Responses to Reviewer's Comments

The manuscript "Extreme haze pollution in Beijing during January 2013: chemical characteristics, formation mechanism and role of fog processing" by K. Huang et al. analyzed a severe pollution episode in Beijing during January 2013, based on Chinese API/AQI data, AERONET data and 15 PM2.5 samples. The monthly averages of pollutants are relatively high compared with those in previous years, which is mainly attributed to the abnormal meteorological conditions. Major aerosol species are found to have an exponential relationship with relative humidity. And aerosol in fog days is found to be more acidic. The topic is interesting. However, there are several major problems:

We gratefully thank for the reviewer's in-depth comments and suggestions. In the revised manuscript, we have added additional data to reinforce the data analysis. A majority part of the manuscript has been re-written and re-organized. All the changes have been highlighted in red color and we hope the reviewer could re-evaluate the revised manuscript. Followings are the response to all the comments point by point:

1. There have been already a number of papers discussing about the haze in the North China Plain. Compared with the previous works, I see few new findings or insights in this manuscript.

Thanks for the comments. In the revised manuscript, we have made more efforts on data analysis and sorted out more findings that are different from most previous studies. Some added findings in this revised manuscript are summarized as below:

1. Most literatures on this severe haze focused on the abundant chemical species of aerosol, i.e. organics, sulfate, nitrate and ammonium. In this study, we added the analysis of trace metals (including both mineral and pollution elements) to supplement the process analysis on explaining the evolution of aerosol levels during the study period. The advantage of using trace metals is due to their primary emission sources rather than the complex secondary formation pathway. Thus, by using different trace metals as chemical tracers, it is easier to apportion the main emissions sources at different stages of the pollution episodes. Please refer to Section 3.3.2 for more details.
2. Hourly measurements made by ACSM (Aerosol Chemical Speciation Monitor) were added to explore the evolution of secondary aerosol formation. Specifically, the partitioning of acidic

aerosol in the salt phase and free acid phase was discussed under different humidity conditions. Please refer to Section 3.4 for more details.

3. We use wind rose to identify three typical atmospheric processing conditions, i.e. cold front, local processing and regional transport. It is assumed that the chemical tracer ratio, i.e. $([X]/Al)_{cold}$ and $([X]/Al)_{local}$ could be used to represent aerosol chemical characteristics during cold fronts and local processing. By using a new developed algorithm, we have estimated the contribution from regional transport to some selected species. Please refer to Section 3.6 for more details.

2. The paper aims to investigate the characteristics and formation mechanism of the severe haze pollution event. Actually, without measurements of basic aerosol physical properties (e.g. number size distribution), I think the current data set is insufficient for such a purpose.

Thanks for the comments. Due to limitations of funding and instruments, we were incapable of conducting the measurement of aerosol physical properties in this field campaign. We hope the reviewer could understand our difficulty. In the revised manuscript, we added high resolution data measured by ACSM, which is the state-of-art aerosol characterization instrument in the world currently. Thus, in this study, we focused on the aerosol chemical properties to investigate the formation mechanism of the severe haze. In the future, we hope to collaborate with scientists whose expertise is aerosol physical properties on advancing the understanding of the haze formation of China in both the physical and chemical aspect.

3. The information on the QC of the data set is missing in the manuscript. The quality of some data is doubtable. For example, in fig. 4(a), the high-frequency fluctuation in visibility and RH before 10th of January seems to be weird. Something must be wrong with the measurements. I do not think the data in this period can be used. I do not know if such problem also existed in other periods which were used for calculating the average.

Thanks for the comments. We have double checked the raw data and found that some missing values for visibility were auto filled with the maximum threshold values (11.8) by using a visibility reformat program and thus caused the “fluctuation” of visibility. In the revised

manuscript, we have corrected this and please refer to the revised figure for the changes. As for RH, we didn't find any improper treatment of the raw data and the quality is assured.

4. There is no clear definition of fog and haze in the manuscript. It seems that the author mixed them together. The author even attributed the severe haze pollution to several fog events. According to the Kohler theory, fog clearly differs from haze. In haze, aerosol undergoes only hygroscopic growth; while in fog, aerosol is activated and grows freely. However, the definition of fog in meteorology is only based on RH and visibility. With high aerosol loading and RH, haze can also cause a visibility of hundreds meters, which might be classified as fog in weather reports. In this manuscript, "the role of fog processing" is discussed. I think the fog should be defined according to its microphysical properties rather than RH and visibility. I believe the "fog events" during the study period are mostly haze.

Thanks for the comments and suggestions. As no microphysical properties were measured in this study, we had to rely on the weather reports to indicate whether there were fog events or not. We agree with the reviewer that fog and haze events sometimes could be mixed together. In the revised manuscript, to avoid the misunderstandings, we changed "the role of fog processing" to be "the role of high humidity on the atmospheric processing". In Section 3.4 and 3.5, we mainly focus to discuss the partition of acidic aerosol in different phases under different RHs rather than the discussion of fog processing. Please refer to the revised manuscript for those changes.

5. It is confusing whether the author wants to discuss one typical severe pollution event or the general condition in January 2013. Section 3.3-3.5 focus on one pollution event in January of 2013, however, the discussion in section 3.1 and 3.2 is based on the average conditions in January. Firstly, a period of one month might only include several synoptic processes, which is too short for a good representative and to make comparisons between different years. Also, Chinese spring festival is in January in some years, which may cause a change of emissions and influence the monthly average. Secondly, I do not think one can prove a pollution event of 7 days (may be longer but not shown in the manuscript) to be "a historical pollution event" and stem from "abnormal meteorological conditions" based on the average data of the whole month of January. The monthly average mixes lasting time and pollution intensity together. A high average can be

yielded from either a long-time light pollution event or a short-time severe pollution event. Concerning the title, I think it makes more sense to focus on one severe pollution event.

Thanks for the comments and good suggestion. In the revised manuscript, we have deleted the discussion about the long-term trends of surface PM_{10} and column AOD in Beijing as the reviewer commented. We still keep the discussion about the spatial distribution of PM2.5 and pollutant gases over China in January, 2013 as we think it is better to give a picture about the severe air pollution on the regional scale in the beginning, and then explicitly discuss the formation mechanism of haze by using a typical city.
In the revised manuscript, as the reviewer suggested, we have more focused on the analysis of one severe pollution episode. More details could be found in Section 3.3.2, 3.4 & 3.6.

6. A lot of discussion is about aerosol hygroscopic growth and the impact of RH on visibility. There are a lot of studies about the relationships between the aerosol hygroscopicity and chemical composition, and the impact of aerosol hygroscopic growth on its optical properties, based on the in-situ measurements of aerosol hygroscopic growth factor (g(RH)) or scattering hygroscopic growth factor (f(RH)) in the North China Plain. The results of these studies are much more clear, specific and in-depth. However, it seems that the author did not notice those papers. Some description and discussion on this issue in the manuscript is therefore unconvincing or inappropriate.

Thanks for the comments. We agree with the reviewer that a lot of studies have demonstrated the relationships between aerosol chemical composition and the aerosol hygroscopic growth. Generally, our results showed consistence with previous studies. In this regard, we think there is no need to address this issue and thus we have deleted the original Section 3.4 “Impact of relative humidity on aerosol chemistry”. Instead, we shifted our focus on how the humidity controlled the aqueous processing and estimation of regional transport contribution by using a new tracer technique. Please refer to the revised manuscript for those changes.