Response to Reviewers' comments

We are thankful to the two referees and Dr. Lin for their thoughtful and constructive comments which help improve the manuscript substantially. Following the reviewers' suggestions, we have revised the manuscript accordingly. Listed below are our point-by-point response in blue to each comment that was offered by the reviewers.

Response to Dr. Chunshui Lin

1. **General comments**

The study by Lei et al., entitled "Fine particle characterization in a coastal city in China: composition, sources, and impacts of industrial emissions", characterizes the aerosol composition and sources near two large steel plants in a coastal city in Shandong using a $PM_{2.5}$ TOF-ACSM. While it is generally well presented, I have a few concerns about the data interpretation which is sometimes questionable. Specifically, in the OA Source Apportionment section, HOA was apportioned to traffic which was shown to have a pronounced morning and evening rush hour peaks. However, HOA was also shown to have a good correlation with NO_x (R^2 =0.82-0.87). The tight correlation between HOA and NO_x means NO_x also had such diurnal trends and could be exclusively from traffic. However, contrasting results appeared in the Industrial Plumes Section where it shows NO_x was also correlated with CO (R^2 =0.83). Later, Lei et al., stated that CO, NO_x, and SO_2 were co-emitted from steel plants. So my question, how is the correlation between CO and HOA, and what on earth NO_x is from. If NO_x is from traffic then other correlated species (i.e., CO and SO_2) are also from traffic, which means the NO_x/SO_2 ratios or NO_x/CO ratios would be invalid diagnostic ratios for steel plant emissions. If NO_x was indeed from steel plant, then the source apportionment of OA factor is questionable. Because the tight correlation between HOA and NO_x, HOA would also be from steel plant, however, which shows morning and evening rush hour peaks. This would be more questionable.

We thank Dr. Lin's comments on our manuscript. First, the correlation coefficient r^2 between HOA and NO_x was incorrectly written, and it was corrected into "r^2 =0.52-0.65" in the revised manuscript. CO and HOA were poorly correlated (r^2=0.04-0.05) and showed quite different diurnal variations (Fig. R1), suggesting their different emission sources. On the contrary, the tight correlation between HOA and NO_x (r^2 =0.52-0.65) and the similarity in their diurnal patterns suggested their common sources. As proved by previous studies, NO_x is produced from the reaction of nitrogen and oxygen gases in the air during combustion, especially at high temperatures. Transportation, industrial activities, power plants and domestic combustion could contribute significantly to NO_x in the atmosphere (Streets and Waldhoff, 2000). The characteristics of diurnal patterns of NO_x in our study indicated that traffic emissions were the main source of NO_x near the sampling site during plume excluded period. As shown in Fig. 8 in the manuscript, the tight correlation between NO_x and CO, NO_x and SO_2 were only found during periods with clear influences from steel plant plumes, suggesting that a large fraction of the three gaseous species were co-emitted from steel plants during plume periods, while they were affected by multiple and different sources during non-plume periods. And due to the high instant concentrations in the plumes, other sources like transportation or domestic coal combustion play minor roles during plume periods, so we concluded reasonably that the ratio of NO_x/CO and NO_x/SO_2 can be used as indicators for steel plant emissions.

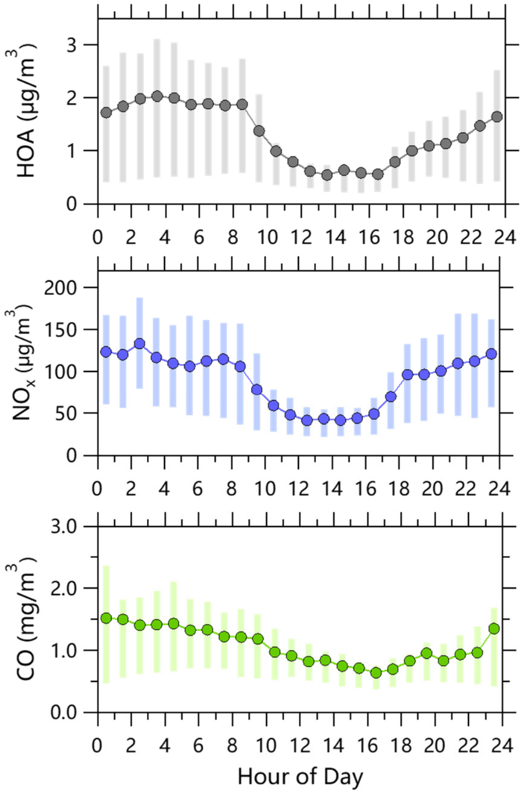

Figure R1. Average diurnal variations of mass concentrations of HOA, NO_x and CO in March 2019, the plume periods have been excluded. The error bars indicate 25th and 75th percentiles.

2. Specific comments

1. My other major concern is how to prove that SO_4 are exclusively from power plant, but not regionally transported. The good correlation between LO/MO-OOA and SO_4/NO_3 indicates regional transport. And the increase of SO_4 above background level before and after the plume could be stronger secondary production during transport. The wind rose only shows a huge source from the southwest region which could be other sources (e.g., general industries

activities) other than power plant. You need more evidence to show the sulfate/ammonium were exclusively from the power plants.

It is a good point. If sulfate was dominantly from regional transport, all aerosol species would be expected to vary similarly. However, during periods with significant influences of steel plants, we only observed substantial increases in ammonium and sulfate, while the mass concentrations of organics and NO_3 remained small changes. Therefore, the sharp increases in mass concentrations of SO_4 and NH_4 were unlikely from the regional transport. The bivariate polar plots also showed that the periods with large increases of sulfate were dominantly from the southwest where the industrial region is located. Based on the available measurements in this study, we cannot quantify the exact contributions of each industry to the increase of sulfate. Considering the dominant industrial emissions in this region, sulfate and ammonium were expected dominantly from the emissions of steel plants.

The correlation coefficients between SO_4 and MO-OOA in Fig. 4 were performed by excluding the periods with significant industrial influences, which is now clarified in figure caption as "Note that the periods with influences from steel plants were excluded when performing correlation analysis between MO-OOA and SO_4 in (a) and (b)." It can be seen that sulfate was generally well correlated with MO-OOA, consistent with the fact that the two components of MO-OOA and SO_4 were mainly from regional transport during periods with small influences of industrial plumes. As shown in Fig. 4, we didn't observe simultaneously large increase in MO-OOA as the plume-related sulfate (Fig. 2), further supporting that the large increases in sulfate could not be from strong secondary production during transport.

2. Are these two power plants coal-powered? Could you comment why CCOA was only resolved in March but not in September. Does it mean the CCOA was actually not from the power plants but from residential sectors? What is the correlation between CCOA and NO_x/CO/SO_2 during the industrial plume? What are the mass spectra during industrial plume, any sign of CCOA? If no sign of CCOA, does it mean the SO4 was not from power plants?

Yes, they were coal-powered steel plants. According to previous studies, the emission factors of carbonaceous aerosol from industrial boilers were significantly lower than those from residential coal combustion by a factor of more than 2 (Zhang et al., 2008). Therefore, CCOA was not resolved in September when residential heating was not important. This is also consistent with many observations in China, e.g., Beijing. For example, CCOA was only resolved in heating season when residential coal combustion was important. However, it cannot be identified in other seasons even the industrial emissions are one of the important aerosol sources (Sun et al., 2016;Sun et al., 2018;Xu et al., 2019). In comparison, CCOA was resolved in March because of the enhanced coal combustion emissions for domestic heating in this season. Thus, we concluded that CCOA was mainly from residential coal combustion instead of steel plants.

Figure R2 shows the correlation of CCOA with $NO_x/CO/SO_2$ during the periods with clear industrial plumes. It can be seen that CCOA was almost completely irrelevant with $NO_x/CO/SO_2$ (r^2=0.02-0.10), suggesting their different emissions during industrial plume periods. Considering that a large fraction of NO_x, CO and SO_2 were emitted from the steel plants during plume episodes, the poor correlation between CCOA and $NO_x/CO/SO_2$ further supported that CCOA could not be dominantly from steel plants.

Figure R3 shows the average OA spectra during the periods with the influences of industrial plume in March. No pronounced PAHs signals were observed in the spectra suggesting that the industrial activities appear not emit CCOA significantly. Even there was no sign of CCOA, it does not mean that sulfate was not from steel plants as well. For example, carbonaceous aerosols can be burned out completely to CO and CO_2 during the high-temperature processes of steel making, while the emissions of SO_2 and SO_3 are still significant which can form ammonium sulfate rapidly in the atmosphere.

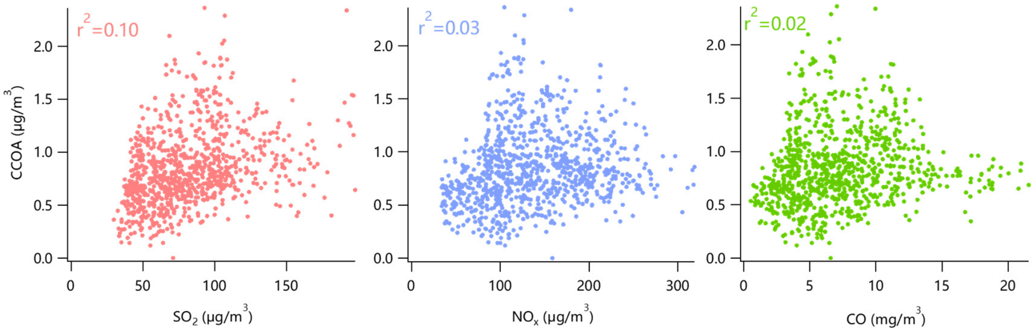

Figure R2. Scatter plots of CCOA with (a) SO_2, (b) NO_x and (c) CO during periods with significant influences from steel plant plumes in March 2019.

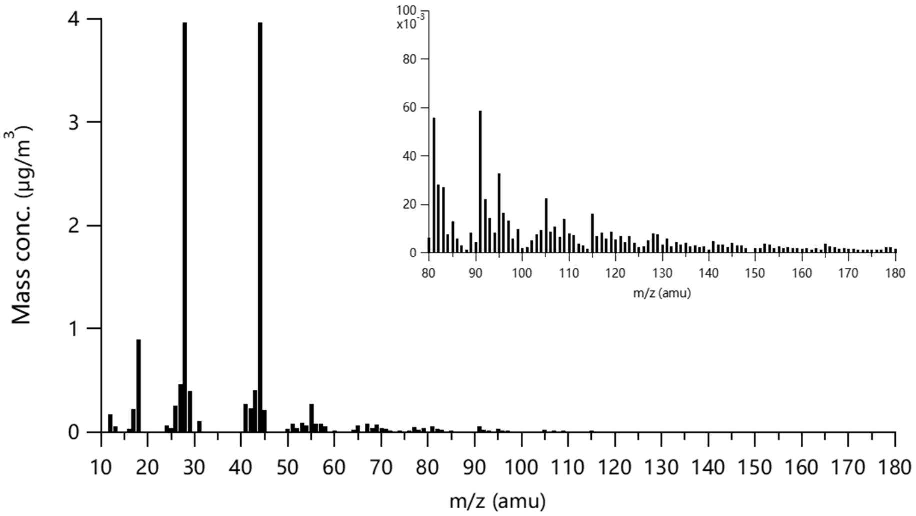

Figure R3. Average OA mass spectra during periods with significant influences from steel plant plumes in March 2019.

3. What are the tools to perform the PMF analysis? How the factors are systematically evaluated? In the Method section, it states "Considering the limited sensitivity of the ToF-ACSM, m/z's larger than 120 and 180 were excluded in PMF analysis" However, in the PMF Section, and Fig. 4, it states "Compared with HOA, CCOA presented a higher mass fraction of larger m/z's (>120) indicating that coal combustion can be an important source of high molecular-weight organic matter during heating period." Is m/z's >120 really excluded?

   The PMF analysis on the organic mass spectra from ToF-ACSM were performed by using the Igor PMF evaluation tool (PET, version 3.04A) (Paatero and Tapper, 2010;Ulbrich et al., 2009), following the data-processing and factors-selecting steps given by Zhang et al. (2011). Factor numbers from 1 to 10 were run in the model with different rotational parameter (f_{peak}) values (i.e., -0.6 to 0.6, stepped by 0.2), and a 3-factor solution with f_{peak}=0 and a 4-factor solution with f_{peak}=0 were determined in September 2018 and March 2019, respectively. More factor solutions would lead to clear splitting of the factors and thus are not selected. We performed PMF analysis on the organic mass spectra in September 2018 and March 2019, and we excluded *m/z's* >120 in September 2018 and *m/z's* > 180 in March 2019, respectively, due to (1) their low signal-to-noise ratios and low contributions to the total OA mass, and (2) low contribution of CCOA in September. Such explanations have been clarified in the manuscript "Considering the limited sensitivity of the ToF-ACSM, *m/z's* larger than 120 and 180 were excluded in PMF analysis in September 2018 and March 2019, respectively due to their low signal-to-noise ratios, and low contributions to the total OA mass."

Response to Reviewer #1

Lei et al. presented time-of-flight aerosol chemical speciation monitor (ToF-ACSM) measurements of fine particles in a coastal city in China that is affected by two large steel plants. The instrument was equipped with the newly developed $PM_{2.5}$ size cut and capture vaporizer. From data sets in spring and fall, differences in mass concentration and species concentration were discussed with both diurnal pattern analysis and positive matrix factorization (PMF). Potential source analysis using bivariate polar plots revealed impacts from the steel plants. Specifically, six plumes were analyzed in detail, and results suggest that ammonium sulfate, NO_x/CO ratio, and NO_x/SO_2 ratio could be used to evaluate the impacts from steel plant emissions. The measurements are new in that 1) a focus on the effects of industrial plumes; and 2) with newly developed $PM_{2.5}$ size cut and capture vaporizer. The analysis is rigorous for standard aerosol mass spectrometric (AMS) practice and PMF analysis, but lacks in verification of certain potential differences between the capture vaporizer (CV) and standard vaporizer (SV), as indicated in Major Comment #1 below. The manuscript is fairly well written, but has some room for improvement. Minor Comments below list a few examples for the authors to consider changing. Overall, I have concerns in the assertions that certain inorganic and gaseous species can be used as diagnostics for steel plant emissions (see Major Comments below). Therefore, I suggest Major Revision, for the authors to lean the discussion more on other major conclusions (which are by themselves quite useful), instead of focusing too much on using these diagnostic indicators that might not be specific to steel plant emissions.

We thank the reviewer’s positive comments.

Major Comments:

1. P11, L14. The authors suggested that there were two plumes in which ammonium bisulfate accounted for 70-80% of PM mass. I suppose the conclusion that the “sulfate” detected by the ToF-ACSM was bisulfate is from the observation that “sulfate” measured was much higher than the ammonium measured on an equivalent basis. If so, the authors should specify that in the manuscript. Another concern I have is whether that was partly due to the usage of CV as compared to the commonly found nearly “neutral” NR-PM in previous AMS studies using SV. There might be difference in both RIE and fragmentation patterns for CV compared to SV, as indicate by Hu et al. (AMT, 2017 and AST, 2017). The authors need to clarify whether these would affect quantification of sulfate using the ToF-ACSM that comes with a CV.

We thank the reviewer’s comments. It was a mistake, and the correct conclusion should be that ammonium bisulfate accounted for ~20%-40% of total PM mass during steel plant plumes. In consideration of the negligible contribution of NO_3 and Chl in the plumes, which was demonstrated by the pie charts in Fig. 7, it is reasonable to estimate the relative fraction of ammonium sulfate and ammonium bisulfate based on the NH_4_meas/NH_4_pred ratios, which was discussed in detail in Zhang et al. (2007). The plots of NH_4 balance for different periods are presented in Fig. R4. As for the quantitation of sulfate by CV ToF-ACSM, it has been validated in both laboratory studies and field campaigns that, even the fragmentation patterns and RIE may be different, the sulfate mass concentrations measured by CV and SV instruments agreed well with each other, as long as the RIE could be calibrated correctly (Hu et al., 2016;Hu et al., 2017). In addition, we calibrated RIEs of ammonium and sulfate before and after the two campaigns, and found that the RIE of sulfate was fairly robust which was 1.1, while that of ammonium changed from 3.1 in September 2018 to 3.8 in March 2019.

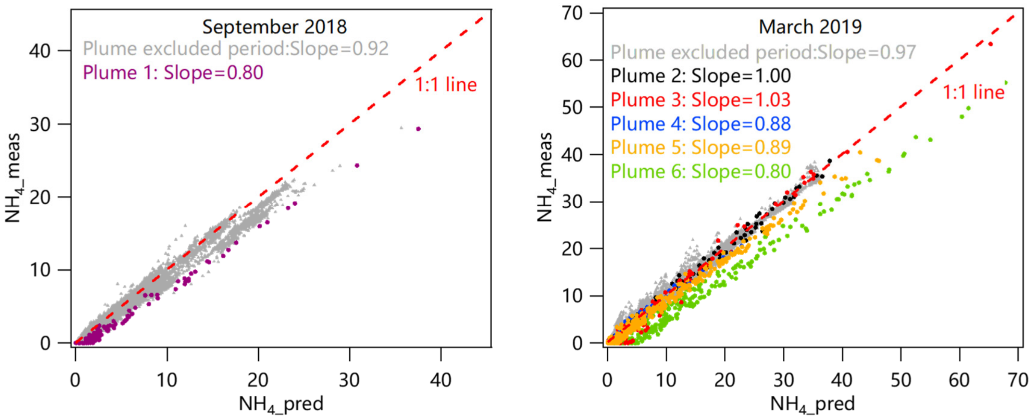

Figure R4. Scattering plots of measured NH_4 vs. predicted NH_4 in steel plant plumes (after subtracting background concentrations, NH_4_pred=18*(2*SO_4/96+NO_3/62+Chl/35.5)).

2. P12, L5-8. The authors suggested that the NO_x/SO_2 and NO_x/CO ratios can be used as diagnostics for impacts of steel plant emissions. First, it is ok for NO_x/SO_2 ratio with both ug/m^3 as the mass concentration unit, and end up in a dimensionless ratio. But CO has a unit of mg/m^3. Should the NO_x/CO ratio be the number stated * 10ˆ-3? Second, the low ratios observed in plumes in this study do not exclude possibilities of similar low ratios from other plumes (such as traffic emissions, power plants, and biomass burning). Can the authors summarize literature values for other typical primary sources before making such a statement that low ratios of these gaseous species can be indicators of steel plant emissions?

   We thank the reviewer's comments. We have changed the NO_x/CO ratios into the correct order of magnitude accordingly. We referred to relevant articles and summarized the literature values of NO_x/CO and NO_x/SO_2 ratios in Table R1. The NO_x/CO ratios in different emission types including vehicle, biomass burning, urban site, rural background site, etc., were investigated relatively more extensively. As presented in Table S1, the NO_x/CO values obtained from other emission sources were apparently higher than that in the steel plant plumes in this study (Schürmann et al., 2007;Fujita et al., 2012;Tiwari et al., 2015;Santos et al., 2018). It’s worth noting that the values of NO_x/CO for on-road motor vehicle emissions (Fujita et al., 2012) were quite close to the values during non-plume period in our study, which supported that our results are reliable. The NO_x/SO_2 value of 1.04 obtained from the fresh flue-gas plume from a coal-fired power plant when FGD and fabric filter were used (Mylläri et al., 2016), which was similar to the flue-gas cleaning conditions of RSP, is close to 1.24 in this study. Unfortunately, the investigations on NO_x/SO_2 were relatively fewer and there are not enough comparable literature values. However, it has been proved by Parrish et al. (1991) that lower NO_x/SO_2 is characteristic of the emissions from coal burning, e.g., in power generation plants or steel making plants, while higher NO_x/SO_2 is generally attributed to vehicle emissions, which is consistent with our conclusions in this study. It needs to be clarified that we treated the steel plant (RSP or SSP) as a whole in this study, rather than focusing on detailed or specific production process including coking, sintering, pickling, etc., or the small-scale coal-fired power plant for the power supply of steel production. Another important thing to note is that plumes emitted by steel plants and power plants are supposed to have common characteristics, such as close values of NO_x/SO_2 as mentioned above, that is because the main source of pollutants emitted from the two kinds of industrial plant could be attributed largely to coal combustions. However, take the fact that there is no any other power plant around our sampling site into consideration, we can conclude reasonably that the lower ratio of NO_x/SO_2, supported also by lower values of NO_x/CO and sharp increase in mass concentration of sulfate and ammonium, can be used as indicators of steel plant emissions. We have also clarified this important conclusion in more detail in the revised manuscript.

Table R1. Summary of literature values of NO_x/CO and NO_x/SO_2.

	Emission type	**NO_x/CO**	**NO_x/SO_2**
Schurmann et al., AE, 2007.	Airport	0.20-0.25	-
Tiwari et al., AR, 2015.	Urban background site	15.0	-
Fujita et al. J. Air Waste Manage. Asso, 2012.	On-road motor vehicle	0.04-0.05	-
Santos et al., ACP, 2018.	Biomass burning	-	-
	Forest background	0.33-1.48	-
	Fresh plume	0.056-8.33	-
	Anged plume	0.1-6.33	-
Mylläri et al., ACP, 2016.	Coal-fired power plant	-	-
	Fabric filter on	-	1.04
	Fabric filter off	-	4.69
This study	Steel plant		
	Non-plume period	0.04	1.55
	Plume period	***0.01***	***1.24***

3. P13, L4. The statement that ammonium sulfate can be used to evaluate and quantify the impacts of steel plant emissions is even less convincing. There would be ubiquitously existence of ammonium sulfate in most environments, many of which with ammonium sulfate as the dominating NR-PM species. If the authors can justify Major Comment #1 above, then it makes a little bit more sense to say that ammonium bisulfate (typo?) can be used for this purpose. Still, other sources (e.g., power plants) can emit (directly or indirectly) large amounts of ammonium bisulfate too. Please clarify.

   We thank the reviewer's comments. First, the quantification of sulfate by CV-AMS has been proved to be highly consistent with SV-AMS. Second, it's true that sulfate and ammonium are ubiquitous in the atmosphere and may be the dominating NR-PM species, and that's why we deducted the background concentrations, which were determined as the average of 1 hr data before and after steel plant plumes to remove the background interference. Besides, the increases in sulfate and ammonium mass concentration are sharp and significant relative to non-plume periods. It's also true that other sources like power plants can emit large amounts of sulfate and ammonium as we stated in the reply of *Major comment 3*, but there is no such possible source emissions near our sampling site, and such rapid and significant increase in mass concentration of sulfate and ammonium is unlikely from secondary formation during the transport.

We totally agree with the reviewer that it was not convincing enough to use ammonium sulfate as a tracer for the impacts of steel plant emissions considering the ubiquitous existence of ammonium sulfate in most environments. In fact, ammonium sulfate can be used as tracer for industrial impacts only in regions nearby, while it would have a large uncertainty in regions far from the steel plants because of the rapid dilution during the transport. In the revised manuscript, we toned town the implications of our results as "Our results might have significant implications for better quantification of industrial emissions using ammonium sulfate and the ratios of gaseous species as tracers in industrial regions and nearby in the future."

Minor Comments:

1. P1, L13: a new sentence should start after "China".

   Corrected.

2. P1, L14: change "have" to "present".

   Corrected.

3. P1, L16: no need to use "seasons". Spring and fall contain the meaning of "season". There are a number of similar cases later. Please check.

   More appropriate expressions were used in the revised manuscript.

4. P1, L21: put a "," after "seasons".

   Corrected.

5. P2, L12: change "important one" to "important sources".

   Corrected.

6. P2, L16: add "concentrations of" before "toxic".

   Corrected.

7. P3, L1: change "contributions" to "effects".

   Revised.

8. P3, L22: add "a" before "capture vaporizer".

   Corrected.

9. P4, L9: change "new" to "newly". There are some similar cases later too. Please check.

   We replaced "new" with "newly" in our revised manuscript.

10. P4, L10: delete “those,”?

    Revised.

11. P4, L15: I would suggest either using “SO4” as the shorthand notation or using the right ionic formula.

    Thank the reviewer’s suggestion.

12. P5, L9: delete “for”.

    Modified.

13. P7, L2: delete “was” before “decreased”.

    Corrected.

14. P7, L5: change “small, less than 2%” to “small at less than 2%”?

    Modified.

15. P8, L4: I would suggest using the right ion formula for the alkyl ion series.

    Modified.

16. P8, L17: a new sentence should start before “for example”.

    Modified.

17. P11, L8: change “nearby” to “near”.

    Modified.

18. P12, L17: add “a” before “higher contribution”.

    Corrected.

19. P12, L18: which one is higher? Daytime or nighttime? It seems contradictory to what was said previously.

    Corrected. It should be “Most aerosol species showed similar diurnal variations in the two seasons with higher concentrations at nighttime and lower values during daytime,”.

20. P13, L4: change “steel plants emissions” to “steel plant emissions”

    Modified.

Response to Reviewer #2

Lei et al. presented a unique dataset on submicron aerosol composition in an area heavily influenced by industrial emissions during two seasons. They conducted PMF analysis for source apportionments using a $PM_{2.5}$-ACSM. They also analyzed several plume events and concluded that the low NO_x/CO, NO_x/SO_2 and the dominance of ammonium sulfate and/or ammonium bisulfate in PM may be used as tracers for identification of emissions from steel plants. This paper is within the scope of ACP. However, there are several areas of ambiguity that should be addressed/clarified in the revision.

Major comments:

1. How did the authors exclude the influences from industrial activities from coal-fired power station, coking state, and pickling process that are also within the region?

   Thank the reviewer's comments. Our study treated the various steelmaking processes as a whole to evaluate the effects of steel plant emissions on air quality instead of evaluating a specific production process separately and precisely. With the available measurements in this study, it is very challenging to separate and quantify the contributions from different plants and processes. Considering that the steel plants, especially RSP, is the largest industry in this area, the impacts of industrial emissions we observed are expected be dominantly from the steel plants as well.

2. Figure 7: Based on the wind rose plots, all six plumes were associated with southerly or southwesterly winds and therefore were not likely to be from the Shandong power plant. How do the authors determine the NO_x/CO, NO_x/SO_2 and ammonium sulfate concentration from the SSP?

   The steelworks plumes from RSP were characterized by dramatic increases in sulfate and ammonium mass concentrations, while maybe due to more advanced purification and emission control technologies, such distinct characteristic was not observed for SSP during northeast wind periods. In order to figure out the impacts of SSP emissions on the air quality nearby, we furtherly explored the gaseous pollutants including NO_x, CO and SO_2 from different directions, and we found some common characteristics during the southwest and northeast wind periods. Specifically, although the SSP emits much less ammonium sulfate particles, and gaseous CO and NO_x, the ratios of NO_x/SO_2 and NO_x/CO were similar between these two steel plants. Thus, we suggested that we can use the combination of sharp increase of sulfate and ammonium, NO_x/CO and NO_x/SO_2 ratios to capture and quantify the effects of steel plant emissions more precisely. The wind rose plots for plume periods and plume excluded periods are shown in Fig. R5 to make a clearer explanation.

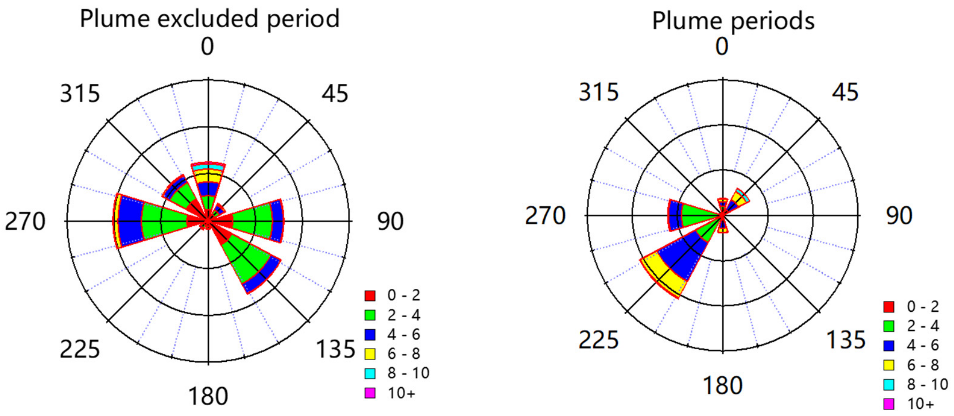

Figure R5. Wind rose plots for plume excluded period and plume periods in March 2019.

3. The authors need to confirm that buildings and street canyons do not affect their wind speeds and direction measurements.

   Thank the reviewer's comments. There are no dense and tall buildings near our sampling site that can affect the wind fields substantially. The west of the sampling site is a large wasteland, and the east is the Yellow Sea, and the north and south of the site are the buildings of steel plants.

Minor comments:

1. Page 2, Line 13: grammatical error in "one of the most important one"

   We revised the expression as "one of the most important sources".

2. Page 4: Line 19: It is usually called electron ionization these days. The electrons don't actually hit the molecules, so "impact" is disfavored.

   Thank the reviewer's comments. The non-refractory constitutes in the particles would be flash vaporized by the vaporizer and subsequently ionized by impacting with electrons emitted by filament (Fröhlich et al., 2013), so "electron impact" is actually a suitable expression in studies with aerosol mass spectrometer.

3. Page 5: Line 13: What about metals?

   The concentrations of metals were not measured in our study.

4. Page 4: Line 7: "while they were dominantly from the north in September and from the west in March" at night?

Yes, it's the description of prevailing winds at night, we have clarified this in the revised manuscript.

5. Figure 1: As a key element of a map, the scale should always be included.

   The scale was added on the maps in the revised manuscript.

6. Figure 2: The units for SO_2 and NO_2 are missing.

   Corrected.

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