Point-to-Point Response to Comments and Questions

on MS No.: acp-2010-965 (Elevated aerosol layer embedded in aged soot particles in a polluted urban atmosphere by G. Shi et al.)

Anonymous Referee #2 (16 March 2011)

General comments:

This manuscript presented a special vertical profile of aerosol particles - layered structure in the Megacity Beijing. Several methods (OPC, TEM and SEM, etc) were implemented in the analysis. The author focused on the description of soot particles, its coating process and interaction with the boundary layer.
The soot particles are undoubtedly important considering its role in the climate change. Though several ground-based measurements of soot particles had been reported in the same area, there was no study addressing its vertical structure with comprehensive analytical methods. Therefore, this study should attract many readership in the aerosol sciences, and is suitable for publication in ACP.
I recommend publication after the authors have considered the comments listed below:

(1) The description about experiments and discussions seems too brief to me, especially how the author defined the "soot particles", and how they did the statistic analysis on TEM results. It would be more precise and less misleading to say "particles with soot inclusions or soot cores" instead of "soot particles".

***Response*:**
The details of particles which were characterized as soot particles and secondary particles (the two groups of most frequently detected particles in the samples) are described in section 3.2 referencing to the pictures shown in Fig. 3. We did the statistic analysis after measuring the shape and size of every particle which we could see in the photos. This is mentioned in line 20 page 1647 of the discussion version. Information on the identification of fly ash and road dust is added in the end of that part, where the two kinds of particles were mentioned (Line 18 page 1650 of the discussion version).
We prefer to use “fresh”, “young” and “aged” to soot particles as mentioned in the manuscript. We are afraid that using "particles with soot inclusions or soot cores" to replace "soot particles" will result in confusion. A major reason is that “soot particles” show too many different shapes under electron microscopes. If we use for example “particles with soot inclusions”, readers may naturally consider “particles without soot inclusions” rather than other kinds of soot particles.

(2) The absorption effect of soot particles were claimed to be the explanation for the inversion and decrease of lapse rate. However, during the observation period, an anticyclone was sitting over a large area, as also mentioned by the authors in the manuscript. The anticyclonic high-pressure systems often lead to a subsidence inversion even without the presence of soot particles. By a glance of the back trajectories, one could also find this period was characterized with a strong subsiding air mass. I suggest the authors to discuss about these absorption effects if they could provide more solid and comprehensive proofs.

***Response*:**

Yes, subsidence can lead to inversion. But this was not the case of the elevated layer we encountered in this study. The reason is that adiabatic subsiding must result in a decrease of RH with the decrease of the altitude and the virtual potential temperature and mixing ratio of the upper part of the EAL should have been approximately similar to that of the upper later. In the present case, RH and mixing ratio in the inversion layer was larger than that in upper layers and the virtual potential temperature gradually increased with height from the upper part of the EAL. In the revision, "The fact that RH and mixing ratio in the EAL were larger than that in the upper layers and the virtual potential temperature gradually increased with altitude from the upper part of the EAL (Fig.1 and Fig.6) indicates that the inversion was not caused by subsidence within the anticyclone." is added in the discussion. Fig.3S is a figure in supplementary figure showing the time series of vertical profiles of downward wind in the revision.

(3) The key issue about EAL is why its aerosol concentration was remarkably larger than those in the lower and upper layers. Though there is a section called "formation of the EAL", this issue seems to be not specifically and fully discussed.

In all, I think the authors could think about strengthening the aerosol and soot characterization parts, giving more component information from TEM and SEM, and making Sect 4.1 more concise.

***Response*:**

In order to show why the concentration of aerosol particles in the EAL was higher than other layers, in the revision, the following descriptions are added into the last paragraph of section 4.1. "Air quality data published by the Beijing Environment Protection Bureau showed that the urban areas of Beijing experienced a heavy pollution episode from December 6 to 7 (maximum concentration of inhalable particulate matter ~ 250 $\mu g\ m^{-3}$) and then the pollution became weaker (~ 150 $\mu g\ m^{-3}$). The air pollution in the nocturnal layer was severe and the upward mixing in the morning diluted the pollutants, which is likely the reason that aerosol concentration in the EAL was higher than in both upper and lower layers."

Specific comments:

1. Abstract: "The lapse rate in the lower part of the EAL had an obvious decrease", "in which the aerosols, as a feedback, enhanced the stability of the layer by absorbing solar radiation." As in the general comments.

***Response*:**

Refer the response to the general comments.

2. Introduction:
Several studies on soot and its optical properties in Beijing should also be mentioned, e.g. (Wehner et al., 2009) et al.

***Response*:**
The following brief descriptions on the soot particles observed on the ground are added to the beginning of the last paragraph of the introduction. "Air pollution caused by soot particles from anthropogenic activities in Beijing, the capital of China, is a public and scientific concern because of the effects of the particles on air quality and regional climate change. Numerous studies with observations on the ground revealed that the particles were from biomass burning, raw coal combustion, diesel vehicle emission and open fire (e.g. Liu and Shao, 2007). Aging of fresh soot particles likely proceeded very fast in the urban atmosphere and resulted in the change of particles from open spherule's chains or agglomerates to shrunk and compact clusters coated with secondary species (Niu et al., 2011). As a consequence, the size of the particles was frequently in the range of 1.0 ~ 2.5 μm and many particles were present as a mixture with other materials and had largely changed their physical and chemical properties in comparison with fresh ones (e.g. Li and Shao, 2010; Wehner et al., 2009)."

3. p1645, l15: "Several observations on the vertical distribution and the structure of the boundary layer were carried out in Beijing, the capital city of China, with tethered balloon flights."
It is a pity that only one day data were presented in this paper making it difficult to judge if the elevated layer is representative or not. I am not sure if "the several observations" here means there are additional measurements on other days.

***Response*:**
Yes, we did the observation on many days from 2002 to 2009. Unfortunately, the case we show in the paper is the only case we got sets of samples successfully in different layers. The reason is the technical difficulty of the observation. Before the observations described in this study, we did not have the assistant of the wind profiler. So we could not pre-design the particle collection altitudes in those observations. Although in two sets we found elevated layers in afterward analyses, we did not collect particles at the altitudes because we did not know there was a different layer. After the observation mentioned in this paper, we did some observations trying to get more data in similar cases. Unfortunately, we have not got another successful case.

4. p1646, l6: "At the site, wind profiles were monitored with a wind profiling system (CFL-16, China Aerospace Science & Industry Corp.), which provided the horizontal and vertical wind from 150m to about 6000m with a vertical resolution of 150m every 6 min."
I suggested showing the wind profiles explicitly either in the manuscript or supplement

***Response*:**
We mentioned it in the manuscript because it was impossible for us to have got the samples exactly in the EAL without the data. Adding these data will make the manuscript tedious. So we add them as supplementary figures (Figs 1S-3S) in the revision. They are also attached in the end of this reply.

5. p1656, l20: “: : : could not be simply attributed to the formation of new particles in accumulation mode” This is misleading because the new particle formation never happens in accumulation mode.

***Response*:**
It is changed into “: : : could not be simply attributed to secondary particles in accumulation mode”.

6. p1656, l24: “Aging had caused the original soot particles to shrink into aggregates and their size shifted to smaller range : : :“
Maybe cite the paper of (Zhang et al., 2008), in which the shrinking was also addressed.

***Response*:**
It is on the page p1652 of the discussion version if we are correct. The mentioned reference is added there in the revision.

Reference:
Wehner, B., Berghof, M., Cheng, Y. F., Achtert, P., Birmili,W., Nowak, A., Wiedensohler, A., Garland, R. M., Pöschl, U., Hu, M., and Zhu, T.: Mixing state of nonvolatile aerosol particle fractions and comparison with light absorption in the polluted Beijing region, J. Geophys. Res., 114, 10.1029/2008jd010923, 2009.
Zhang, R., Khalizov, A. F., Pagels, J., Zhang, D., Xue, H., and McMurry, P. H.: Variability in morphology, hygroscopicity, and optical properties of soot aerosols during atmospheric processing, Proceedings of the National Academy of Sciences, 105, 10291-10296, 10.1073/pnas.0804860105, 2008.

p.s. According to the request of Dr. Guangyu Shi, the order of the first two authors is exchanged in the revision.

Thank you very much for your helpful comments and questions.

Supplementary data for

Elevated aerosol layer embedded in aged soot particles in a polluted urban atmosphere

by Daizhou Zhang, Guangyu Shi, Biao Wang, Bin Chen, Maromu Yamada, and Hongya Niu

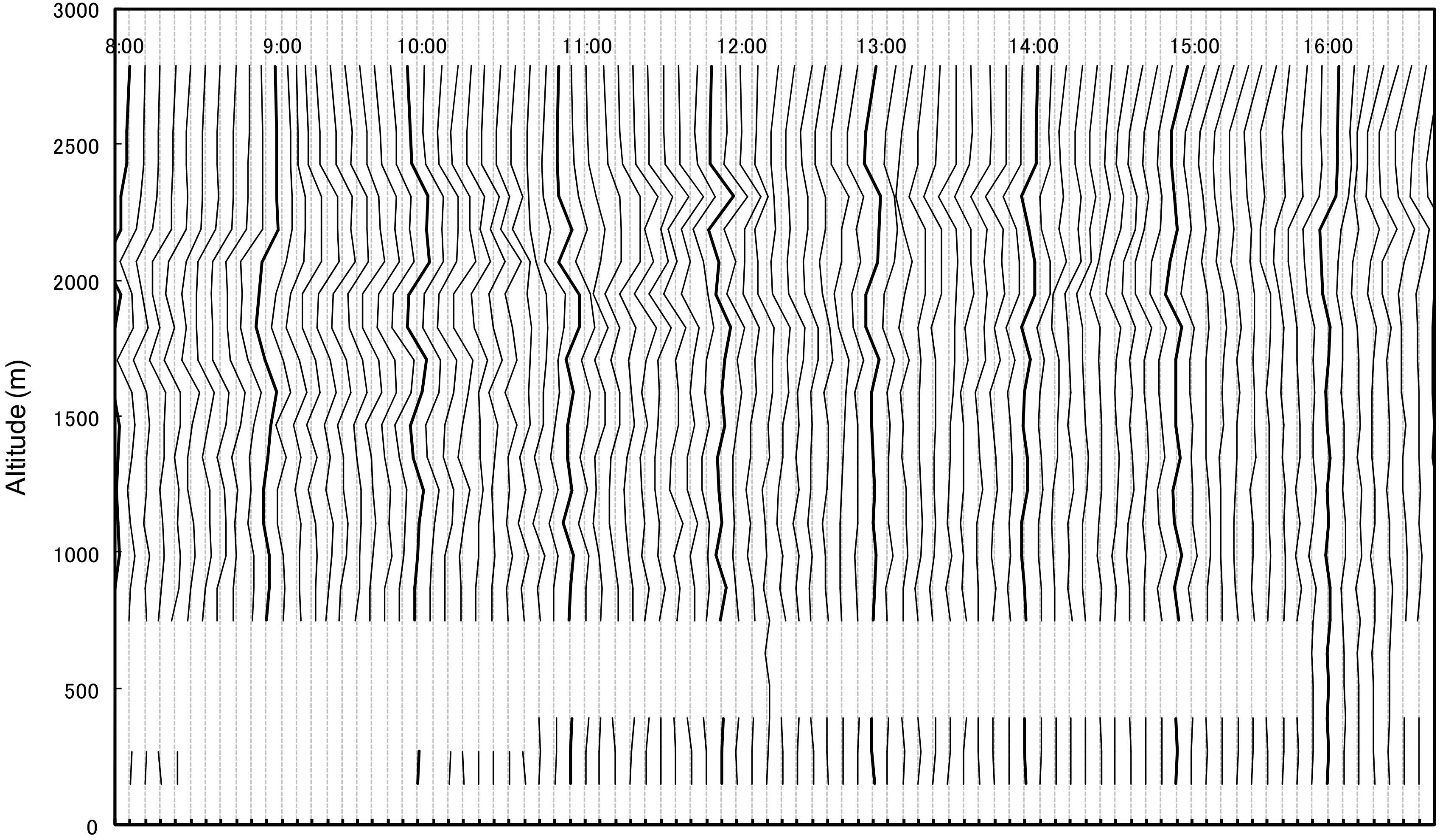

Fig. 1S Profiles of eastward wind at the site every 6 minutes on the observation day. Increments show the scale of 10 m s^{-1}.

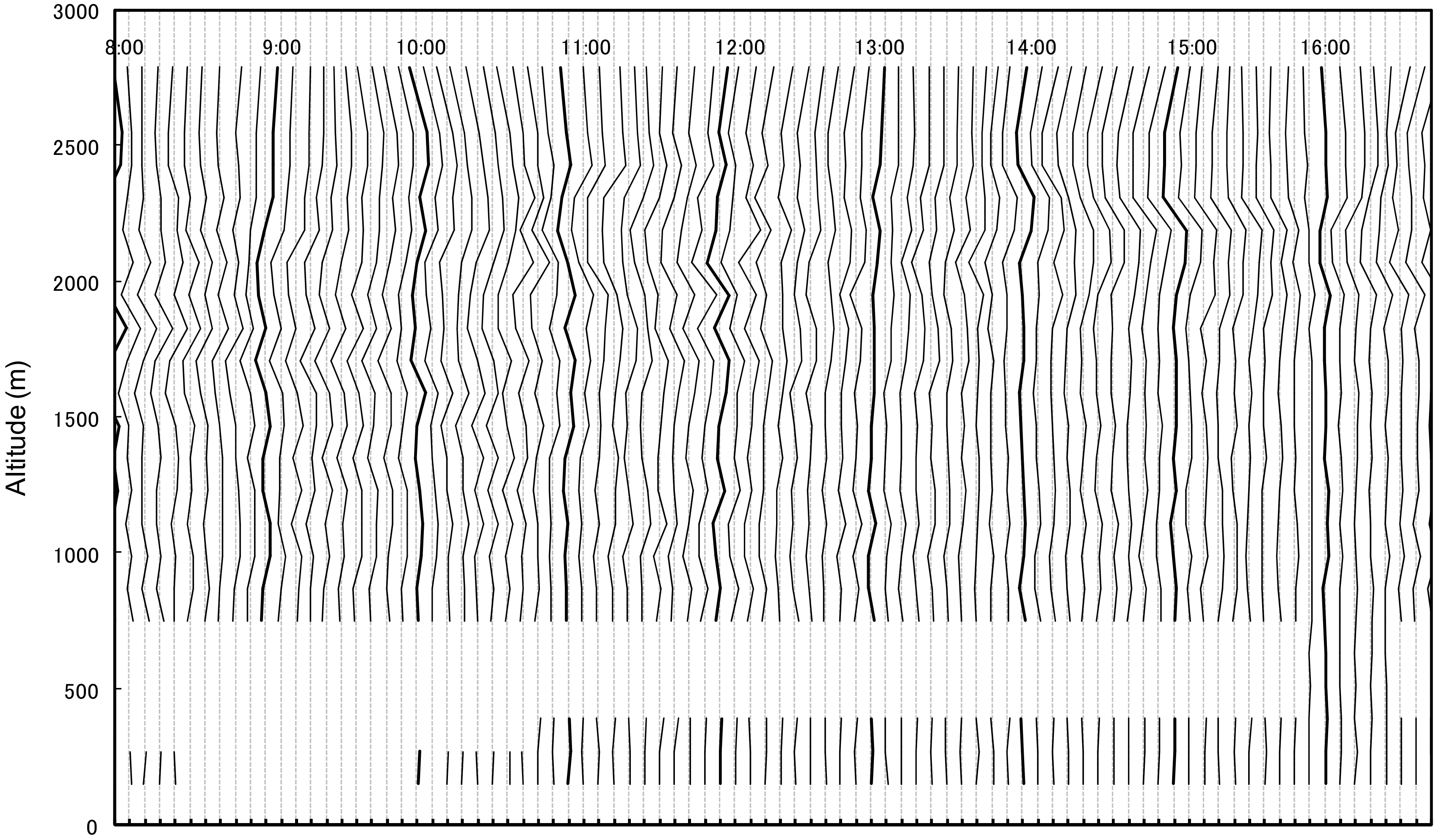

Fig. 2S Similar to Fig. 1S but the profiles of northward wind.

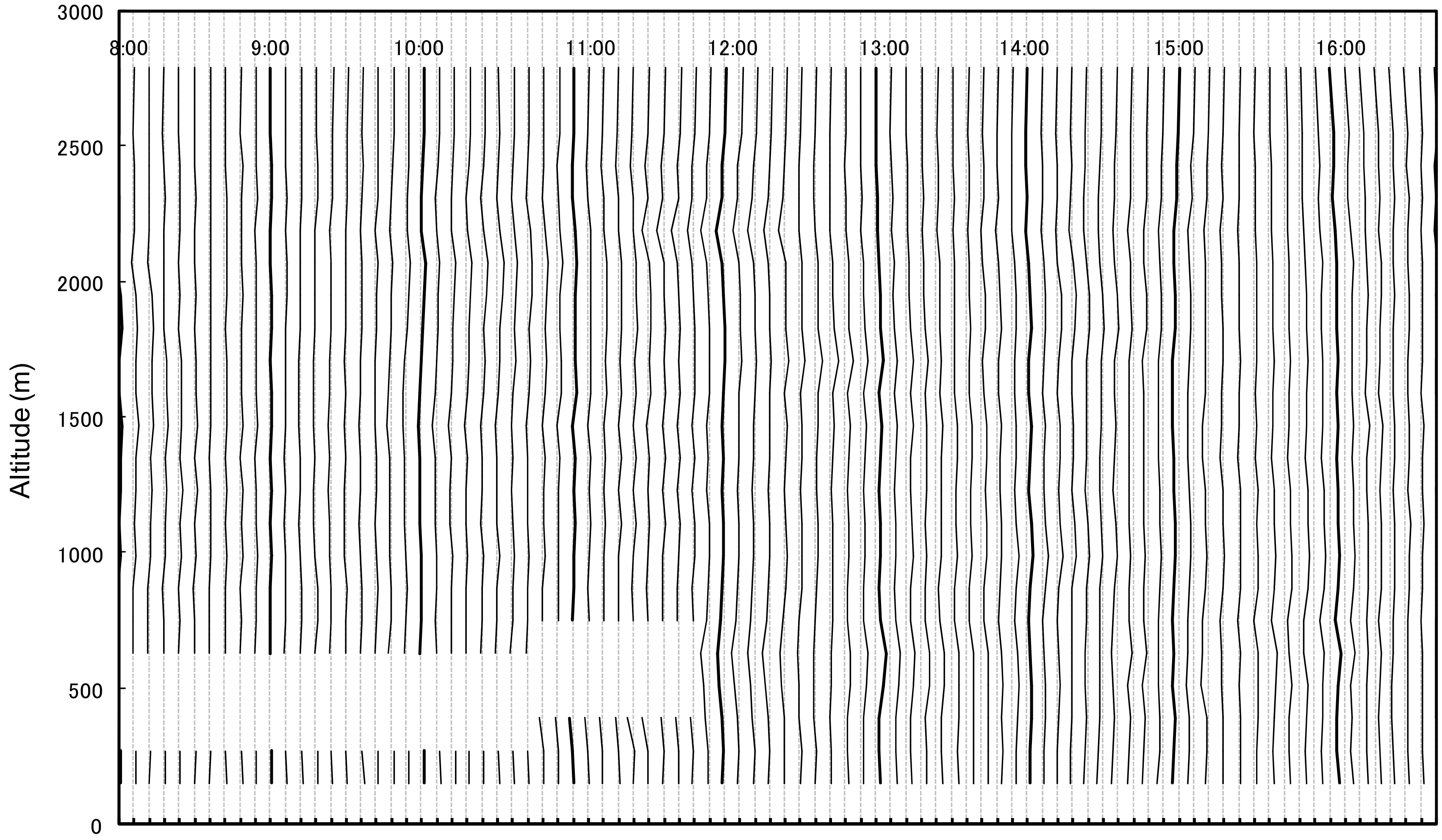

Fig. 3S Similar to Fig.1S but the profiles of downward wind and the increment scale of 0.5 m s[-1].