# Authors' Response to Anonymous Referee #3

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**Overall:**

We thank the reviewer for the careful reading and the valuable comments that helped improving our paper.

In the original manuscript, there were two problems argued by the three reviewers. One was section 3.3.4 "New particle formation", the other was section 3.2.6 "Visibility empirical equation". We thought over and over about these two questions and analyzed the data in depth. Lastly, we reorganized the original section 3.2.4 "New particle formation" with section 3.2.4 "Gas to particle conversion and aerosol growth". We merged the section 3.2.6 "Visibility empirical equation" into the section 3.2.5 "Impact of hygroscopic growth for aerosol scattering f(RH)" in the revised manuscript. In addition, we tried our best to analyze the meteorological condition in depth. We also deleted some incorrect sentences and conclusions in the revised manuscript.

We would like to answer the comments and suggestions one by one as following.

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This paper reports a very interesting episode of high pollutant concentrations that developed during a period of high pressure over the region around Beijing. The study is generally well documented and described in this paper; however, the conclusions that are drawn as to the cause of the high concentrations are not necessarily correct. Some of what is concluded seem to be supported by the measurements but there are several factors that have not been sufficiently explored in the analysis.

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***Response:***

Of course, there should be many factors affecting the formation and evolution of the regional haze. In the revised manuscript, the causes of the high concentrations of air pollution were summarized as following: meteorological condition, high intensity of local pollution, shallow PBL, gas to particle conversion, and hygroscopic growth for aerosol scattering. We tried our best to explore these five factors in depth and also rectified some incorrect statements in the revised manuscript.

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Two of the main conclusions that are reported in the abstract and summary, and that are developed in the main text, are that it was mainly the high intensity of local pollutions that led to this particular haze event and that it was new particle formation that was the most important factor contributing to this haze.

Although neither of these conclusions are necessarily incorrect, I would argue that since the emissions from Beijing sources probably change very little from day to day or month to month, it was the meteorological factors that led to the haze event, not any change in the sources of emissions.

The meteorology is partially discussed but is in need of much more analysis that includes looking at the vertical structure of the temperature, humidity and winds using the local radiosonde information. The introduction of the surface maps was a good start, as was the use of the lidar to look at how the aerosols were distributed. This is not enough however to tell the complete story. The vertical profile of temperature and dew point temperature allow you to calculate the potential temperature and hence the stability of the boundary layer.

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***Response:***

We accepted this suggestion and intensified the analysis of meteorological conditions in the revised manuscript, especially in section 3.2.1 “Meteorological analysis”, section 3.2.2 “Local source of pollutions”, and section 3.2.3 “Development of PBL”.

We are sorry for no radiosonde data illustrating the vertical structure of the temperature, humidity and winds. But, we add the analysis of surface maps of air pressure in the revised manuscript.

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The boundary layer is defined by its thermodynamic structure and not by its aerosol population. Perhaps there were morning inversions also associated with the increased aerosol concentration?

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***Response:***

We agreed with the reviewer’s comment. The boundary layer is defined by its thermodynamic structure and not by its aerosol population. But, the presence of the boundary layer and inversion layer will inevitably be reflected in the distribution of aerosol profiles, so, we retrieved the height of the boundary layer by aerosol profile.

Lidar has been used for many years to provide accurate measurements of the top of the boundary layer (Boers et al., 1984; Sasano, 1985; Menut et al., 1999). Several methods have been employed to extract the PBL height from lidar data. These include critical threshold techniques (Melfi et al., 1985), gradient techniques (Hayden et al., 1997; Hoff et al., 1996), wavelet analyses (Cohn et al., 1998) and techniques using idealized profiles (Steyn et al., 1999). The fundamental premise takes advantage of

the large gradient in aerosol concentration generally evident between the boundary layer aerosols and those found in the free troposphere.

As an example, mixed layer depths were derived from potential temperature profiles from aircraft, high-altitude balloon sonde and tethersonde measurements taken during the Pacific '93 field study in the Lower Fraser Valley of southern British Columbia (Hayden et al., 1997). These lidar-derived aerosol depths compared well with the meteorologically derived mixed layer depths (Hayden et al., 1997).

**References:**


Boers, R., Eloranta, E.W., Coulter, R.L., Lidar observation of mixed layer dynamics: tests of parameterized entrainment models of mixed layer growth rate. Journal of Climate and Applied Meteorology 23, 247–266, 1984.

Cohn, S.A., Mayor, S.D., Grund, C.J., Weckwerth, T.M., Senff, C. The Lidars in Flat Terrain (LIFT) experiment. Bulletin of the American Meteorological Society. 79(7), 1329-1343, 1998.

Hayden, K. L., Anlauf, K. G., Hoff, R. M., Strapp, J. W., Bottenheim, J. W., Wiebe, H. A., Froude, F. A., Martin, J. B., Steyn, D. G., McKendry, I. G. The vertical chemical and meteorological structure of the boundary layer in the Lower Fraser Valley during Pacific '93. Atmospheric Environment, 31(14), 2089-2105, 1997.

Hoff, R.M., Guise-Bagley, L., Staebler, R.M., Wiebe, H.A., Brook, J., Georgi, B., Dusterdiek, T., Lidar, measurement over the Paris area. Applied Optics 36 (6), 945–954, 1996.

Melfi, S.H., Spinhirne, J.D., Chou, S.-H., Palm, S.P., Lidar observation of vertically organized convection in the planetary boundary layer over the ocean. Journal of Climate Applied Meteorology 24, 806–821, 1985.

Menut, L., Flamant, C., Pelon, J., Flamant, P., Urban boundary layer height determination from lidar nephelometer and in-situaerosol experiments in Southern Ontario. Journal of Geophysical Research Atmosphere 19(D10), 199–209, 1999.

Sasano, Y., Observational study on the atmospheric mixed layer and transition layer structure using a Mie lidar. Meteorological Society of Japan 63 (3), 419–435, 1985.

Steyn, D.G., Baldi, M., Hoff, R.M. The detection of mixed layer depth and entrainment zone thickness from lidar backscatter profiles. Journal of Atmospheric and Oceanic Technology, 16, 953-959, 1999.

Strawbridge, K.B., Snyder, B.J. Planetary boundary layer height determination during Pacific 2001 using the advantage of a scanning lidar instrument. Atmospheric Environment, 38, 5861-5871, 2004.


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The low wind speeds and shallow boundary layer were certainly the primary cause of intensified haze; however, the primary driving force of the decreased depth of the boundary layer and the outflow seen on the AOD maps was the larger scale circulation and high pressure over that region. This is the story that needs to be told.

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***Response:***

We accepted this suggestion add the corresponding analysis in section 3.2.3 “development of PBL” in the revised manuscript. For example, we added this sentence “Large scale high pressure moved over the Beijing-Tianjin-Tangshan Region as described in section 3.2.1, subsidence airflow restricted the development of the height of the PBL. Meanwhile, the development of the PBL was driven by thermal

(air temperature) and dynamic (wind speed) factor from the local perspective." in section 3.2.3 "development of PBL".

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High pressure means convergence aloft, divergence on the surface. This is the reason that as the high pressure moved over the region the boundary layer depth decreased, the concentration of particles increased, humidity increased and temperature decreased leading to deliquescence of the particles and visibility decreased. This is a classic pollution event seen often in the wintertime but is not restricted to just cold weather. Yes, winds were low because there were no strong pressure gradients; however, the divergence at the ground led to large scale outflow seen by the increase AOD over time in Fig. 7.

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***Response:***

Thanks a lot for reviewers' analysis on synoptic condition and we accepted this suggestion adding the meteorological analysis in the revised manuscript. For example, We added this sentence "Large scale high pressure moved over the Beijing-Tianjin-Tangshan Region as described in section 3.2.1, subsidence airflow restricted the development of the height of the PBL." in section 3.2.3 "development of PBL". We also added this sentence "The divergence airflow at the ground (section 3.2.1) led to large scale air outflow from Beijing to down-wind area. According to the trend of the isobars in figure 4, pollutants from Beijing were diffused in the northeast - southwest direction, as seen by the increased AOD over time in figure 7." in section 3.2.2 "local source of pollutions".

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This is a very nice example that should be published but it has to be better documented with respect to the actual meteorological factors that led to the high pollution event. The authors are correct that this case can be quite instructional, especially for those forecasting such events since they are clearly tied to meteorology that can be forecast fairly accurately. If none of the authors are experience meteorologists, than I strongly recommend that they consult with one.

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***Response:***

Thanks a lot and we consulted with a meteorologist. We added the analysis on synoptic condition in the revised manuscript.

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With respect to the claim that new particle formation is the most important contributor to the high particle concentrations, I have to ask that the authors do more research on the conditions that are necessary to bring about gas to particle conversion. A case of new particle formation is described by Dunn et al (2004) in Mexico City, an urban

area similarly polluted as Beijing. They concluded that three criteria consistently characterize new particle formation events within the city: the events occur during daylight hours, while $SO_2$ is elevated, and when particulate matter mass concentrations are at significantly lower values than their averages.

The criteria of elevated $SO_2$ and low PM is critical to new particle formation. There has to be high $SO_2$ vapor pressure and at the same time, very low available surface area to which the $SO_2$ will preferentially diffuse, since that will have a higher probability than forming clusters of $SO_2$ molecules that grow into particles. In the present case, on the days that are being called new particle formation days, there is high concentrations of PM and low $SO_2$. The $SO_2$ concentrations are actually increasing after the says that are being called new particle formation.

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***Response:***

We accepted the reviewer's suggestion and studied the relationship between new particle formation and $SO_2$ and $PM_{2.5}$. The result was illustrated by the following figure.

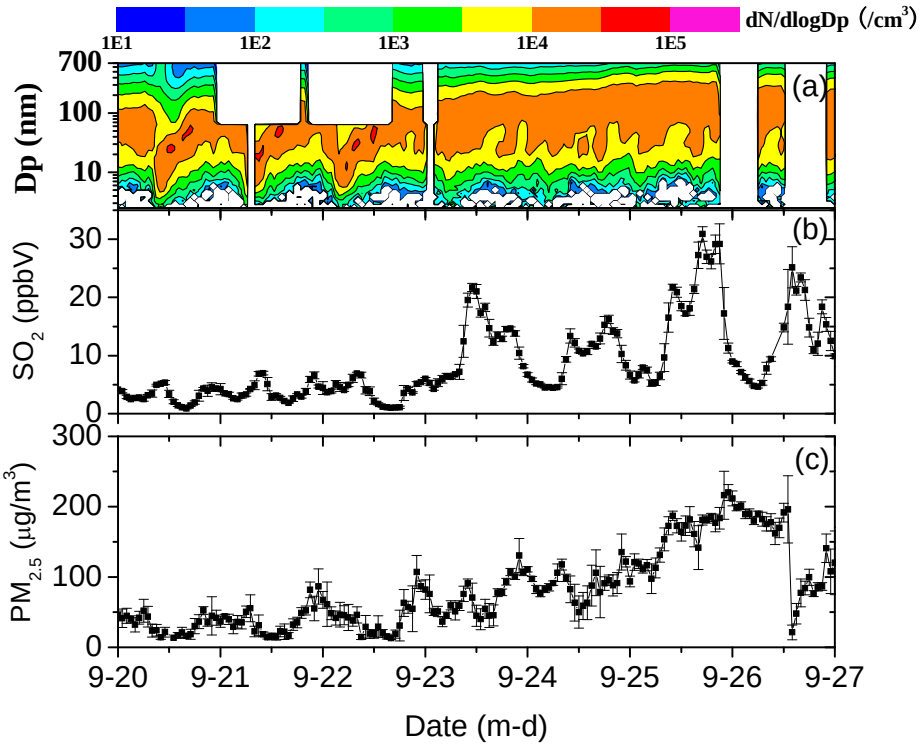


**Figure 1. Temporal variance of (a) particle size distribution, (b) $SO_2$ mixing ratio, and (c) $PM_{2.5}$ mass concentration from September 20 to September 27.**

The characterization of new particle formation in this study was consistent with the result by Dunn et al (2004) in Mexico City: the events occur during daylight hours,

$SO_2$ is elevated, and when particulate matter mass concentrations are at significantly lower values than their averages.

In the revised manuscript, we replaced the original figure 9 with this figure and cited the result by Dunn et al (2004).

**References:**

Dunn, M. J., Jiménez, J.-L., Baumgardner, D., Castro, T., McMurry, P. H., and Smith, J. N.: Measurements of Mexico City nanoparticle size distributions: Observations of new particle formation and growth, Geophys. Res. Lett., 31, L10102, 2004.

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## Other comments:

Page 16264: Line 7 – What were the objectives of the field experiment? Why only 8 days?

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***Response:***

As described in the abstract, the main objective of this study was to investigate the formation and evolution mechanism of the regional haze in megacity Beijing by analyzing the process of a severe haze that occurred 20–27 September 2011. This case analysis would be useful to better understand the causes of the poor air quality over megacity Beijing, which was a first step for ultimately improving it in the future. For example, the government could reduce emissions when synoptic conditions were expected to cause recurrences of the phenomenon discussed in this study.

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Page 16264: Lines 18&20 – The TEOM and meteorological sensors are not in the instrumentation table.

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***Response:***

We accepted this suggestion and added the TEOM and meteorological sensors in table 1 in the revised manuscript.

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Page 16265: Line 1 – What was the model number of the CPC?

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***Response:***

The model number of the CPC was 3010. We rectified the sentence in the revised manuscript with “Particles of different sizes were classified with differential mobility analyzer (DMA, Hauke-type) and their concentrations were measured with a condensation particle counter (CPC, TSI Model 3010)”.

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Page 16265: Line 10 – "Aerosol absorption coefficient was then calculated by the product of mass concentration of BC by specific absorption coefficient (6.6 $m^2g^{-1}$), which was from the manufacture guide." This seems backward to me. The MAAP measures light attenuation, not BC. Why not use the absorption directly measured?

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***Response:***

The MAAP serves to determine the mass concentration of black carbon suspended in the ambient atmosphere. The principle of determination is based on the light attenuation (absorption) by BC. The MAAP directly give the mass concentration of BC in unit of $\mu g/m^3$ using specific absorption coefficient (6.6 $m^2g^{-1}$) introduce by the manufacture. So, aerosol absorption coefficient was calculated by the product of mass concentration of BC by specific absorption coefficient (6.6 $m^2g^{-1}$).

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Page 16267: Line 5 – Suggest including the CN concentration on one of the time series plots.

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***Response:***

We accepted this suggestion and added the aerosol number concentration for nucleation mode, Atiken mode, and accumulation mode in the revised manuscript. This figure as following is clarified in section 3.2.4 "Gas to particle conversion and aerosol growth".

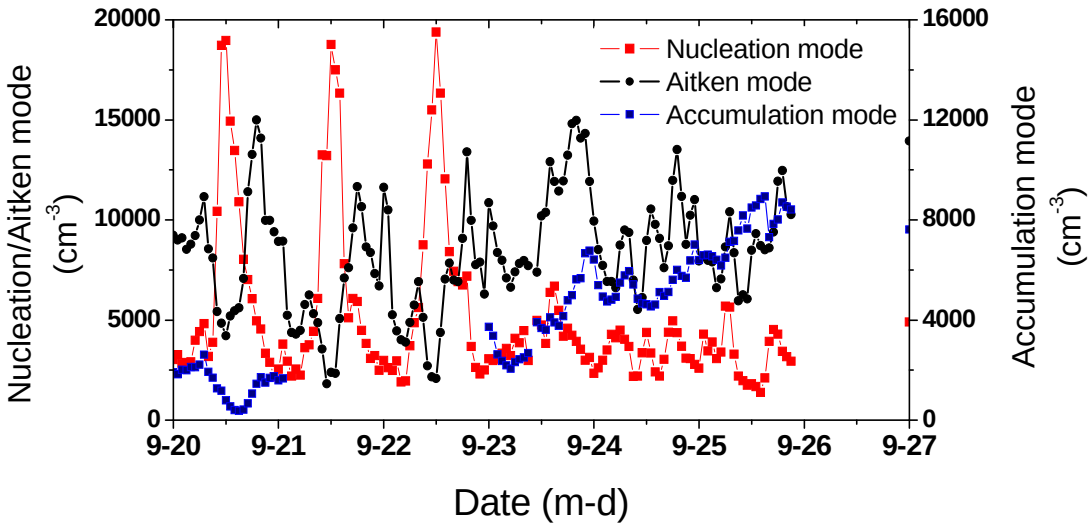


**Figure 2. Temporal variations of aerosol number concentration for nucleation mode, Atiken mode, and accumulation mode.**

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Page 16269: Line 7- This is a weak argument. Maximum temperature were still in the upper 20's, more than enough to heat and grow the boundary layer. As I discuss in my opening paragraphs, larger scale forcing is the most likely reason. The air temperature doesn't "sharply decrease".

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***Response:***

We accepted this comment and deleted the sentence "In addition, the air temperature sharply decreased on September 24 and the decreasing trend kept till September 26, which would cause the lack of the thermal dynamics for the development of the PBL (discussed in section 3.2.3)." in the revised manuscript. Additionally, we intensified the analysis of meteorological conditions in the revised manuscript.

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Page 16270: Line 4 – The lidar backscatter signal doesn't actually give you the top of the boundary layer. The boundary layer is defined by its thermodynamic structure not the aerosol properties. The black line here is only an estimate.

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***Response:***

We agreed with the reviewer's comment. The boundary layer is defined by its thermodynamic structure and not by its aerosol population. But, the presence of the boundary layer and inversion layer will inevitably be reflected in the distribution of aerosol profiles, so, we retrieved the height of the boundary layer by aerosol profile.

Lidar has been used for many years to provide accurate measurements of the top of the boundary layer (Boers et al., 1984; Sasano, 1985; Menut et al., 1999). Several methods have been employed to extract the PBL height from lidar data. These include critical threshold techniques (Melfi et al., 1985), gradient techniques (Hayden et al., 1997; Hoff et al., 1996), wavelet analyses (Cohn et al., 1998) and techniques using idealized profiles (Steyn et al., 1999). The fundamental premise takes advantage of the large gradient in aerosol concentration generally evident between the boundary layer aerosols and those found in the free troposphere.

As an example, mixed layer depths were derived from potential temperature profiles from aircraft, high-altitude balloon sonde and tethersonde measurements taken during the Pacific '93 field study in the Lower Fraser Valley of southern British Columbia (Hayden et al., 1997). These lidar-derived aerosol depths compared well with the meteorologically derived mixed layer depths (Hayden et al., 1997).

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Hoff, R.M., Guise-Bagley, L., Staebler, R.M., Wiebe, H.A., Brook, J., Georgi, B., Dusterdiek, T., Lidar, measurement over the Paris area. Applied Optics 36 (6), 945–954, 1996.

Melfi, S.H., Spinhirne, J.D., Chou, S.-H., Palm, S.P., Lidar observation of vertically organized convection in the planetary boundary layer over the ocean. Journal of Climate Applied Meteorology 24, 806–821, 1985.

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Strawbridge, K.B., Snyder, B.J. Planetary boundary layer height determination during Pacific 2001 using the advantage of a scanning lidar instrument. Atmospheric Environment, 38, 5861-5871, 2004.

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Page 16270: Section 3.2.4 – Unless the authors can present a more compelling argument than just the size distributions, I strongly suggest removing this entire section and any references to new particle formation in the text. The real story here is the development of the haze event due to meteorological factors.

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***Response:***

We thought over and over on this question. In our opinion, the development of this haze event was not only due to meteorological factors but also due to the high intensity of local pollutions. We only focused on the new particle formation in the original manuscript, but, we added the analysis of aerosol growth and its impact on extinction in the revised manuscript. In theory, the contribution of extinction by the new-formed aerosol could be ignored regarding visibility impairment. But, these new-formed particles would quickly grow to bigger sizes and the scattering ability of these aerosols in accumulation was strong, which resulted in the deterioration of visibility. So, the section 3.2.4 was renamed by "Gas to particle conversion and aerosol growth" and added the corresponding analysis in the revised manuscript.

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Page 16272: Line 11 – The hygroscopic factor was introduced by Hegg et al back in the early 1990's. Please give credit to the ones that originated the idea.

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***Response:***

We accepted this suggestion and cited the original lecture by Hegg et al. in 1993 in the revised manuscript.

**References:**

Hegg, D., Larson,T., Yuen, P-F., A theoretical study of the effect of relative humidity on light scattering by tropospheric aerosols. Journal of Geophysical Research 98(D10), 18,435-18,439. 1993.

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Page 16272: Line 19 – But the average RH in Beijing is not 80%

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***Response:***

Generally, researchers (Sheridan et al., 2002; Xu et al., 2002; Yang et al., 2004; Kim et al., 2006) give the value of f(RH=80%) and compare it with each other. So, we gave the value of f(RH=80%) in the manuscript.

Notes in parentheses "the mean value for the near-surface atmosphere in the Northern Hemisphere (Charlson et al., 1991)" might be redundant and was deleted in the revised manuscript.

**References:**

Kim, J.Y., Yoon, S.C., Jefferson, A., Kim, S.W., 2006. Aerosol hygroscopic properties during Asian dust, pollution, and biomass burning episodes at Gosan, Korea in April 2001. Atmospheric Environment, 40 (8), 1550-1560.

Sheridan, P.J., Jefferson, A., Ogren, J.A., 2002. Spatial variability of submicrometer aerosol radiative properties over the Indian Ocean during INDOEX. Journal of Geophysical Research 108 (D13): Art. No. 8011.

Xu, J., Bergin, M.H., Yu, X., Liu, G., Zhao, J., Carrico, C.M., Baumann, K., 2002. Measurement of aerosol chemical, physical and radiative properties in the Yangtze delta region of China. Atmospheric Environment 36(2), 161-173.

Yang, H., Xu, J., Wu, W.-S., Wan, C.-H., Yu, J.Z., 2004. Chemical characterization of water-soluble organic aerosols at Jeju Island collected during ACE-Asia. Environmental Chemistry 1, 1–5.

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Page 16272: Section 3.2.6 – This complete section is incomprehensible for me. No references for the equations that are introduced and no explanation for why it is in this manuscript. Unless its relevance can be explained and netter document, it would be better left out.

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***Response:***

Section 3.2.5 addressed the importance of RH on aerosol scattering by f(RH), section 3.2.6 tended to introduce the relationship between PM, RH and visibility degradation

based on the result of section 3.2.5. These two sections were closely connected in content. So, we merged section 3.2.5 with section 3.2.6 in the revised manuscript.
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**Lastly, we would express our appreciation to anonymous reviewer and editor for their warm-hearted help and useful suggestions. Thank you very much!!!!**