A point-by-point response to the reviews

Thank you for your valuable comments. The followings are our responses to your comments.

Response to Reviewer #1

Comment 1: This manuscript by Liu et al. reports $PM_{2.5}$ and its major components in winter and spring seasons at four sites in North China. Chemical compositions and spatial difference are discussed. The major sources are also attributed. Generally, the study is well-designed and the manuscript should be published after my concerns are addressed.

Answer: Thank you for your positive evaluation of our work. The followings are our responses to your comments.

Comment 2: Line 144-159. The authors claim that Cl/Na ratio is 1.4 in coal combustion. And if the ratio high than 1.4, atmospheric Cl and Na would be considered to be totally from coal combustion. In fact, biomass burning, including wild fires, open straw burning and biofuel combustion also emits Na and Cl with the Cl/Na ratios of 1-6 (e.g. Schauer ES&T 2001). Moreover, the OC/EC ratios in biomass burning samples are as high as those in coal combustion (Table 3). Since biofuel combustion for heating is also enhanced during winter in the northern China, why and how did the authors rule out the influence of biomass burning on $PM_{2.5}$ at the four sites?

Answer: Yes, you are right. Biomass burning is indeed an important source for atmospheric Na^+ and Cl^-, with the Cl^-/Na^+ ratios of 1-6 (Schauer et al., 2001). However, biomass burning in the NCP region is mainly focusing on the harvest seasons in summer and autumn (Zong et al., 2016), and few farmers are currently combusting crop straws for household cooking and heating because of the inconvenience of biomass with respect to coal and liquid gas.

The ratios of OC and EC to K^+ from biomass burning had been measured to be about 3.9 and 0.8, respectively (Li et al., 2007; Yao et al., 2016), which was about one order of magnitude less than those (34.0 ± 9.3 for OC/ K^+ and 6.9 ± 1.7 for EC/ K^+) measured in this study. Assuming the atmospheric K^+ measured in this study was totally from biomass burning, the contribution of biomass burning to atmospheric carbonaceous aerosols could be roughly estimated to be only 2.8%-5.2% in $PM_{2.5}$ based on the typical ratios of OC and EC to K^+ from biomass burning and the mass proportions of K^+ (0.6%-1.1%, Fig. 11). Therefore, biomass burning during the sampling period in this study made minor contribution to atmospheric $PM_{2.5}$. According to your pertinent comment, the corresponding paragraph has been rephrased in the revised manuscript.

Comment 3: The authors discuss the spatial difference of $PM_{2.5}$ and the major components at the four sites. Did the meteorological conditions, such as planetary boundary layer (PBL), cause such a spatial difference?

Answer: The meteorological conditions, especially wind speed and planetary boundary layer (PBL), play pivotal roles in the dispersion and accumulation of atmospheric pollutants, which can cause spatial and temporal difference of pollutants. As for the sampling sites of BD, WD and DBT, the meteorological conditions could be considered as the same because of the short distances (< 36 km) among them, and hence the spatial difference of $PM_{2.5}$ and the major components at the three sampling sites was rationally ascribed to the different source strengths. Although the distance between the sampling sites of BJ and BD is about 156 km, there was no significant difference of the wind speeds between the two sampling sites during the sampling period (1.4 ± 1.4 m/s for BJ and 1.7 ± 1.1 m/s for BD, Fig. 3). Therefore, the spatial difference of $PM_{2.5}$ and the major components between the sampling sites of BJ and the other three could not be ascribed to the difference of the wind speeds. Because the information of PBL was not available in the region of Baoding, it is difficult to discuss the impact of PBL on the spatial difference of the pollutants. According to your pertinent comment, the corresponding paragraph has been rephrased in the revised manuscript.

Comment 4: SOC is estimated by the EC-tracer OC/EC method. However, previous studies have demonstrated that this method could overestimate SOC under the influence of coal combustion and biomass burning, especially in wintertime. As discussed in the manuscript, coal combustion (maybe also biofuel combustion) had significant impact on PM at the sampling sites in wintertime. Thus, the concentrations and mass fractions of SOC in the winter (Figure 11) should be overestimated.

Answer: Yes, we totally agree with your comment. Because the lowest 10 % percentile of OC/EC ratios (3.5) measured during the sampling period were obviously less than that (13.1, Table 3) from residential coal combustion, POC could be underestimated by the product of the lowest OC/EC ratio and EC measured, and SOC could be overestimated through the subtraction of POC from OC. The overestimation of SOC by the EC-tracer OC/EC method has been noted by previous studies (Ding et al., 2012; Cui et al., 2015). The statement has been inserted in the revised manuscript.

Comment 5: The citation need to be re-formatted throughout the main text. For example, in line 48, the citation formats for the two references are different (Huang et al., 2014; P. Liu et al., 2016).

Answer: According to the citation style of the ACP, "Huang et al., 2014" is commonly the standard format. However, "P. Liu et al., 2016", "J. Liu et al., 2016" and "C. Liu et al., 2016" are often used to distinguish the references that first authors are different but both the last names of first authors and the publication years of the papers are the same.

Comment 6: Line 230-232. Is the difference in concentrations statistically significant? Please add p-value to show the significance of the observed difference.

Answer: Yes. The one-way ANOVA analysis results of the concentrations of OC, EC, NH_4^+, NO_3^-, SO_4^{2-}, Cl^- and K^+ at the four sampling sites are list in Table R1. The statistically significant differences among them were found with the p-values all lower than 0.01. The p-value ($p < 0.01$) has been added in our revised manuscript.

Table R1. The one-way ANOVA analysis for the concentrations of OC, EC, NH_4^+, NO_3^-, SO_4^{2-}, Cl^- and K^+ at the four sampling sites.

		Sum of Squares	df	Mean Square	F	Sig.
OC	Between Groups	56870.407	3	18956.802	19.096	.000
	Within Groups	76439.111	77	992.716		

	Total	133309.519	80			
EC	Between Groups	3036.393	3	1012.131	18.014	.000
	Within Groups	4326.303	77	56.186		
	Total	7362.696	80			
NH_4^+	Between Groups	2029.908	3	676.636	7.820	.000
	Within Groups	6662.556	77	86.527		
	Total	8692.465	80			
NO_3^-	Between Groups	1254.055	3	418.018	4.188	.008
	Within Groups	7685.732	77	99.815		
	Total	8939.787	80			
SO_4^{2-}	Between Groups	2003.050	3	667.683	4.205	.008
	Within Groups	12227.563	77	158.800		
	Total	14230.613	80			
Cl^-	Between Groups	934.896	3	311.632	14.889	.000
	Within Groups	1611.608	77	20.930		
	Total	2546.503	80			
K^+	Between Groups	73.109	3	24.370	19.524	.000
	Within Groups	96.109	77	1.248		
	Total	169.218	80			

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