Authors' Reponse to Anonymous Referee #2

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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 presents measurements of aerosol properties and meteorological conditions during a severe 8-day haze period occurred in Beijing, China, with the aim of identifying the formation and evolution mechanisms of haze. Chemical, physical, and optical measurements of the gas- and particle-phase aerosols are described, and satellite images are used to support the analysis. The main conclusion is that the formation of the haze is the result of a combination of several factors, including high-level pollutants, stagnant air, and the new particle formation, with the new particle formation being the most important factor. While the high-level pollutants and the meteorological conditions are relatively well-established reasons for the formation of haze, the new contribution from this study is that new particle formation is the most important factor that causes haze formation. However, there is lack evidence or analysis that supports this argument, and more in-depth analysis is needed to justify this conclusion.

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Response:

We recognized that new particle formation was not the direct cause of haze formation,

but, gas to particle conversion and new-formed aerosols growth was an important factor resulting in the formation and evolution of regional haze. So, the original section 3.2.4 was rectified with section 3.2.4 “Gas to particle conversion and aerosol growth” in the revised manuscript.

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In addition, despite the multiple measurements, e.g., gas-phase, light scattering/absorption, and satellite images, they are presented independently, in other words, these measurements are not integrated to substantiate the main results. For example, is there quantitative relation of new particle formation and light scattering/extinction? Therefore, more comprehensive analysis could be done to support the conclusions. There are also some descriptions in the text that are not consistent with what the figures show. Specific comments are presented below.

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Response:

The objective of this study was to investigate the formation and evolution mechanism of the regional haze in megacity Beijing. There should be many causes of this haze episode. So, we tried our best collecting various data to clarify the mechanism of haze formation. So many measurements were presented independently. We also tried our best to integrate these independent measurements in the revised manuscript. For example, we added the analysis of meteorological condition in other four sections. We merged the section 3.2.5 with section 3.2.6 to analyze the relationship among visibility, $PM_{2.5}$ and RH.

We had calculated the scattering coefficient of aerosols by Mie model (Bohren and Huffman, 1998) based on the size distribution of aerosols which were measured by SMPS and APS. The contribution of light extinction by new-formed particle could be ignored regarding visibility impairment. But, the new-formed particles subsequently grew in diameter, which would result in the enhancing extinction. We accepted this suggestion and added the calculated scattering coefficients based on aerosols size distribution. That is, the quantitative relation of new particle formation and light scattering was clarified in the revised manuscript.

We recognized that some descriptions in the text were not incorrect and rectified it in the revised manuscript, which were also listed in the section “specific comments”.

References:

Bohren, C.F., Huffman, D.R. Absorption and scattering of light by small particles. John Wiley & Sons, Inc., 1998.

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Specific comments：

1. New particle formation is considered to be the most important factor contributing the formation of haze. This is argument is not justified in the manuscript. There are

three paragraphs in Section 3.2.4 discussing new particle formation, but only the third paragraph describes the new particle formation observed during this study (Fig.9). It is not stated how and why new particle formation is the more important than the other parameters, such as the synoptic weather system. Also Fig. 9 shows that particle grew from 100 nm during non-haze period (Sep 20-23) to 200 nm in haze period (Sep 24-27), the 200 nm particles are too small to significantly enhance aerosol light scattering. To me, it seems the high-level pollutants and the meteorological conditions are the most important parameters for haze formation as well as new particle formation.

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Response:

Firstly, we reorganized the original section 3.2.4 "New particle formation" with section 3.2.4 "Gas to particle conversion and aerosol growth". Secondly, we recognized that the statement "new particle formation is the more important than the other parameters" was incorrect. We deleted this statement in the revised manuscript.

We investigated the relationship among new particle formation, SO_2 and $PM_{2.5}$. The result was illustrated by the following figure.

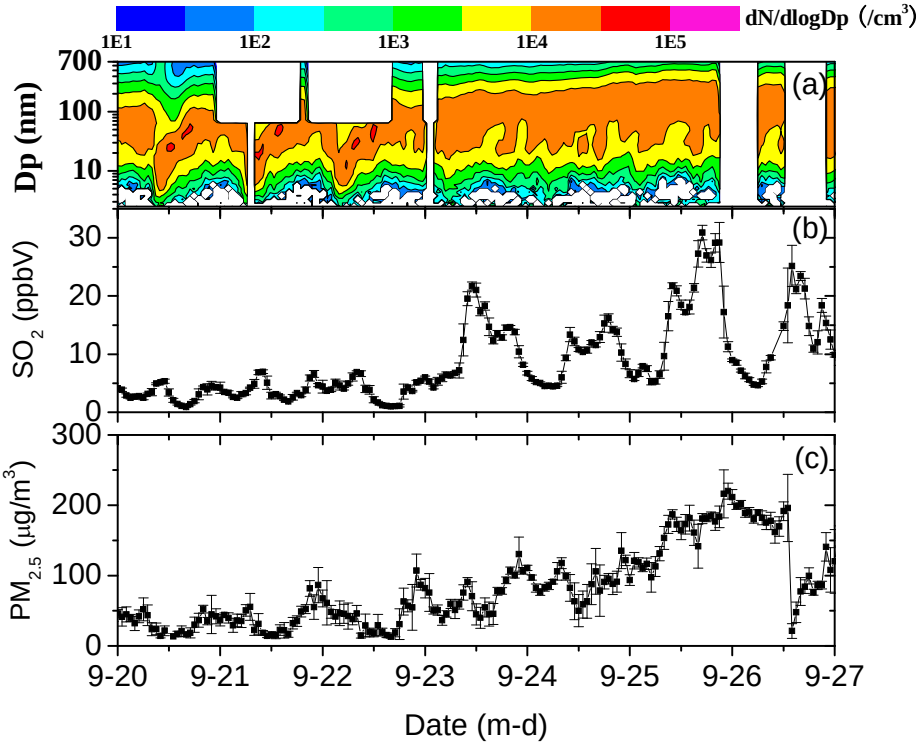

Figure 1. Temporal variance of (a) particle size distribution, (b) SO_2 mixing ratio, and (c)

$PM_{2.5}$ mass concentration from September 20 to September 27.

In the revised manuscript, we replaced the original figure 9 with this figure.

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.

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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2. Section 3.2.5, the discussion of the impact of hygroscopic growth on aerosol scattering is quite general. There is no analysis of how hygroscopic growth during this measurement affects the aerosol scattering and contributes to the haze formation or evolution.

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Response:

In the revised manuscript, 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)". So, hygroscopic growth of aerosols on scattering and lastly on visibility (haze) were analyzed in depth, and the formation and evolution of regional haze can be clearly attributed to the hygroscopic growth of aerosols.

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3. Did the optical measurements give consistent results? For example, b_{ext} is the sum of b_{sp} and b_{ap}, but this is not support by Fig. 2.

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Response:

In this study, the directly measured visibility (*Vis*) was transformed to atmospheric extinction coefficient b_{ext}(RH) at 550nm in unit of inverse megameter (Mm^{-1}) by equation 1 (Koschmieder, 1924; Seinfeld and Pandis, 2006).

$$b_{ext}(RH) = \frac{3.912 * 10^3}{Vis} * (\frac{550}{\lambda}) \quad (1)$$

That is, the b_{ext} was directly measured by visibility sensor (Vaisala FD12) and was not

the sum of b_{sp} and b_{ap}.

References:

Koschmieder, H.: Theorie der horizontalen Sichtweite, Beiträge zur Physik der freien Atmosphäre, 12, 33-53, 1924.

Seinfeld, J. H. and Pandis, S. N.: Atmospheric Chemistry and Physics: from Air Pollution to Climate Change, 2nd ed., John Wiley & Sons, Inc., New York, USA, 2006.

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4. P16267, L13, "Pollutants SO_2, NO_2 and CO, being the emissions from biomass, fuel and coal burning, had the same increasing trend from 20–27 September."

It seems the SO_2 and NO_2 are quite stable and increasing trend is not clear. The trends of SO_2/NO_2 and CO are also different as shown in Fig. 1.

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Response:

The temporal trends of SO_2, NO_2 and CO from September 20 to September 27 were illustrated by the following figure. The red line was the linear fit and the increase of SO_2, NO_2 and CO was obvious.

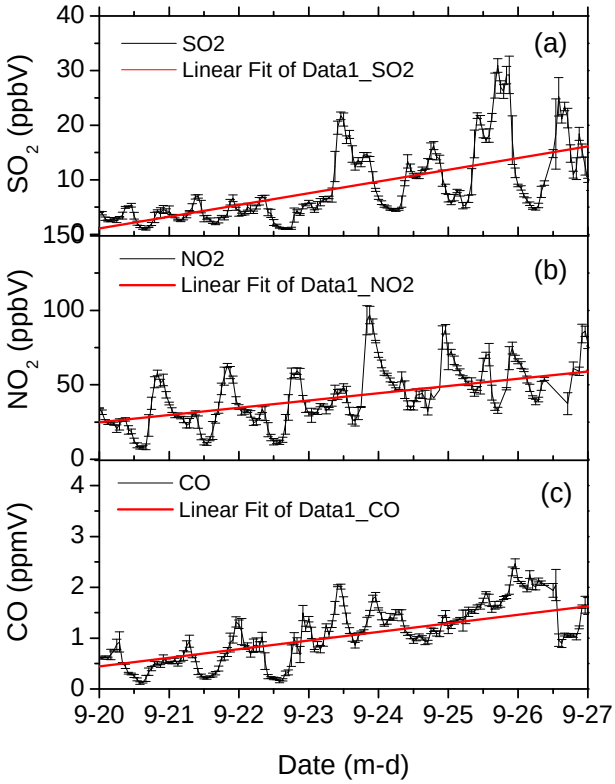

Figure 2. The temporal trend of the mixing ratio of (a) SO_2, (b) NO_2, and (c) CO from September 20 to September 27, 2011.

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5. P16269, L7, "In addition, the air temperature sharply decreased on 24 September, and the decreasing trend remained till 26 September, which could have caused the lack of the thermal dynamics for the development of the PBL"

The temperature on September 25-26 is 1-2 degrees lower than on September 20-23, so the sharp temperature decrease did not last till 26 September (Fig. 6).

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Response:

We deleted the word "sharply" in the sentence in the revised manuscript.

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6. P16269, L9, "The RH increased from 24 September, which could have resulted in the hygroscopic growth of aerosol scattering."

The increase of RH from 24 September is not obvious (Fig. 6), and there is no relation of RH and visibility or light scattering.

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Response:

The temporal trend of the RH from September 20 to September 27 was illustrated by the following figure. The red line was the linear fit of the RH and the overall trend of RH from September 20 to September 27 was increasing. From figure 6, the RH on September 22-23 was 60% with minimum of 10%, but, from September 24, the RH increased with peak 75-80% and minimum of 30-40%.

We rectified this sentence in the revised manuscript with "The RH increased from 24 September, which resulted in the hygroscopic growth of aerosol scattering."

The relationship among RH, visibility, and $PM_{2.5}$ was illustrated in figure 12 in the revised manuscript.

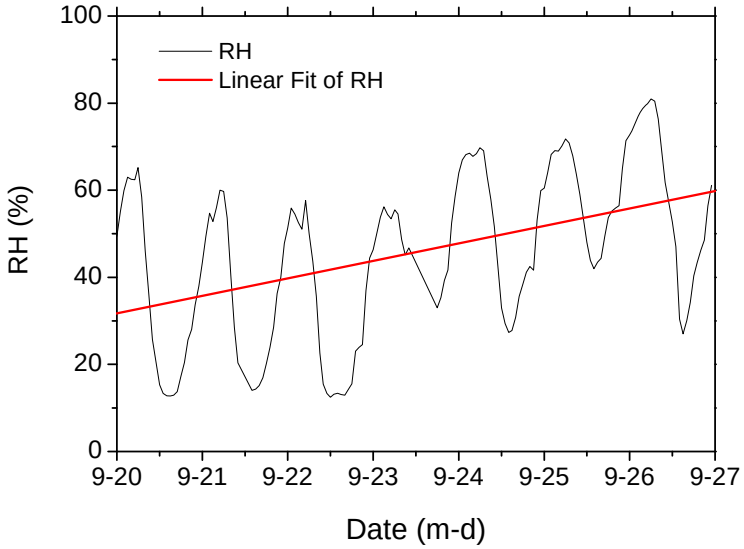

Figure 3. The temporal trend of the RH from September 20 to September 27, 2011.

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7. P16269, L2, "low wind speed provided a longer contact time among aerosols and trace gases, and could possibly enhance the formations of new particles"

Low wind speed favors accumulation of aerosols and hence aerosol surface area increases under low wind speed conditions, which could inhibit new particle

formation. This can be seen from Fig. 6 and Fig. 9: When wind speed is low during September 23-27, new particle formation did not occur. How does low wind speed enhance new particle formation?

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Response:

This sentence "low wind speed provided a longer contact time among aerosols and trace gases, and could possibly enhance the formations of new particles" was cited from Wang et al. (2006). We thought over the reviewer's comment and re-analyze the data of this study, we recognized that this statement may be unsuitable for our research and we deleted this sentence in the revised manuscript.

References:

Wang, Y., Zhuang, G. S., Sun, Y. L., and An, Z. S.: The variation of characteristics and formation mechanisms of aerosols in dust, haze, and clear days in Beijing. Atmos. Environ., 40, 6579-6591, 2006.

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8. P16271, P12, "High temperature and high humidity promoted the photochemical formation of particulate matters."

I don't think high humidity promotes photochemical formation of particles. What is the basis of this argument?

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Response:

We thought over about this question. This sentence in the original manuscript was redundant, furthermore, this statement contradicted with the sentence "At the noon time with sunny and dry conditions, the photochemical reaction was generally vigorous and new particle formed (Wu et al., 2007)" at line 19, P16271. So, we deleted this sentence "High temperature and high humidity promoted the photochemical formation of particulate matters." in the revised manuscript.

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9. P16273, Section 3.2.6, this section describes the relation of visibility and PM2.5, which is not related to the "formation and evolution mechanisms of regional haze " of Section 3. So Section 3.2.6 could be Section 4.

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Response:

We accepted this suggestion and modified the section 3.2.6. In the revised manuscript, research on the relation of visibility, RH, and $PM_{2.5}$ was merged with section 3.2.5 "Impact of hygroscopic growth for aerosol scattering f(RH)".

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10. P16275, L1, "At the beginning, plenty of sunlight and higher temperature provided favorable conditions for photochemical reactions and generated more ozone and fine particles."

The temperature is not significantly higher at the beginning of the measurement (Fig. 6) and less ozone (not more ozone) was formed at the beginning (Fig. 1).

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Response:

We recognized that this statement is incorrect and this sentence "At the beginning, plenty of sunlight and higher temperature provided favorable conditions for photochemical reactions and generated more ozone and fine particles." was deleted in the revised manuscript.

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Technical corrections

1. The APS data are not shown in the text or in the figures, so APS should be removed from the experimental section and Table 1.

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Response:

Section 3.2.4 "New particle formation" was modified with name "Gas to particle conversion and aerosol growth" in the revised manuscript. We add the analysis of aerosol growth from nucleation mode (3-25nm) to Aitken mode (25-100nm), accumulation mode (0.1-1μm), and coarse mode (>1μm). So, the instrument APS was not deleted in Table 1.

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2. The figure legends in Figure 4 and Figure 7 are too small and hard to read.

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Response:

The figure legends in figure 4 and figure 7 were redrawn for clarity in the revised manuscript.

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3. Figure 7, add (a), (b): : : to the figure legends.

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Response:

We added legends (a), (b), (c), and (d) to the figure 4 and figure 7 in the revised manuscript.

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4. P16260, L8, remove "(RH)" in the abstract since it is defined at L26 on this page.

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Response:

Thanks and we revised accordingly in the revised manuscript.

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5. P16261, L17, change "frequently has occurred" to "has frequently occurred."

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Response:

Thanks and we revised accordingly in the revised manuscript.

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6. P16262, L3, change "yr" to "years."

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Response:

Thanks and we revised accordingly in the revised manuscript.

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7. P16262, L4, change “extremely haze” to “extreme haze.”

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Response:

Thanks and we revised accordingly in the revised manuscript.

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8. P16262, L6, “Haze episodes have been now characterized by increasing frequencies, longer duration and expanding sphere.” Are there references for this sentence?

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Response:

We added references to this sentence “Haze episodes have been now characterized by increasing frequencies, longer duration and expanding sphere (Wu et al., 2005; Zhang et al., 2008; Kang et al., 2012).”

References:

Kang, H., Zhu, B., Su, J., Wang, H., Zhang, Q., Wang, F.: Analysis of a long-lasting haze episode in Nanjing, China. Atmos. Res., http://dx.doi.org/10.1016/j.atmosres.2012.08.004, 2012.

Wu, D., Tie, X., Li, C., Ying, Z., Lau, A. K.-H., Huang, J., Deng, X., and Bi, X.: An extremely low visibility event over the Guangzhou region: a case study. Atmos. Environ., 39(35), 6568-6577, 2005.

Zhang, Y. H., Hu, M., Zhong, L. J., Wiedensohler, A., Liu, S. C., Andreae, M. O., Wang, W., and Fan, S. J.: Regional integrated experiments on air quality over Pearl River Delta 2004 (PRIDE-PRD2004): Overview, Atmos. Environ., 42, 6157-6173, 2008.

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9. Section 2.2 re-order the description of the measurements in this section or in Table 1 to make them in the same order for smooth reading.

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Response:

Thanks and we accepted this suggestion. In the revised manuscript, the order of the instruments in table 1 was re-ordered by the description of the measurements in section 2.2.

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10. P16266, equation (1), define "*Vis*" in the equation as visibility here rather than on P16268.

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Response:

We accepted this suggestion and rectified the manuscript accordingly.

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11. P16266, "In this study, we used the coefficient of variance of LIDAR signals calculated by Eq. (2) to detect the height of the PBL." This sentence is not very clear. Also, are there references for this method?

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Response:

In the revised manuscript, we described the method in detail and tried our best to clearly articulate the retrieval method of the height of the PBL. We also cited some literatures in the revised manuscript.

The top of the PBL has been given many names; the most popular being inversion height, mixing height and mixed-layer depth. The commonality exists in that they all refer to the greatest depth to which atmospheric constituents are well mixed.

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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12. P16269, L10, “which could have resulted in the hygroscopic growth of aerosol scattering.” The “hygroscopic growth of aerosol scattering” in this sentence is not clear.

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Response:

We accepted this suggestion and rectified the sentence with “The RH increased from September 24, which resulted in the hygroscopic growth of aerosol scattering (discussed in section 3.2.5)” 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!!!!