Interactive comment on “The measurement of aerosol optical properties at a rural site in Northern China” by P. Yan et al.

P. Yan et al.

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We would like to thank the two reviewers for their time in reading our manuscript and correcting the errors, and their valuable comments. They are indeed very helpful to improve the quality of the manuscript.

Reply to Anonymous referee #1

(1) Page 13005 on instrumentation: Detailed description of the sampling system should be given as such information is important for determining any particle loss during sampling. In particular the material type, length and diameter of the sampling line should be given. Heating inside the nephelometer is expected to cause loss of not only ammonium nitrate but also semi-volatile organic aerosols. The later tend to have larger contribution to scattering compared to nitrate aerosol.
The description of the sampling system has been added in the revised manuscript, and the statement on the heating loss of aerosol has been changed.

(2) Section 3.1 on comparison with other studies: As different instruments/method may give different readings for BC, it is important to keep this in mind when comparing the data from different investigators.

In the revised manuscript, we have added a column to the Table 3, and listed the instruments used by the different investigators.

(3) Section 3.2 on seasonal variation: Different seasonal pattern in southern China (Cape D Aguilar) (i.e. winter-high and summer-low) is mainly due to large scale changes in transport of pollution. In winter northerly winds brings polluted continental air masses while southern flows in summer contained clean oceanic air masses from the tropics.

In the revised manuscript, we added some explanations to the statement.

(4) Section 3.3 on diurnal variation: the author examined the diurnal pattern using yearly average data. It would be better to examine the pattern in different seasons. Northern Beijing can experience upslope and down-slope flow during summer time, thus it is possible that if the summertime data are examined, higher values of scattering and absorption can occur during afternoon not at nighttime. Such a feature has been observed for ozone, SO2 and NOx (Wang et al., GEOPHYSICAL RESEARCH LETTERS, VOL. 33, L21806, doi:10.1029/2006GL027689, 2006.)

We have analyzed the diurnal variations for each season, the results are quite similar to those calculated using the yearly average data. So we do not show the diurnal pattern for each season in the revised manuscript, but we add some discussions to explain the reasons. Although the SDZ site can experience upslope and down-slope flow during summer time, the diurnal variations for aerosol absorption or scattering coefficient does not show higher values during afternoon as reported by Wang (Wang et al., GEO-
PHYSICAL RESEARCH LETTERS, VOL. 33, L21806, doi:10.1029/2006GL027689, 2006). In Wang’s work, the site is relatively close to the urban area (about 50 km away from the center of Beijing city), the terrain induced circulation could have significant impact on the transport of the pollutants to the site, however, SDZ site is farther away from the urban area (about 150 km northeast to the Beijing urban area), There are only limited anthropogenic sources within a 30 km range. The lack of enhancement of aerosol extinction in summer afternoon at SDZ, suggests that upslope flow did not transport urban sources to SDZ.

(5) Section 3.4: the description on cluster analysis result can be shortened. Table 8 shows that the carbonaceous mass fraction in PM2.1 was low (9-13% after multiplying a factor of 1.6 to OC) in the two samples collected on polluted days. Urban Beijing has much larger mass fraction of carbonaceous materials. Were the two samples collected on foggy days?

We have condensed the first part on cluster analysis by removing the paragraph on the description of the method, and also we have added a brief discussion on the measured fraction of OC, EC and other chemical compositions.

As the referee pointed out, the fractions of OC in the two samples were lower than those measured in Beijing urban areas, however, the concentration levels were still comparable to the measurements made in Beijing urban regions, for example, Yu et al.(Yu J., et al.: Characteristics of carbonaceous particles in Beijing during winter and summer 2003. Advances in Atmospheric Sciences, 23(3), 468-473, 2006.) reported the OC and EC concentration were 9.4±2.1μg/m3 and 4.3±3.0μg/m3, respectively, for the summer campaign in Beijing in 2003; He et al.(He K.B., et al.: The characteristics of PM2.5 in Beijing, China. Atmos. Environ., 35, 4959-4970, 2001) reported the average OC and EC concentrations were 13.42μg/m3 and 6.27μg/m3 at Beijing urban site in the summer of 1999, respectively. The low fraction of OC in the two samples collected under polluted conditions presented in our work was mainly due to the high concentration of sulfate and other compositions in the PM2.1 particles.
1. (In the Abstract and in various sections of the paper) A crucially important issue to address in this paper: Why are more polluted areas or sources giving rise to higher values of aerosol single scattering albedo [SSA]? A more detailed explanation is required. The results from this work would appear to contradict findings from previous work of several authors. Secondary aerosol formation is alluded to as a possible explanation of higher SSA values, but reason(s) for such is (are) not given. In addition, evidence for secondary aerosol formation is not provided. Could it be that the nephelometer is reading too high an aerosol scattering coefficient value?

For the Nephelometer measurement, during the experiment, we applied the strict quality control procedure (SOP) to the operation of the instrument. That is, zero check was done automatically by pumping in particle-free air once each day, and weekly span check was performed manually by the operator using pure HFC-134a gas. The results of the zero/span check indicated that the bias for zero check was less than 2 Mm⁻¹, and the bias for span check was less than 5%. There is no clear evidence indicating that the nephelometer is reading too high an aerosol scattering coefficient value. For the moment, the exact reason for the air masses coming from more polluted areas or sources giving rise to higher values of aerosol single scattering albedo (SSA) is not totally elucidated in this paper, however, there are some indirect information or evidences that support our measurement and give some hints to understand our results:

(1) The emission inventory for OC and BC reported by Cao et al. (Cao, G., Zhang, X. and Zheng, F.: Inventory of black carbon and organic carbon emissions from China Atmospheric Environment 40(34), 6516-6527, 2006) showed that, in the northern sector of our site, such as Inner Mongolia (or Nei mongol), the dominant emission sources for BC and OC are from emissions of residential coal combustions. They are different
from the emissions in more polluted regions in the southern sector of our site (more industrial emissions). The combustion process of coal at a low temperature (residential emissions) could release more BC than the industrial sources, and might lead to more light absorption of aerosol when air masses come from northern regions of the site. (2) Limited analyses of aerosol chemical composition indicated that more sulfate, ammonium and nitrate aerosols presented when the samples were collected under polluted conditions. This implies that aerosols under polluted conditions can be more scattering, although the relationship between the single scattering albedo (SSA) and the aerosol chemical compositions are much more complicated.

In the revised manuscript, we added some discussions on aerosol chemical compositions (including OC) and their effect on aerosol optical properties despite the insufficient measurement on this issue.

2. Explain why BC aerosol is of particular importance with respect to the hydrological cycle (Introduction, line (L) 8).

In the revised manuscript, we added some explanation about why BC aerosol is important to the hydrological cycle.

3. Explain the basis for the wavelength correction for aerosol absorption coefficient (page 7), and give details of how it is applied.

In the revised manuscript, we added the formula and its application of the light absorption coefficient wavelength correction.

4. "scattering coefficients are more variable than absorption coefficients, which reflect the different sources and formation processes in these regions" [p 7]: Surely will not the sources be the same? "More variable scattering" needs to be clarified and a better explanation is required to explain the differences between scattering and absorption coefficient.

In the revised manuscript, this description was removed, since it was not very relevant.
5. One could question the appropriateness of presenting seasonal characteristics of aerosol parameters based on just 5/6 full seasons of data. Due comment should be made on this in view of likely variability of seasonal characteristics from one year to another.

In the revised manuscript, we just select one year data with four complete seasons and confined only to this specific time period to discuss the seasonal variations and the factors affecting the variations. Our focus is to explain the influencing factors for the seasonal variations, rather than to present a representative seasonal climatology of the site. To give more representative seasonal distributions of aerosol at the site, long-term measurements are needed.

6. Statement (p 8) that for the spring period there is enhancement of absorption and scattering is not the case. Greater values prevail for the summer season (Table 4) and for the fall season in the case of aerosol scattering. This statement needs to be modified and clarified.

Revised

7. Section 3.2, p 9, L 11: "heterogenic"? or is it meant to be "heterogeneous"; if so, how do you know that heterogeneous aerosol production is occurring during transport?

"Heterogenic" was corrected to "heterogeneous".;. The expression was changed to "Under the fog/mist weather conditions the growth and/or heterogeneous production of aerosol was favored and could significantly enhance the aerosol scattering coefficient."

8. 3.3 Explanation/clarification needed for why does turbulent dilution give rise to the highest values of absorption coefficient at night ? if this is the reason, why is it that highest values of aerosol scattering coefficient do not also occur over the same period?

Revised. The new explanation is presented in the revised manuscript. The explanation for the diurnal pattern of the measurement is due to the evolution of the planetary boundary layer, not just the turbulent dilution.
9. The winter season seems to me [I may have mis-interpreted their choice] to have been chosen from months January, February of 2004 and from December 2004. Surely should not the 3 contiguous months have been chosen: that of December 2003, and January & February 2004? The authors apparent choice of months January, February of 2004 and from December 2004, may well lead to different results from that of the more orthodox sequence. If this is the case, at the very least additional analysis should be undertaken to compare results from the 2 different 3-month combinations.

Revised. We re-calculated the trajectories and made the clustering analysis for the new winter period (from Dec. 2003 to Feb. 2004). The conclusion is similar with that for period from months January, February of 2004 and December 2004, although the mean trajectory of each cluster is not the same.

10. No account is given of the filter sampling results for the winter period [Feb 2 - 20]. I am curious why reference to aerosol filter sampling results for this period is not made in the paper?

In the revised manuscript, the description about the filter sampling for the winter period was removed, because the chemical composition data in the winter period sampling analysis need further validation.

11. No serious reference is made in the paper to organic carbonaceous (OC) aerosol, and what effect OC could have on SSA for example. Lack of measurement of OC could be construed as a relatively major gap in this work. Comment on OC should be included at least in the Conclusions section of the paper.

In the revised manuscript, we added some discussions on the OC and its effect on the aerosol optical properties. OC is another important contributor to the light scattering of aerosol, but our filtering sampling analysis revealed that OC accounted for only a small fraction of the fine particles for the summer samples (as presented in Table 8 of the manuscript). The effects of OC on our measured SSA during the summer period could be expected to be small.
12. Typographical errors or other amendments/queries
In the revised manuscript, necessary amendments are made according to the referee’s comments and corrections.