then each particle was tracked backward in time for 48 hours. The residence time of all particles in 0-100 m above the ground were recorded to obtain the footprint. This model has undergone rigorous theoretical discussion and verification and more detailed principle and calculation methods of the model can be found in Cai et al. (2007).

2.2.3 The Nested Air Quality Prediction Model System (NAQPMS)

In this study, the NAQPMS model was applied to analyze the contribution of local emissions and regional transport to $PM_{2.5}$ in winter in Beijing. NAQPMS is a 3-D Eulerian chemical transport model with terrain-following coordinates, developed by the Institute of Atmospheric Physics, Chinese Academy of Sciences (IAP/CAS) and has been validated by the Ministry of Environmental Protection of China (CMEP, 2013). The main modules in the model include horizontal and vertical advection and diffusion, dry and wet deposition, and gaseous, aqueous, aerosol and heterogeneous chemistry (Li et al., 2007; Li et al., ~~2017b~~2017c). A more detailed description of the model can be found in Li et al. (2008; 2014; 2016; 2017c~~b~~).

Three nested model domains were used in this study. The coarsest domain (D1) covered most of China and East Asia with a 27 km resolution. The second domain (D2) included most anthropogenic emissions within the North China Plain with a 9 km resolution. The innermost domain (D3) covered the Beijing-Tianjin-Hebei region at a 3 km resolution. The first level of model above the surface is 30 m in height, and the average vertical layer spacing between 30 m and 1 km is around 100 m. The MIX (http://www.meicmodel.org/dataset-mix.html) anthropogenic emission inventory was used (Li et al., 2017d~~e~~), with the original resolution of 0.25 °(about 25 km at middle latitudes) and the year of 2010. The

NAQPMS meteorological fields was provided by the Weather Research and Forecasting model (WRF-ARWv3.6.1) (http://www.wrf-model.org/) driven by the National Centers for Environmental Prediction (NCEP) Final Analysis (FNL) data. The simulation was conducted from November 10 to December 15, 2016.

2.2.4 The combination of multiple models

The footprint model was used to provide the direction of source regions while the NAQPMS model was run to calculate the contribution of local emission and regional transport. To verify the consistency between the two models, the footprint with a time resolution of 6 hours was divided into four types (local, south, north and east) according to the direction of potential source regions, and average local contributions of different types obtained from NAQPMS were calculated (See Table S3~~1~~). A typical example of different types of footprint can be seen in Fig. ~~S2~~S5. The average local contribution estimated by NAQPMS was highest for the local footprint (85%) and lower for south (68%), north (63%) and east footprints (66%). The results of the two models correlated well with each other.

Based on online measurement of $PM_{2.5}$ species including specific source tracers, the receptor model (PMF) could be used to obtain precise source apportionment result but with no information upon regional transport. Therefore, the footprint and NAQPMS model were simultaneously conducted and combined with the PMF model to link the source type and contribution to $PM_{2.5}$ in Beijing by receptor models with different source regions.

3. Results and discussion

3.1 Mass concentration and chemical composition of $PM_{2.5}$

Temporal variation of chemical composition of $PM_{2.5}$ during the field campaign was shown in Fig. 1. The statistical summary of identified species of $PM_{2.5}$ in the entire sampling period was summarized in Table S1. ~~Organic matter (OM) was calculated as OM= 1.8 ×OC (Pitchford et al., 2007). Mineral~~

~~species was calculated as Mineral= 1.89 Al +2.14 Si + 1.4 Ca + 1.43 Fe + 1.66 Mg (Zhang et al., 2003). The concentrations of Al, Si, Fe and Mg were calculated by the concentration of Ca and the composition of urban soils of Beijing: Al= 1.7Ca, Si= 7.3Ca, Fe=0.7Ca, Mg= 0.3Ca (An et al., 2016). Since the concentration of Al and Si were not directly measured by Xact, the calculated mineral component might be underestimated. "Others" were calculated by subtracting OM, EC, Mineral and secondary inorganic aerosol (SIA, including SO_4^{2-}, NO_3^-, NH_4^+) concentration from total $PM_{2.5}$ concentration.~~ Figure 1 shows that SIA and OM were the predominant $PM_{2.5}$ components in winter in Beijing, accounting for ~~55~~57% and ~~27~~24% of total $PM_{2.5}$ mass, respectively. The average concentration of OC was 20. 8±17.0 μg m^{-3}, and the average concentration of EC was 5.6~~59~~ ±4.4~~3~~ μg m^{-3}. The OC/EC ratio is often used to indicate the contribution of primary emission sources and secondary organic aerosols (SOA) (Lim et al., 2002; Zheng et al., 2014). In this study, the OC/EC ratio ranged from 1.36 to 7.92 with an average ratio of 3.91±0.91, which was lower than that in the winter of Beijing in 2013 (5.73 ±2.47) (Yan et al. 2015). SO_4^{2-} is the predominant ion in SIA with an average concentration of 23.5±20.8 μg m^{-3}, which was ~~higher than~~similar with that of NO_3^- (22.0±23.3 μg m^{-3}). The concentration of elemental components ranked from high to low as K> Fe> Ca> Zn> Pb> Mn> Ba> Cu> As> Cr> Se> Ni, with K contributing 2% to $PM_{2.5}$. In general, the large contribution of SIA, OM as well as the high OC/EC ratio indicated the importance of secondary formation in winter in Beijing (Sun et al., 2016b), while the high concentration of species like SO_4^{2-} and K suggested a significant contribution of combustion sources including coal combustion and biomass burning to $PM_{2.5}$ (Achad et al., 2018; Chen et al., 2017; Li et al., 2017b).

Figure 2 shows the large differences in chemical composition of $PM_{2.5}$ concentration between non-haze and haze episodes. The average concentration of $PM_{2.5}$ and identified species in different haze and non-haze periods were summarized in Table S4. Under low $PM_{2.5}$ concentration (< 50 μg m^{-3}), ~~the contribution of sulfate~~SO_4^{2-} was one of the major components of $PM_{2.5}$ ~~increased significantly~~ with the contribution of ~~(up to~~ around –24%~~)~~. When $PM_{2.5}$ was from 50~150 μg m^{-3}, OM was the dominant composition (about ~~40~~38%). When $PM_{2.5}$ was greater than 150 μg m^{-3}, the contribution of SIA increased with the concentration level (up to 55%). The contribution of mineral components decreased from 8% to 2% when $PM_{2.5}$ concentration increased from below 50 µg m^{-3} to over 250 µg m^{-3}. The proportion of K, Pb, As and Se, which were tracers of biomass burning and coal combustion (Achad et al., 2018; Chen et al., 2017; Vejahati et al., 2010), increased with $PM_{2.5}$ concentration. While the contribution of Ca, Ba, Fe, tracers of dust source (Amato et al., 2013; Shen et al., 2016), decreased with $PM_{2.5}$ concentration. Taken together, all these variations of source specific $PM_{2.5}$ compositions suggested more significant influence of combustion sources to $PM_{2.5}$ in haze episodes and relatively higher contribution of dust source in non-haze periods.

3.2 Source apportionment during haze and non-haze periods

To conduct high-time resolution source apportionment in Beijing, a PMF model was applied to 1-h online measurement data. The six-factor solution gave the best performance. The factor profile for each factor is shown in Fig. ~~S3~~S7. Contribution of different factors to $PM_{2.5}$ were estimated after considering major sources in Beijing, the similarity of the PMF source profiles with relevant source emission profiles, and distinctively different marker species for different sources. Factor 1 was heavily weighted by secondary inorganic ions (SO_4^{2-}, NO_3^- and NH_4^+) and moderately weighted by OC, which was typical of the secondary source profiles (Gao et al., 2016; Peng et al., 2016; Shi et al., 2017). Factor 2 was highly loaded on metal species including Mn, Fe, Cu and Zn, which were mostly used as indicators for industrial sources (Hu et al., 2015; Li et al., 2017e; Pan et al., 2015; Yu et al., 2013). Factor 3 presented high loading of Ca, Ba, and Fe, and the two crustal elements were mainly emitted from dust sources (Amato et al., 2013; Shen et al., 2016). Factor 4 was mostly loaded by EC, OC and moderately loaded by Cu and Zn, which were mainly emitted from lubricant additive of vehicles (Kim et al., 2003; Tao et al., 2014) and wear of brake and tyre (Pant and Harrison, 2013; Perez et al., 2010). High loading of As, Se and moderate loading of OC, EC were observed in Factor 5, indicating a typical source profile of coal combustion (Vejahati et al., 2010). Factor 6 was characterized by high loading of K, SO_4^{2-} and OC, which were identified as indicators of biomass burning (Duan et al., 2004). In previous studies, cooking source could be one of the important sources of $PM_{2.5}$, contributing to about 10% on average in East Asia (Chafe et al., 2014; Sun et al., 2013), but in this study cooking source was not identified by PMF due to the lack of organic tracers. The relationships between the tracers of identified sources and sources mass concentrations were shown in Fig. S8.

The source apportionment result of PMF in winter in Beijing was shown in Fig. 3. During the campaign, the source contribution in Beijing ranked as secondary source (44%) > traffic source (18%) > coal combustion (16%) > biomass burning (9%) > industrial source (8%) > dust (5%). The high contribution of secondary sources in winter was similar to previous studies (Gao et al., 2016; Peng et al., 2016; Zhang et al., 2013), which might be attributed to regional transport and heterogeneous reactions (Ma et al., 2017).

Considering data integrity and representativeness, four typical pollution episodes (EP1-4) and two non-haze periods (NH1 and NH2) were selected. The average $PM_{2.5}$ concentrations in four haze episodes were all above ~~75~~ 97 μg m^{-3} (see Table S4~~1~~). EP1 (Nov. 14-19) and EP2 (Nov. 24-27) represented the pollution episodes in November, and EP3 (Dec. 1-5) and EP4 (Dec. 16-21) were two severe pollution processes in December. The four pollution episodes were characterized by low wind speed around 2 m s^{-1} and high relative humidity (RH) compared to non-haze periods (see in Table 1). The chemical composition and sources of the four pollution episodes varied from each other, but relatively high contribution of secondary sources was observed in all episodes (32-57%), and the contribution increased with $PM_{2.5}$ concentration (see Fig. 2). EP4 was characterized by the highest contribution of secondary source (57%). The contribution of coal combustion and industrial sources in EP1 was the most significant compared with the other episodes, which were 22% and 17% respectively. The traffic source contribution in EP2 and EP3 was higher than other pollution episodes, accounting for about 22%. The source contribution in non-haze periods is significantly different from that in pollution episodes. The contribution of secondary sources in the two non-haze periods, NH1 (Nov. 22-23) and NH2 (Dec. 13-15), decreased to 18% and 25%, while traffic and dust source contribution to $PM_{2.5}$ increased to about 30% and 10%, which could be influenced by local emission and regional transport from northern areas to Beijing.

Generally, secondary source was predominant (~50%) to $PM_{2.5}$ in pollution episodes, while traffic source (~30%) became more important in non-haze periods. However, source contribution of $PM_{2.5}$ could vary from episode to episode. EP1 was more influenced by primary sources while EP4 was characterized by high secondary source contribution (57%).

3.3 Evolution of different types of haze episodes

The high-time resolution source apportionment result by PMF was combined with the NAQPMS and footprint modelling outcomes to investigate the variation of source types and contributions with source regions in different haze episodes in Beijing. EP1 and EP4, with the longest duration and significantly different source compositions, were selected as two case episodes for further analysis.

3.3.1 A haze episode dominated by local emission

Figure 4 shows the variation of sources and local contribution and Fig. 5 shows the footprint regions and daily source apportionment results by PMF in EP1. The spatial mass concentrations of $PM_{2.5}$, wind speed and wind direction during EP1 by NAQPMS could be found in Fig. S9. It can be seen that EP1 was characterized with high local contribution (69%-89%) and primary source contribution to $PM_{2.5}$. On November 14, the footprint located in the northeastern part to Beijing (mainly Inner Mongolia) with low $PM_{2.5}$ concentration while the contribution of dust source was significant (52%). On November 16 when the formation stage of EP1 started, the footprint concentrated in local areas of Beijing and the local contribution by NAQPMS (82%) increased simultaneously. The daily average source contribution ranked as traffic source (29%) > coal combustion (28%) > industrial source (15%) > dust and secondary source (12%) > biomass burning (6%). The contribution of primary sources especially for traffic source increased when footprints were primarily located in local area.

The relationship between source apportionment (1 h) and the footprint model (6 h) could also be found in daily variation of November 17 (see Fig. 5). From 01:00 to 12:00 of the day, the footprint remained in local areas while primary sources were predominant. However, with the footprint changed to southwestern areas to Beijing from 13:00 to 18:00, the contribution of secondary sources increased significantly to 42%. After the footprint changed back to local type from 19:00 to 24:00, the secondary source contribution decreased to previous level (19%).

3.3.2 A haze episode dominated by regional transport

Figure 6 shows the variation of sources and Fig. 7 shows the footprint regions and daily source apportionment results by PMF in EP4. Different from EP1, the footprint in EP4 was mostly located in the southwestern area to Beijing, where there were heavy polluted cities including Baoding and Shijiazhuang (see Fig. 7). The daily local and regional contribution by NAQPMS of this episode was not provided due to lack of data. From the formation stage (December 16 -17) to the peak (December 20) of EP4, the contribution of secondary sources increased from 34% to 58%, while the contribution of coal combustion and biomass burning were also significant among primary sources (see Fig. 6). Figure 7 shows that the footprint on December 17 was more concentrated in local and eastern areas to Beijing, while it gradually moved to southwestern areas along with the increase of $PM_{2.5}$ concentration and the secondary source contribution.

The above results confirmed that high-time resolution source apportionment result could be integrated with footprint and NAQPMS model to identify the rapid evolution of different episodes - EP1 was an episode mainly caused by local emission from transportation and coal combustion while EP4 was typical for regional transport from southwestern areas to Beijing with increasing contributions of secondary sources.

3.4 Relationship of $PM_{2.5}$ sources by PMF with regional transport estimated by NAQPMS

~~In order to further determine the relationship of $PM_{2.5}$ sources by PMF with regional transport in winter in Beijing, the results of PMF and NAQPMS during the whole campaign were combined for analysis.~~

~~3.4.2~~3.4.1 Sources dominated by local emission and regional transport

Receptor models which are used for source apportionment have the limitation that it could not quantify the local or regional transport contribution. Therefore, the receptor model was combined with the chemical transport model NAQPMS to investigate the correlation of source contribution with local/regional transport. As shown in Sect. 3.2, secondary and combustion sources were predominant in haze episodes in Beijing. To better control those major sources in winter, it is essential to determine correlation of source contribution with the contribution of local emission or regional transport. Figure 8 shows the correlations of relative contribution of secondary sources, coal combustion and biomass burning sources by PMF with local contribution by NAQPMS during the sampling period. The results showed that for $PM_{2.5}$ in Beijing, secondary source contribution decreased when local emission was more significant ($p<0.05$, $r=0.4$) while coal combustion, as a primary combustion source, showed an increasing trend along with local contribution estimated by NAQPMS ($p<0.05$, $r=0.3$). Comparing with Fig. 8 (b) and (c), the two primary combustion sources showed opposite relationship with local contribution, indicating that the pollutants from biomass burning were mainly transported from surrounding areas outside of Beijing while those from coal combustion were more influenced by local emission. According to previous studies, biomass burning was an important source in provinces around Beijing including Shandong, Hebei and Inner Mongolia (Khuzestani et al., 2018; Sun et al., 2016a; Zhang et al., 2010; Zhao et al., 2012; Zong et al., 2016). The surrounding provinces and cities of Beijing are shown in Fig. ~~S4~~S10. The results suggested that local-emitted coal combustion contributed significantly to $PM_{2.5}$ in Beijing in winter 2016 and the strict control strategies for coal combustion were essential to improve air quality in Beijing. In the meantime, more control of biomass burning and precursors of secondary source~~s~~ in surrounding areas are also needed to mitigate air pollution in Beijing.

~~3.4.3~~3.4.2 Sources dominated in different potential source regions

The combination of PMF result with footprint model was used to further identify specific source type and contribution in different source regions. As mentioned in Sect. 2.2.4, the footprint with the time resolution of 6 hours was divided into four types (local, south, north and east) according to the direction of potential source regions. The typical examples of different types of footprint were shown in Fig. ~~S2~~S5. The local footprint referred to the cases with source region located within Beijing. The south footprint mainly covered southwest areas in Hebei province including Baoding, Shijiazhuang and Xingtai. The north footprint included Zhangjiakou and Inner Mongolia. The east footprint covered the north part of Hebei such as Tangshan and Qinhuangdao and the south part of Liaoning province. The local footprint was predominant in winter in Beijing (N=79) with the contribution of 38%, followed by north and south footprint (N=51, 45 respectively). The amount of east footprint was the lowest in winter. The average value and box chart of source contribution in four types of footprint during the whole sampling period were shown in Fig. 9. It could be seen that local footprint was characterized by traffic (23%) and coal combustion sources (25%), while the contribution of secondary source (26%) was the lowest among the four types. On the contrary, secondary source was predominant in south footprint cases with the contribution of 53%, while the contribution of traffic source decreased to 15%. The results corresponded well with the analysis of two typical episodes in Sect. 3.3. North footprint was characterized by the highest contribution of dust source (11%), which could be influenced by dust transported from Inner Mongolia (Hoffmann et al., 2008; Park et al., 2014). East footprint, which mainly covered heavy industrial areas such as Tangshan and Shenyang, showed higher contribution of industrial sources (10%) and coal combustion sources (18%). Figure 9 (b) shows that the variation of source contribution was the smallest in local footprint, indicating a relatively stable local emission of Beijing, while the source contribution varied more significantly with east footprint.

The results of PMF and footprint model showed that source contribution in winter in Beijing was influenced by the potential source regions, and the predominant source could change specifically for different footprint type, which might suggest that source apportionment and footprint analysis need to be combined to better control specific sources from different source regions.

3.5 Future prospect

In this study, the high-time resolution online measurement was conducted by Xact, IGAC and the Sunset OCEC analyzer, which could measure inorganic species including water-soluble ions, elemental components, OC and EC. As a result, most of the tracers selected for PMF source apportionment were inorganic species. In previous studies based on online measurement, organic tracers are also not commonly used due to current technical difficulty in carrying out online and quantitative measurements of organic species with high-time resolution (Gao et al., 2016; Li et al., 2017e; Peng et al., 2016). However, some organic tracers are believed to be more specific for certain sources, such as levoglucosan for biomass burning, hopane and sterane for traffic source, and cholesterol for cooking source (Fraser et al., 2000; Yin et al., 2010; Zhao et al., 2015). Therefore, future online measurement of organic species could be conducted, which will be very helpful in identifying sources. Besides, vertical measurement of $PM_{2.5}$ is important for better understanding of sources and regional transport of $PM_{2.5}$ in Beijing. Li et al. (2017) found that the height of regional transport ranged from 200 to 700 m above ground level using the NAQPMS model. In the future, the integration of ambient measurement with the air quality model should be considered in a vertical level as well.

4. Summary

High-time resolution online measurements of $PM_{2.5}$ were conducted during the APHH winter campaign in Beijing. Considering the limitation of receptor models which could not identify and quantify regional transport, the receptor model PMF was combined with multiple models including NAQPMS and footprint model to analyze the specific sources from different source regions during haze episodes in Beijing. The source apportionment results by PMF during our sampling period showed that secondary source was predominant (~50%) to $PM_{2.5}$ in pollution episodes, while traffic source (~30%) became more important in non-haze periods. Source contribution of $PM_{2.5}$ could vary from episode to episode.

The multiple models were combined to analyze the evolution of two typical pollution episodes in Beijing. The high-time resolution results indicated that source contribution could vary rapidly and significantly with source regions within different type of haze episodes. EP1, with local concentrated footprint and high local emission, was characterized by coal combustion and traffic sources while EP4 with more southwestern footprint was typical for high secondary source contribution. The relationship of $PM_{2.5}$ sources by PMF with regional transport during the whole sampling period was further investigated. As the predominant sources of $PM_{2.5}$ in Beijing, secondary and biomass burning source were more influenced by regional transport while coal combustion source increased with local contribution. The source regions of $PM_{2.5}$ in Beijing were classified into four types and source contribution varied significantly with potential source regions, with traffic source dominated in local footprint, secondary source dominated in south footprint, dust and industrial source increased in north and east footprint, respectively. The results suggested that source contribution of $PM_{2.5}$ in winter, Beijing could change significantly along with the contribution and direction of regional transport. Therefore, the combined use of receptor model, meteorological model and chemical transport model was important to identify specific sources from different source regions.

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Tables and Figure Legends

Table 1. ~~$PM_{2.5}$ and m~~Meteorological conditions during pollution episodes and non-haze periods.

Figure 1. Chemical composition of $PM_{2.5}$ during sampling period (red for SO_4^{2-}, blue for NO_3^-, yellow for NH_4^+, green for OM, black for EC, pink for mineral, and grey for others).~~Chemical composition of $PM_{2.5}$ during sampling period (red for sulfate, blue for nitrate, yellow for ammonium, green for OM, black for EC, pink for mineral, and grey for others).~~

Figure 2. Variation of (a) chemical composition and (b) elemental species with $PM_{2.5}$ concentration (the white bars represent the frequency of $PM_{2.5}$ concentration).~~Variation of chemical composition with $PM_{2.5}$ concentration.~~

Figure 3. Source contribution of $PM_{2.5}$ (a) in the whole sampling period and (b) in different pollution episodes and non-haze periods (yellow for dust source, green for biomass burning, pink for industrial source, red for coal combustion, black for traffic source, and blue for secondary source).

Figure 4. Variation of sources and local contribution during EP1. The above pie charts show the daily local (Beijing as BJ) and regional contribution (labeled as Others). The pie charts below show the daily source type and contribution.

Figure 5. (a) Source regions by the footprint model and (b) daily source apportionment results by PMF in EP1.~~Source regions by the footprint model and daily source apportionment results by PMF in EP1.~~

Figure 6. Source contribution in EP4. The pie charts show the daily source type and contribution.

Figure 7. (a) Source regions by the footprint model and (b) daily source apportionment results by PMF in EP4.~~Source regions by the footprint model (every 6 h) and daily source apportionment results by PMF in EP4.~~

Figure 8. Correlations of local contribution by NAQPMS with the relative contribution by PMF of (a) secondary source, (b) coal combustion source and (c) biomass burning source.

Figure 9. (a) The average source contribution (in percentage) for each type of footprint, and (b) box chart

of source contribution in four types of footprint during the whole sampling period. N in (a) represents for the number of cases. The capital letters in (b) stands for the type of footprint (L for local; S for south; N for north; E for east) and the lowercases stands for different sources (s for secondary source, c for coal combustion, t for traffic source, i for industrial source, d for dust, and b for biomass burning).

Table 1. ~~$PM_{2.5}$ and m~~Meteorological conditions during pollution episodes and non-haze periods.

	EP1	EP2	EP3	EP4	Non-haze	Average
~~$PM_{2.5}$ (μg m^{-3})~~	~~98~~	~~144~~	~~115~~	~~242~~	~~11.1~~	~~110~~
Wind speed (m s^{-1})	2.24	2.26	2.36	2.04	4.17	2.48
Temperature (°C)	7.48	2.94	5.36	2.63	-2.05	3.42
Relative humidity (%)	54.5	38.2	38.8	49.4	24.1	43.5
Pressure (hPa)	1012.5	1016.5	1016.3	1016.1	1027.9	1017.4

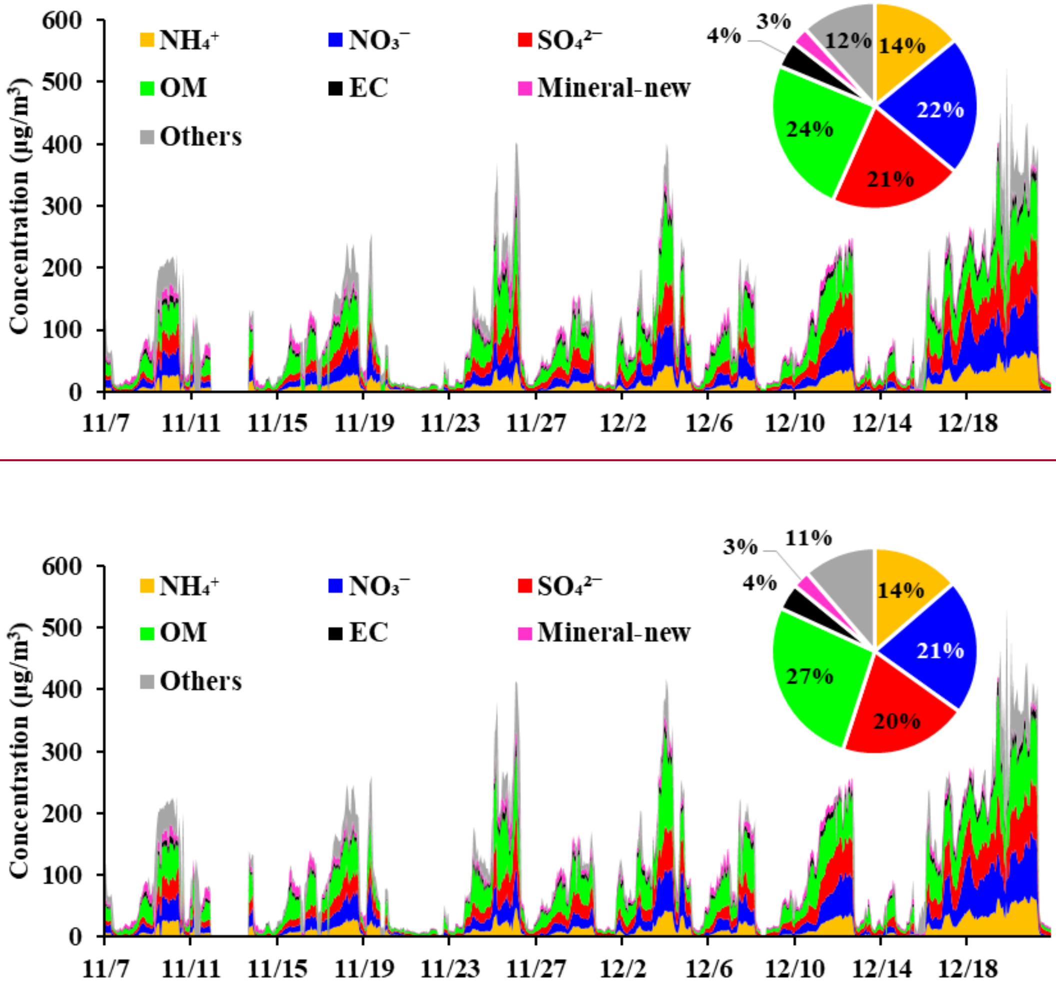
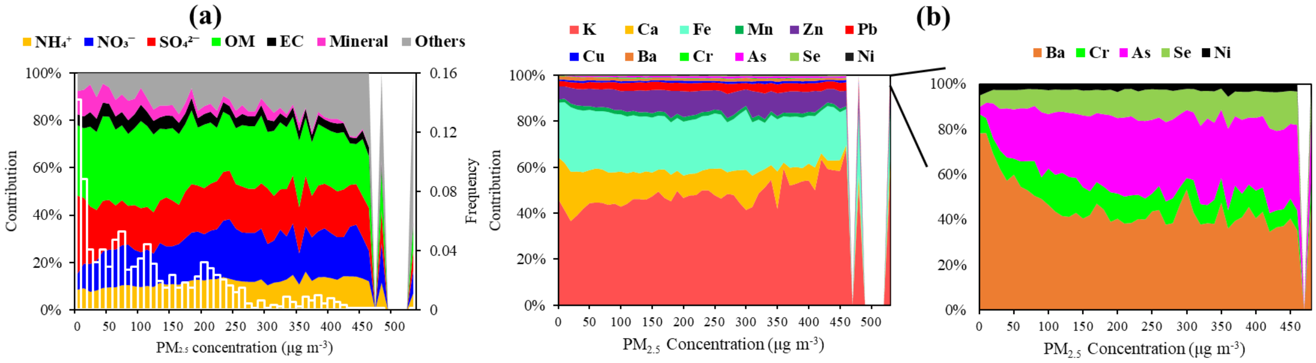


Figure 1. Chemical composition of $PM_{2.5}$ during sampling period (red for SO_4^{2-}~~sulfate~~, blue for NO_3^-~~nitrate~~, yellow for NH_4^+~~ammonium~~, green for OM, black for EC, pink for mineral, and grey for others).



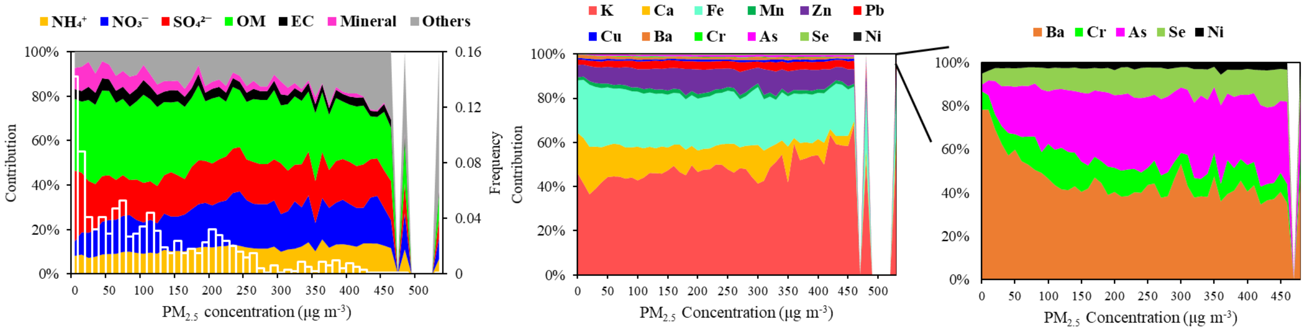
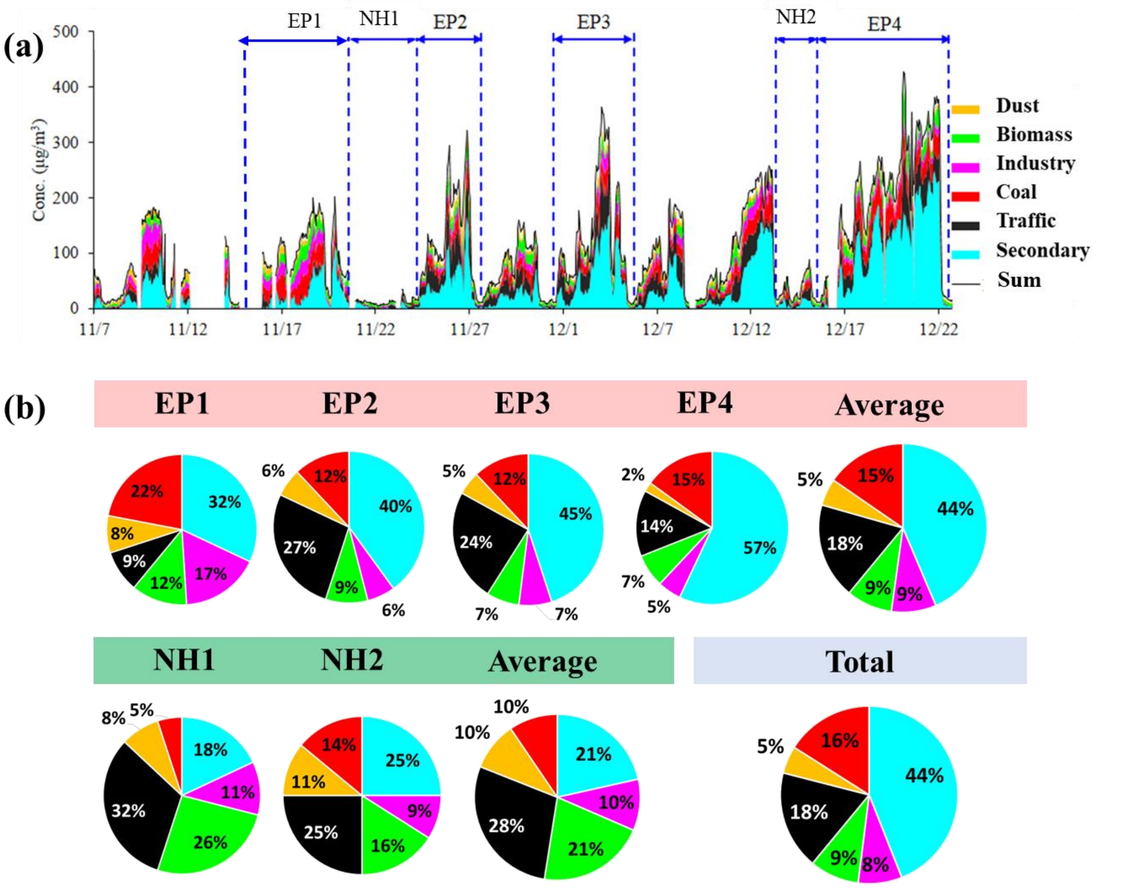


Figure 2. Variation of (a) chemical composition and (b) elemental species with $PM_{2.5}$ concentration (the white bars represent the frequency of $PM_{2.5}$ concentration).



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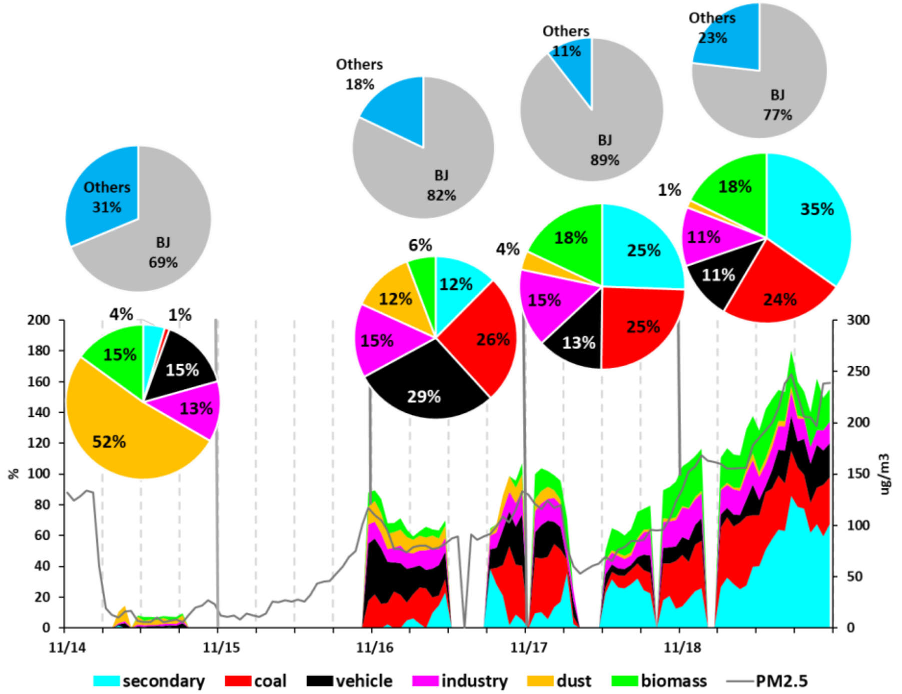
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BJ 69%
Others 18%
BJ 82%
Others 11%
BJ 89%
Others 23%
BJ 77%
4%
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52%
6%
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ug/m3
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140
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secondary
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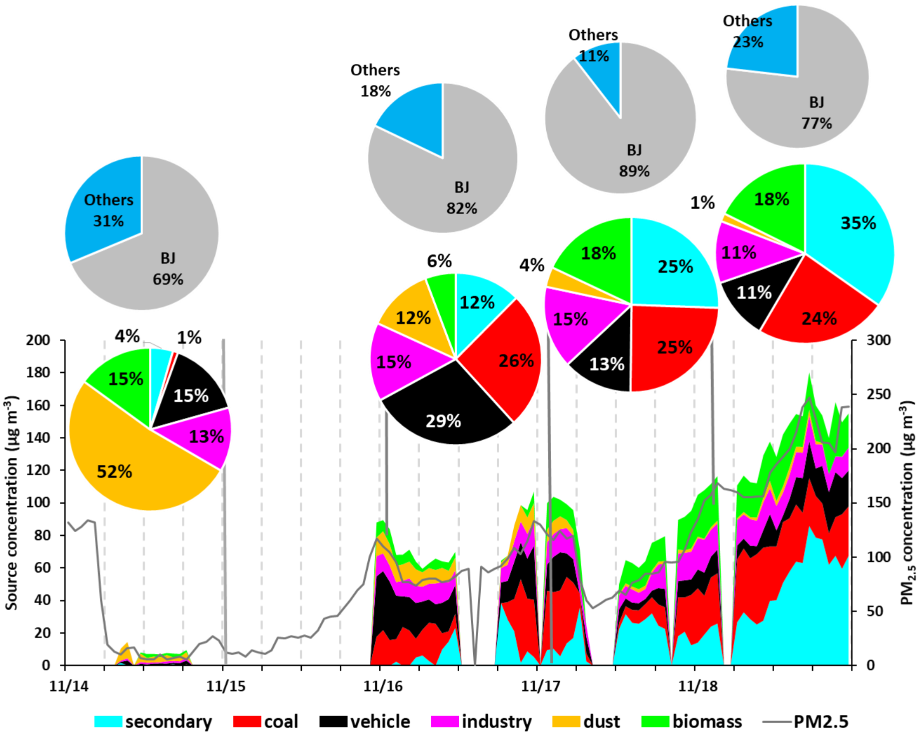
PM2.5


Others 31%
BJ 69%
Others 18%
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Source concentration (μg m⁻³)
PM2.5 concentration (μg m⁻³)
0
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80
100
120
140
160
180
200
0
50
100
150
200
250
300
11/14
11/15
11/16
11/17
11/18
secondary
coal
vehicle
industry
dust
biomass
PM2.5

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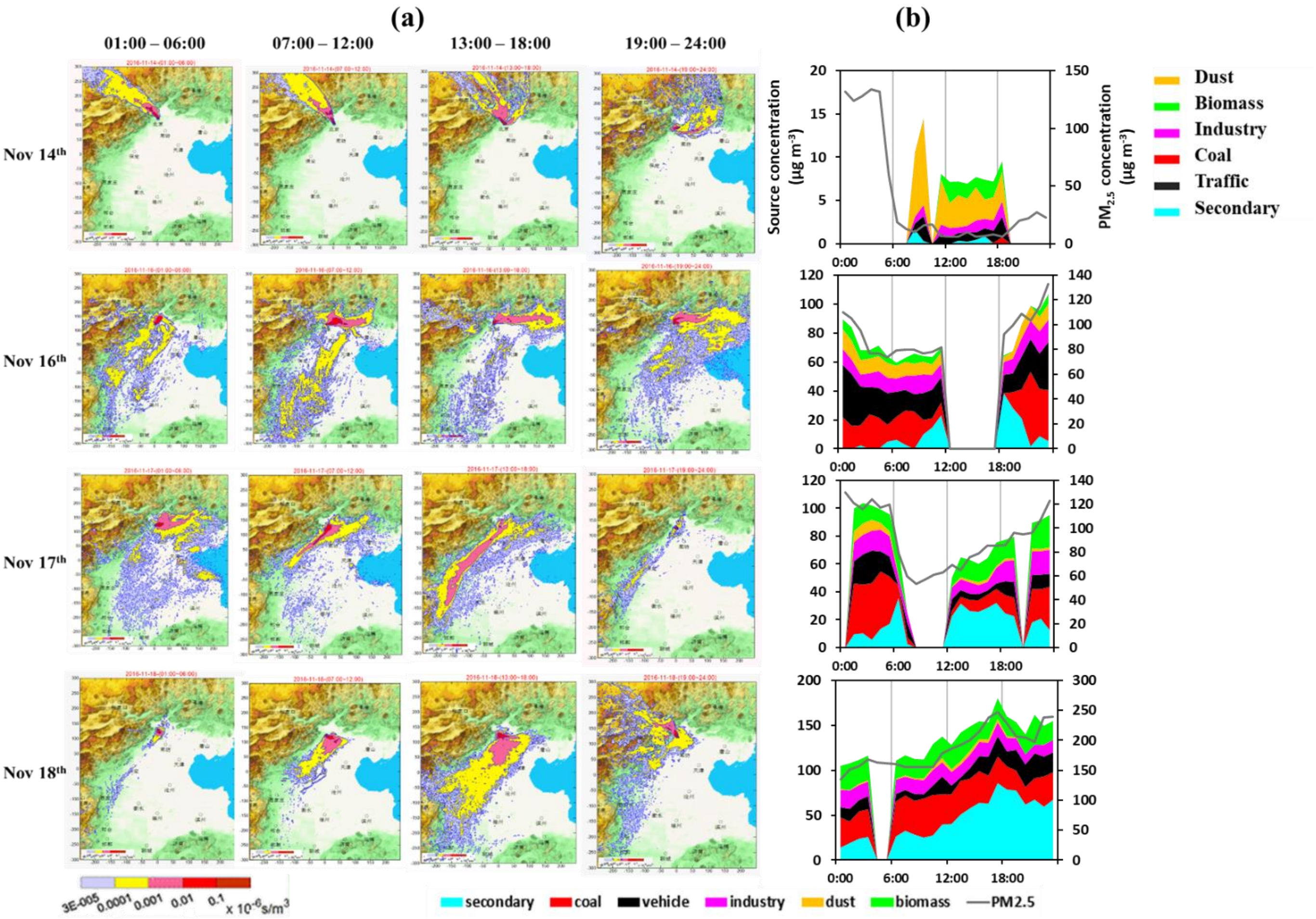


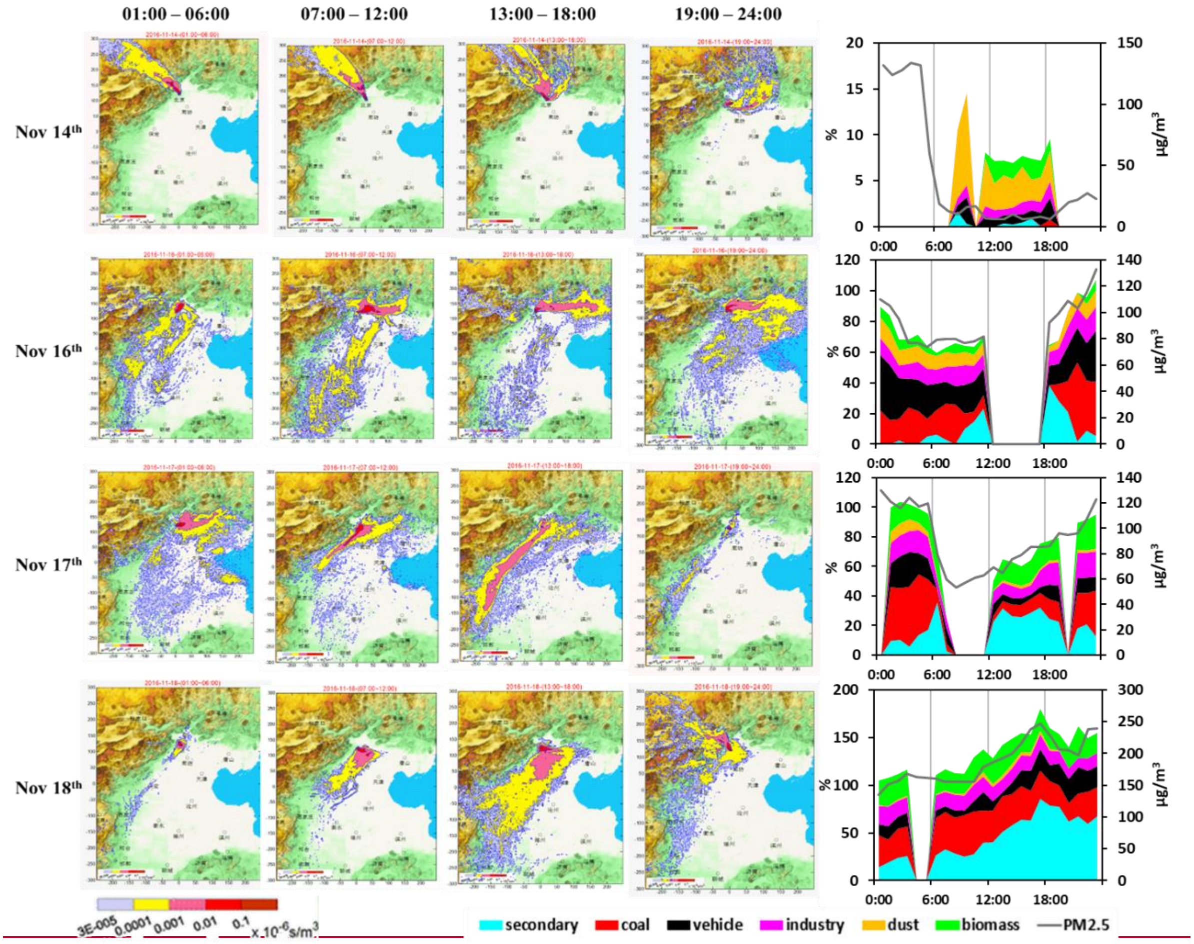
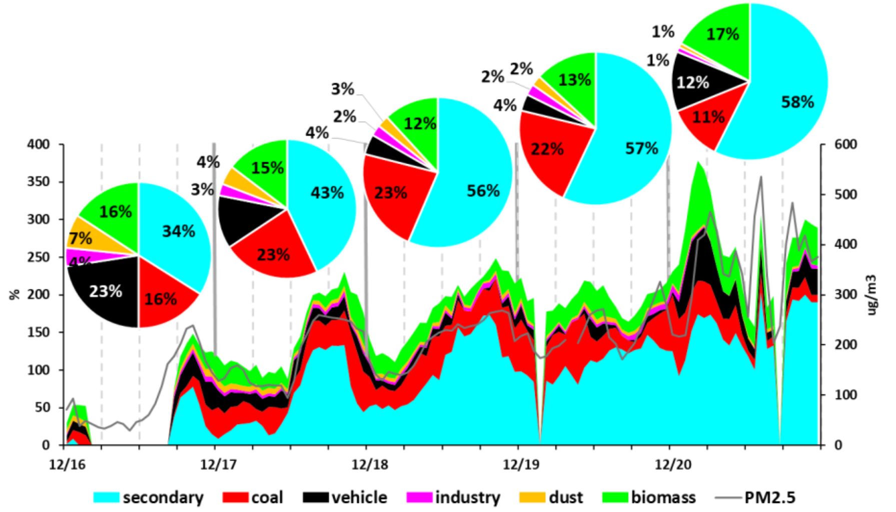


Figure 5. (a) Source regions by the footprint model and (b) daily source apportionment results by PMF in EP1.



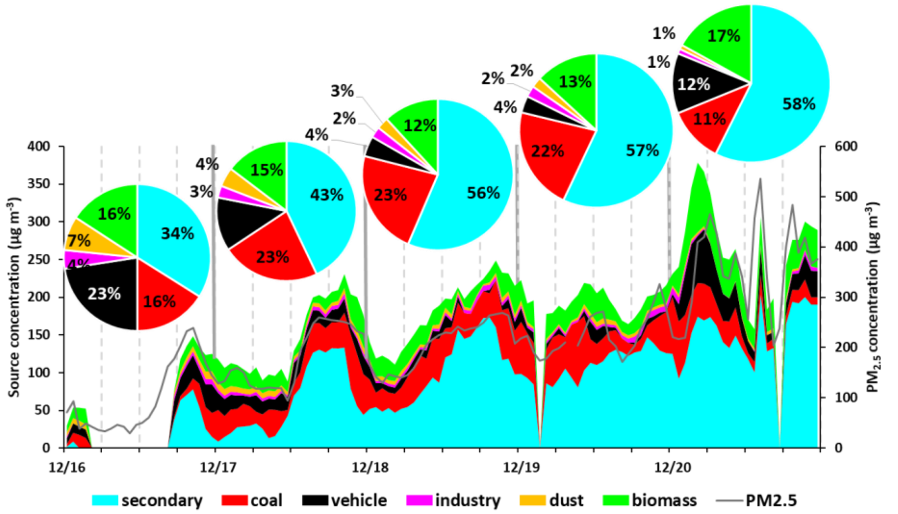
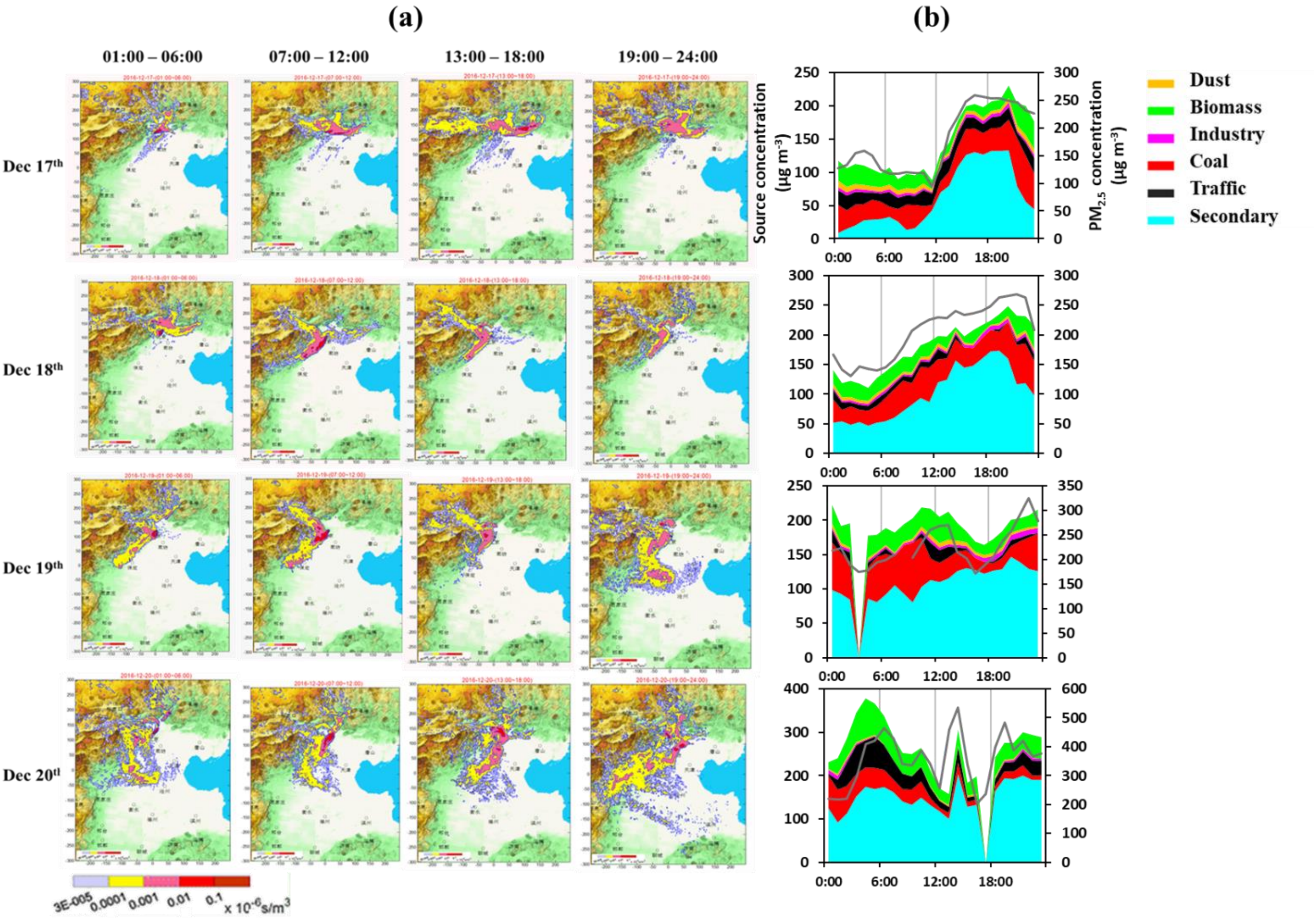


Figure 6. Source contribution in EP4. The pie charts show the daily source type and contribution.


(a)
(b)
01:00 – 06:00
07:00 – 12:00
13:00 – 18:00
19:00 – 24:00
Dec 17th
Dec 18th
Dec 19th
Dec 20th
3E-005
0.0001
0.001
0.01
0.1
x 10⁻⁶s/m³
Source concentration (μg m⁻³)
PM2.5 concentration (μg m⁻³)
0:00
6:00
12:00
18:00
Dust
Biomass
Industry
Coal
Traffic
Secondary

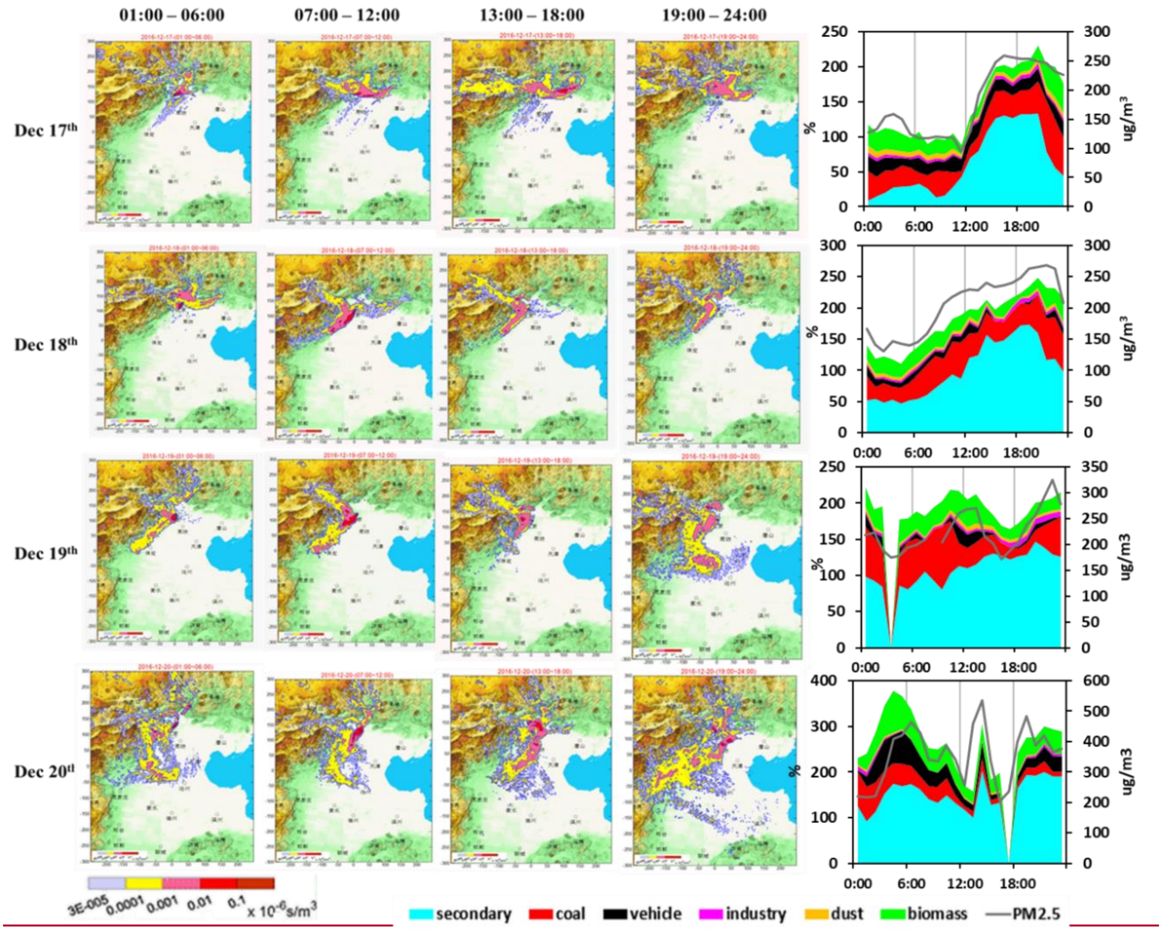
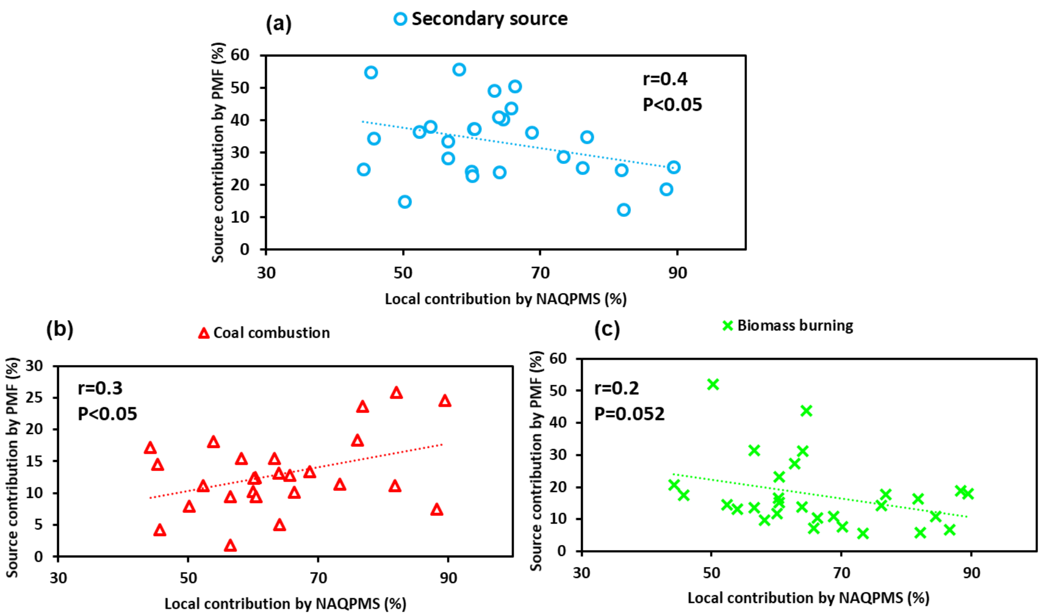
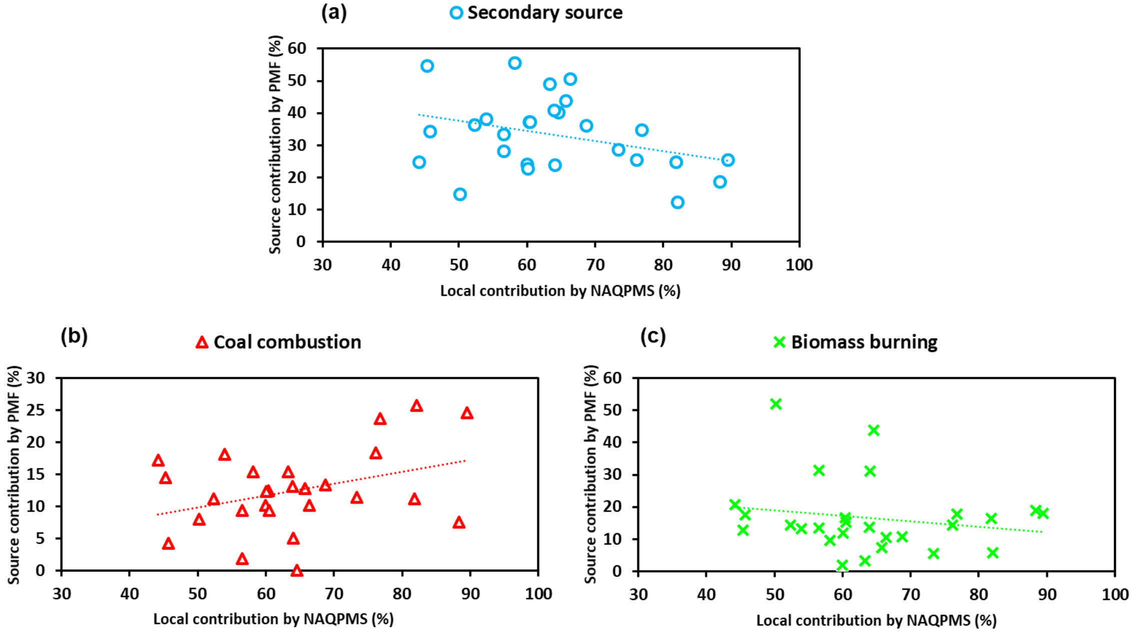


Figure 7. (a) Source regions by the footprint model ~~(every 6 h)~~ and (b) daily source apportionment results by PMF in EP4.





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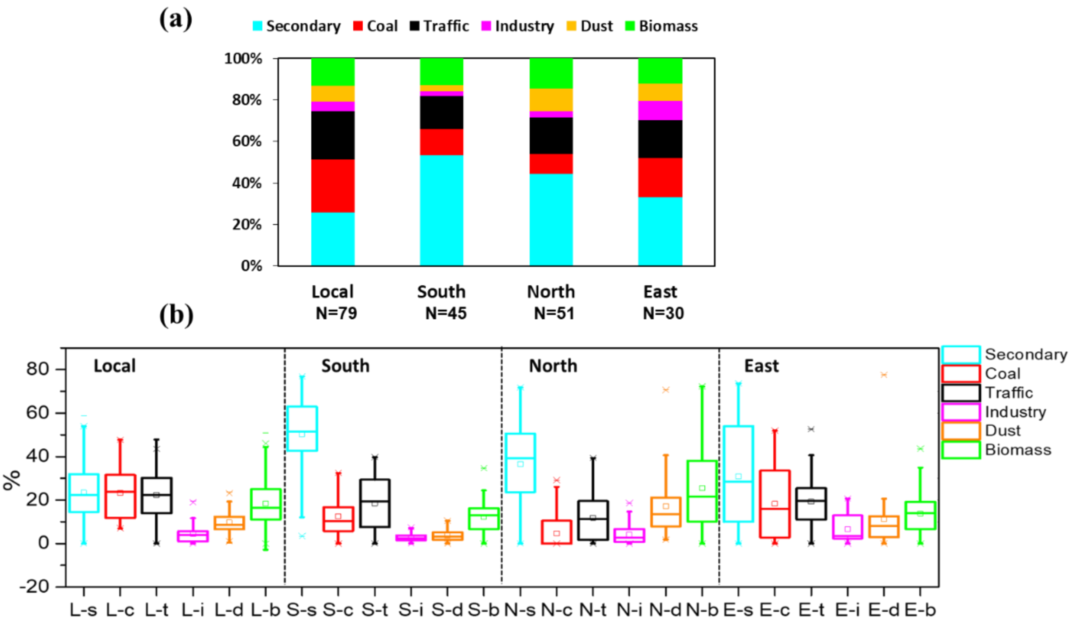

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