Manuscript # acp-2017-455

Reply to Referee #3

We are very grateful to all important and helpful comments from the referee. The followings are our responses to each comment in detail.

Summary: In this article, the authors analyzed and quantified the relationships among $PM_{2.5}$ concentration, visibility and PBL height for the haze and fog-haze mixed events using the data from several state-of-the-art instruments, and then showed the corresponding meteorological conditions for the two typical cases. Similar analyses have been implemented by many previous studies and the novelty of this study is actually not enough. However, the detailed estimations of this study can still provide some valuable information for the haze early warnings. I suggest it to be accepted after several corrections. Note I am not an expert on the atmospheric chemistry, so my assessment on this part may not be accurate.

Specific comments:

1. For any journal, the first requirement is that the abstract of the article should be briefly and concisely. However, the abstract of this study is too redundant and including some valueless information that would be lowering the readability. So, this part of the article is suggested to be re-worded in the next version that just the highlights from this research are needed.

Reply 1:

Thanks for the suggestion, the abstract has been re-worded in the revised manuscript, shown as below:

"The air quality and visibility are strongly influenced by aerosol loading, which is driven by meteorological conditions. The quantification of their relationships is critical to understanding the physical and chemical processes and forecasting of the polluted events. We investigated and quantified the relationship among $PM_{2.5}$ (particulate matter with aerodynamic diameter is 2.5 μm and less) mass concentration, visibility and planetary boundary layer (PBL) height in this study based on the data obtained from four long–lasting haze events and seven long–lasting fog–haze mixed events from January 2014 to March 2015 in Beijing city. The statistical results show that there was a negative exponential function between the visibility and the $PM_{2.5}$ mass concentration for both haze and fog–haze mixed events. However, the fog–haze events caused a more obvious decrease of visibility than that for haze events due to the formation of fog droplets that could induce higher light extinction. The $PM_{2.5}$ concentration had inversely linear correlation with PBL height for haze events and negative exponential correlation for fog–haze mixed events, indicating that the $PM_{2.5}$ concentration is more sensitive to PBL height in fog–haze mixed events. The visibility had positively linear correlation with the PBL height with the R^2 of 0.35 in haze events and positive exponential correlation with the R^2 of 0.55 in fog–haze mixed events. We also investigated the physical mechanism responsible for these relationships among visibility, $PM_{2.5}$ concentration and PBL height through typical haze and fog–haze mixed event, and found that a double inversion layer formed in both typical events and played critical roles in maintaining and enhancing the long–lasting polluted events. The upper–level stable inversion layer formed by the persistent southwest warm and humid airflow caused the $PM_{2.5}$ accumulation and subsequent surface cooling as well as the formation of a weak low–level inversion layer. The formation of low–level inversion layer further enhanced the $PM_{2.5}$ accumulation and surface cooling process, and induced a strong descending process of the upper–level inversion layer with warm and humid air, which significantly strengthened the PBL stability and formed a deep stable PBL in the daytime, and in return rapidly increased the $PM_{2.5}$ concentration. This positive feedback was particularly strong when the $PM_{2.5}$ mass concentration was larger than 150–200 μg m^{-3}. Therefore, the formation and subsequent descending processes of the upper–level inversion layer should be an important factor in maintaining and strengthening the long–lasting severe polluted events, which has not been revealed in previous publications. The interactions and feedbacks between $PM_{2.5}$ concentration and PBL height linked by radiation process caused an obvious and more rapid increase of $PM_{2.5}$ concentration and a significant and long–lasting deterioration of air quality and visibility in fog–haze mixed events."

2. Since 2013, increased studies have addressed the impact of climate changes on the haze pollutions over China. For example, weakened East Asian winter monsoon (Li, Qiang, et al., 2016: Interannual variation of the wintertime fog-haze days across central and eastern China and its relation with East Asian winter monsoon. Int. J. Climatol., 36, 346-354), reduced Arctic sea ice (Wang, Huijun, et al., 2015: Arctic sea ice decline intensified haze pollution in eastern China, Atmos. Oceanic Sci. Lett.,8, 1–9), Tibetan Plateau warming (Xu, X., et al., 2016: Climate modulation of the Tibetan Plateau on haze in China. Atmos. Chem. Phys., 16, 1365-1375), ENSO variability (Gao, Hui, et al., 2015: Influences of El Nino Southern Oscillation events on haze frequency in eastern China during boreal winters. Int. J. Climatol., 35, 2682-2688), etc. all showed important roles on the haze occurrences across China. I think this part of the work should be reviewed in the introduction. Additionally, there are also some studies presented the meteorological conditions for the haze pollutions from climatological perspectives (Zhang, Renhe, et al., 2014: Meteorological conditions for the persistent severe fog and haze event over eastern China in January 2013. Sci. China Earth Sci., 57, 26–35; Chen, Huopo, et al., 2015: Haze days in North China and the associated atmospheric circulations based on daily visibility data from 1960 to 2012. J. Geophys. Res. Atmos., 120, 5895-5909), which can be compared with the case analyses in this study, further increasing the readability.

Reply 2:

Thanks for the important suggestion. We have added some recent papers in the introduction section of the revised manuscript, as "In addition, the interactions between aerosol pollution and climate change have been substantially addressed in recent publications, for example, anthropogenic climate change (Cai et al., 2017), reduced Arctic sea ice (Wang et al., 2015; Zou et al., 2017), the Tibetan Plateau warming (Xu et al., 2016), influences of ENSO events on haze frequency in eastern China (Gao and Li, 2015), weakened East Asian winter monsoon (Li et al., 2016), decadal weakening of winds (Yang et al., 2016), and enhanced thermal stability of the lower atmosphere (Zhang et al., 2014; Chen and Wang, 2015). The dust–wind interaction (Yang et al., 2017a) and upwind transport (Yang et al., 2017b) could also intensify haze events in China."

3. The difference of the separating criterions of the long-lasting haze and fog-haze mixed events is the different value of minimum visibility that minimum visibility larger than 1km for haze events and smaller than 1km for fog-haze mixed events. This is the self-criterion or from the other research? The humidity is a key factor for the separation of the fog and haze events, why the relative humidity has not been considered?

Reply 3:

Thanks for the comment. As you said, the RH is critical factor for separating haze from fog events. Actually, the definition we used is from international definition of fog event. Fog is an observed horizontal visibility below 1000 m in the presence of suspended water droplets and/or ice crystals (NOAA, 1995), which means that when the horizontal visibility is below 1000 m, the fog events occur. Since the horizontal visibility for atmospheric haze event is usually larger than 1000 m, only the fog occurs the visibility can decrease to be less than 1000 m. So that is why we use the minimum visibility to define fog and haze events.

Theoretically, when a fog event occurs, the RH has to reach over 100 %. However, it is difficult to measure RH accurately, so in most cases, we use RH value of 90 % or 95 % as criterion to separate fog and haze. In fact, in the study region, when the RH is high enough, the fog and haze are usually co-existed. The haze aerosols can be transformed to fog droplets under certain conditions according to the Köhler curve (Köhler, 1936). It should be noticed that the situations such as heavy rain event or light fog events, which cause the horizontal visibility to be below or above 1000 m are not considered here. In addition, since we focus on long–lasting severe fog and haze event, we also include factors such as the lasting time and $PM_{2.5}$ mass concentration as additional criteria. To be more clearly, the corresponding text have been modified in the revised manuscript.

We also revised Table 1 to include more parameters such as duration and maximum RH of the pollutant events, shown as below:

Table 1: The long–lasting haze and fog–haze mixed events from January 2014 to March 2015 in Beijing city

Type	Starting date / time	Ending date / time	Minimum visibility (m)	Duration (h)	Maximum $PM_{2.5}$ ($\mu g\ m^{-3}$)	Maximum RH (%)	Weather phenomenon
Haze events	2014.01.21/ 15:00	2014.01.24/ 15:00	1364	73	264	68	–
	2014.04.11/ 22:00	2014.04.14/ 23:00	1113	74	304	89	–
	2015.02.12/ 21:00	2015.02.16/ 10:00	1667	86	263	77	–
	2015.03.04/ 22:00	2015.03.08/ 10:00	1886	83	266	72	–
Fog–haze mixed events	2014.02.19/ 21:00	2014.02.26/ 20:00	647	168/76[b]	269	92	02.26/16:00–21:25 Drizzle rain
	2014.03.22/ 22:00	2014.03.28/ 14:00	664	137/13[b]	417	94	3.28/4:30–6:20 Drizzle rain
	2014.10.06/ 22:00	2014.10.11/ 18:00	500	117/48[b]	391	100	10.08/6:40–7:50 10.08/10:30–11:50 Drizzle rain
	2014.10.16/ 21:00	2014.10.20/ 23:00	964	99/3[b]	322	100	–
	2014.10.22/ 4:00	2014.10.26/ 4:00	258	97/24[b]	379	100	–
	2014.10.28/ 23:00	2014.11.01/ 5:00	837	79/1[b]	184	100	10.29/23:00– 10.30/00:10 10.31/15:10–16:30 Drizzle rain
	2015.01.12/ 17:00	2015.01.16/ 3:00	526	83/8[b]	297	93[a]	01.14/10:00–10:20 snow

[a] the maximum RH of all valid data except missing measurements.

[b] fog–haze mixed event duration / fog duration.

4. In the context, the authors mentioned that "The PBL height derived by MPL is usually used as a reference in detecting the aerosol vertical distribution by more advanced and powerful lidars.", however, the authors also mentioned in the following paragraph that there are also some uncertainty existed for the MPL to determine the PBL height. This seems to be conflict.

Reply 4:

Thanks for the comment. Generally, MPL is a reliable tool to retrieval PBL height, however, it cannot work well in some situations such as that the aerosol concentration becomes uniform in the vertical direction (Tang et al., 2016). We revised this part based on your comment, as "The PBL heights retrieved by the attenuated backscatter profile of MPL and CL31 still exist some uncertainties (Tang et al., 2016; Geiß et al., 2017). Tang et al. (2016) founded that PBL height cannot be correctly obtained through sudden changes in the attenuated backscatter profiles. Such as in situation that the strong northerly winds with dry and clear air masses prevail in observation site, the atmospheric aerosols spread rapidly and became uniform in the vertical direction, the PBL height was substantially underestimated."

5. How about the statistical relationship between $PM_{2.5}$ ma concentration and PBL height from CL lidars?

Reply 5:

Good comment. Since MPL is much more powerful tool in retrieving PBL height, we used the data from MPL to investigate the relationship between $PM_{2.5}$ mass concentration and PBL height. When using CL data, we found that the basic relationship could be the same as that by using data from MPL, however, the correlation coefficients decreased substantially. The figure S2 below shows that the $PM_{2.5}$ concentration has inversely linear correlation with the PBL height with the R^2 of 0.2 for haze events and negative exponential correlation with the R^2 of 0.34 for fog–haze mixed events. R^2 are both lower than that determined by MPL. We have added these descriptions to section 3.2 in the revised manuscript and added Fig. S2 to the supplement.

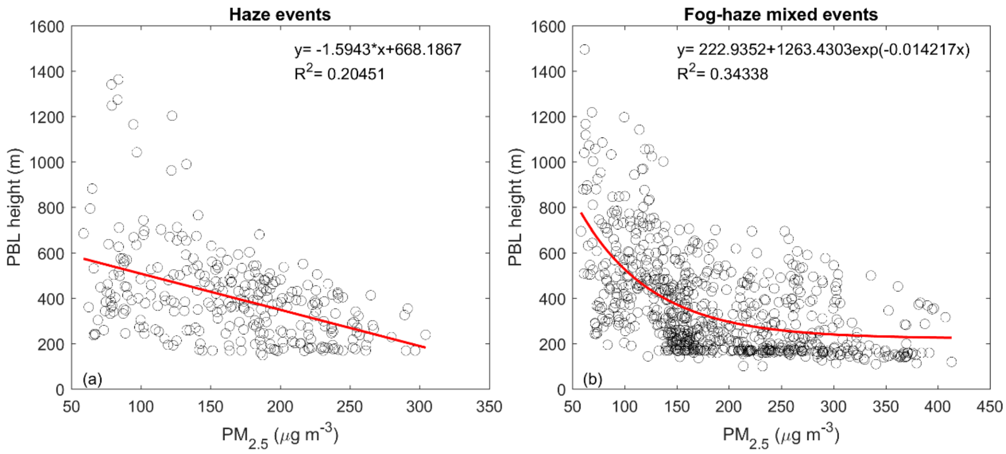

Figure S2: Relationship between PBL height derived by CL31 and $PM_{2.5}$ mass concentration for (a) haze and (b) fog–haze mixed events from January 2014 to March 2015 in Beijing city

6. In this study, the authors just showed the meteorological conditions for two typical haze events? Why chose these two cases from 11 cases? The composite analysis method is suggested for the further analysis if conveniently.

Reply 6:

Thanks for the comment. The main reason to choose two typical cases for detailed analyses is to consider their representativeness and data completeness. This study focuses on the long–lasting haze and fog–haze mixed events, we choose typical long–lasting pollutant events and also consider whether the data for these cases is complete or not. Since the most data we used in this study were observed and supported by just a research project, it is not an easy thing to maintain these advanced instruments and obtain a complete data case from its occurrence to the end.

We have added some descriptions for this comment in the revised manuscript, as "To clarify the physical mechanism responsible for the relationship among $PM_{2.5}$, visibility and PBL height obtained above, two typical cases of long–lasting haze and fog–haze mixed events are presented and further investigated in considering their representativeness and data completeness of all cases (Table 1). In all haze events, the haze event observed from 11 to 14 April, 2014 was highly polluted with the maximum $PM_{2.5}$ concentration of 304 μg m^{-3} and minimum visibility of 1113 m. For all fog–haze mixed events, the fog duration was considered firstly. Two cases are chosen, in which the fog duration accounted for more than 40 % of the total. One was observed from 19 to 26 February 2014, and the other was occurred from 6 to 11 October 2014. The maximum $PM_{2.5}$ concentration was more higher and the maximum RH reached to 100% in the fog–haze event occurred from 6 to 11 October 2014, which was chosen as typical fog–haze event for the following study."

7. The authors presented detailed analyses on the meteorological conditions for the long-lasting haze and fog-haze mixed events. However, I am still not clear what the difference for the meteorological conditions between these two cases. So, the comparison discussion in this aspect should be added in the section of Conclusion and Discussions, not just showing the common features as the current MS did.

Reply 7:

Thanks to the referee for the suggestion. The main differences of meteorological condition for two cases are humidity and duration. Although both cases occurred in a stable weak pressure field covering northern China, the haze event was relatively drier with shorter duration while the fog–haze mixed event was highly humid with longer duration. Since the fog droplets was formed in the fog–haze mixed event, the radiation reduction at surface was more obvious and stronger, which caused stronger descending process of the upper inversion layer, and formed a more stable PBL.

Based on your comment, we added some descriptions relevant to the differences of meteorological conditions between two cases in the section of conclusions and discussions in the revised manuscript, as "The main differences of meteorological condition for two cases are humidity and duration. Although both cases occurred in a stable weak pressure field covering northern China, the haze event was drier and duration was shorter while the fog–haze mixed event was more humid and had a longer duration. Since the fog droplets were formed in the fog–haze mixed event, the radiation reduction at surface was more obvious and stronger, and caused stronger descending process of the upper inversion layer. In most cases, light precipitation (drizzle rain or light snow) occurred during the fog–haze mixed event while in all haze events during the observation period, there was no precipitation. The fog–haze mixed event was more favorable to form extremely high mass concentration of $PM_{2.5}$ (>300 μg m^{-3}) than the haze event."

8. To increase the readability, the location of Beijing is suggested to be highlighted in Figure 11 and 17.

Reply 8:

Thanks for the suggestion. The location of Beijing has been added in the Fig.S3 and Fig.S4 in the revised manuscript. The revised figures are shown as below:

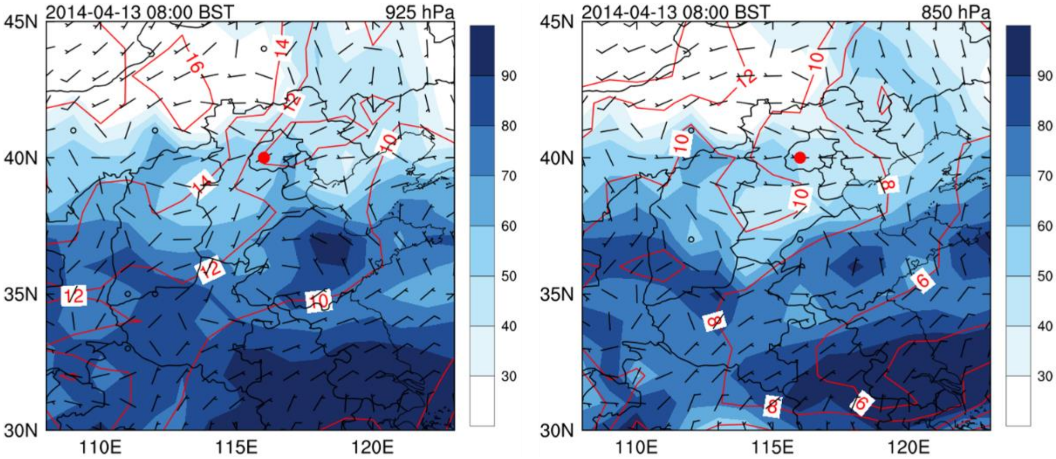

Figure S3: Temperature (red contour lines, units: ℃), RH (color shading, units: %) and wind (wind bar) distribution in (a) 925 hPa and (b) 850 hPa at 08:00 BST on 13 April 2014. The red dot represents the site of CMA

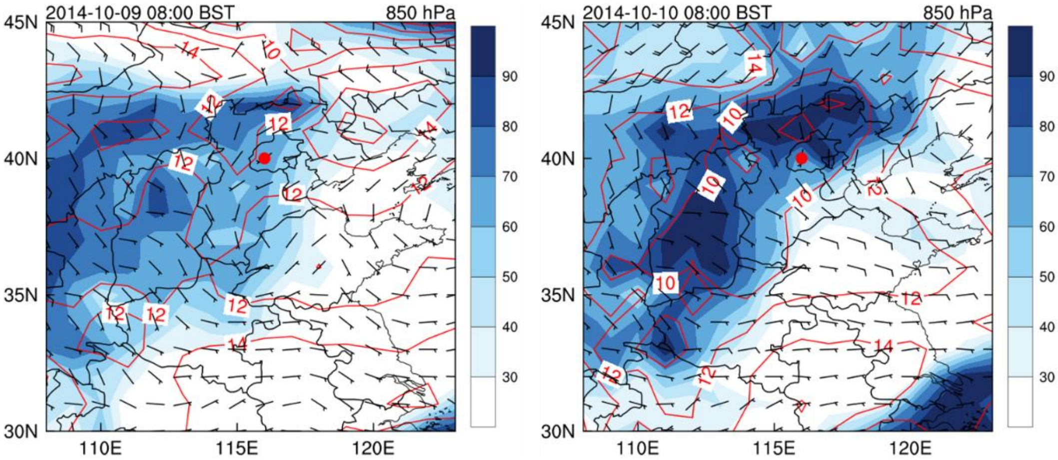

Figure S4: Temperature (red contour lines, units: ℃), RH (color shading, units: %) and wind (wind bar) distribution in 850 hPa at 08:00 BST on (a) 9 October and (b) 10 October 2014. The red dot represents the site of CMA.

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