

Thanks for your comments. Our responses for the general comments are provided below:

(1) On the boundary layer height in relation to conversion of column mass BC to surface concentration.

In the paper, our comparison focus mainly on the time variation trend of remotely sensed column BC with in situ BC (the correlation coefficient is about 0.77 for the studies samples). Moreover, if one wants to convert column BC to surface BC concentration, we propose to assume a uniformed BC layer (UBCL) distributed vertically homogenously from surface to a certain height. Conversely, if it is in a unit of kilometer, this UBCL height = $BC(\text{Remote Sensing}) / BC(\text{in situ at the ground})$, which is confirmed by their units (RS in mg/m^2 and in situ in $\mu\text{g}/\text{m}^3$). Lack of explanation on the difference between UBCL height and the planetary boundary layer height (PBLH) might be the reason causing misleading of this part. So we changed texts in the revised paper:

(i) Replace “we can estimate that the boundary layer height is about 1 km when comparing surface and column BC concentration” with “we can estimate that the assumed BC column height is about 1 km when comparing surface and column BC concentration”.

(ii) Supplement “Considering in the case of heavy haze, most of aerosols can be constrained somewhat uniformly near the surface (Lv et al., 2013), the planetary boundary layer height (PBLH) can be treated as a proxy of the BC column Height roughly”.

Moreover, Zhang et al. (2009) observed PBLH from radiosonde measurements (related texts in the paper are changed following reviewer’s opinion) and their results (Beijing winter haze concentrates somewhat uniformly below 1 km) are confirmed by some recent Lidar observation at Beijing. One example is provided below:

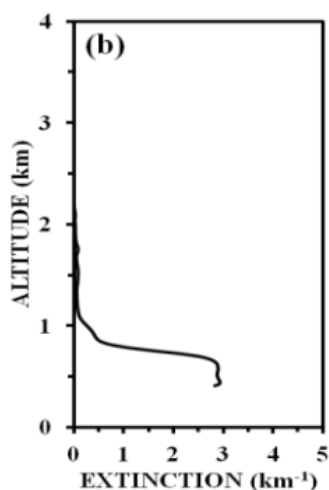


Fig S1. Beijing winter haze aerosol extinction profile observed by Lidar at 532nm on 14 Jan, 2013 (Lv et al., Joint use of ground-based lidar and sun-sky radiometer for observation of aerosol vertical distribution, *J. Rem. Sen.*, Accepted, 2013).

(2) On the data representativeness.

This work is a case study, so we changed possible misleading expression (e.g. deleting words like “model”) in the paper. Also, the chemical component retrieval approach is a very new technique that will be tested firstly by case studies before applying to long-term historical data. Here we want to show Beijing 2013 haze results (averaged from 1 to 31 Jan, 2013) in Figure S2 and Table S1. By comparing haze results with that of Figure 10 and Table 1 in the paper, we can find general agreements considering the standard deviation presented in Table 1. The differences in DU and AS can be explained by relatively less influences of dust or fly ashes in Beijing Jan 2013 and partly uncertainties in the DU and AS fraction retrieval (please see the supplemented error assessment section in the paper), which will be investigated carefully in the next works.

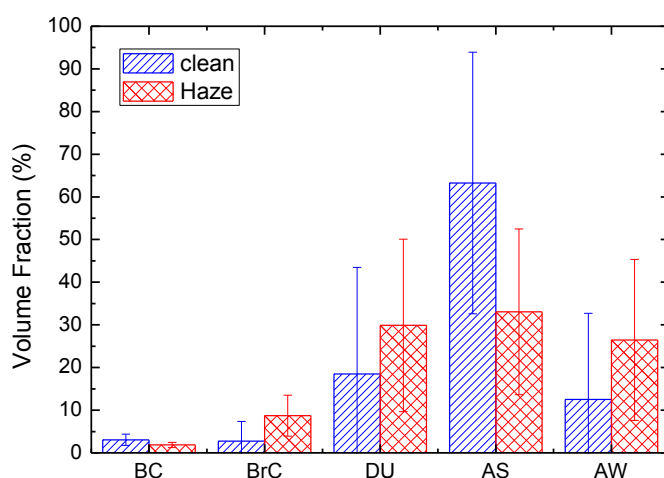


Fig S2. Volume fractions of aerosol components retrieved from sun-sky radiometer measurements at Beijing under clean (5 days) and haze (9 days) conditions in Jan 2013.

	Volume fraction (%)				
	BC	BrC	DU	AS	AW
clean	3 (±1)	3 (±5)	18 (±25)	63 (±32)	13 (±20)
Haze	2 (±1)	9 (±5)	30 (±20)	33 (±19)	26 (±19)

Table S1. Volume fraction numbers (and standard deviations) of aerosol components retrieved from sun-sky radiometer measurements at Beijing in Jan 2013.

(3) On the relationship between PM_{2.5} and AOD in the haze condition.

We agree the reviewer’s opinion on the representativeness of our PM_{2.5} and AOD regression equation, i.e. it is only valid for cases studied in this work. So we deleted the equation of the PM_{2.5} and AOD in the paper to avoid possible misunderstanding and misuse. However, we want to mention that even the problem on the representativeness of linear equations, the linear regression is still the most popular way (e.g. Wang and Christopher, 2003; Engel-Cox et al., 2004; Koelemeijer et al., 2006; Liu et al., 2007; Gupta et al., 2008; Schaap et al., 2009; Zheng et al., 2011)

to illustrate PM_x and AOD relationship considering it's merit of simplicity. We added also more discussion in the paper on this issue:

“Moreover, it is well accepted that aerosol vertical distribution and ambient humidity corrections can improve the correlation between AOD and PM_{2.5} (e.g. Kotchenruther et al., 1999; Tsai et al., 2011). In this meaning, the observation of AOD from ground, simultaneously with PM_{2.5}, aerosol profile and other environmental factors (e.g. humidity) is very useful to study on the AOD-PM_{2.5} conversion which can be used in remote sensing of PM_{2.5} by satellites from space.”

Additionally, we want to provide more details to reviewer's comment on “800% uncertainty in 2013 event”. Firstly, we want to mention that up to now the PM_{2.5} and AOD sites of Beijing city are not located in the same place. The AOD number mentioned by the reviewer should correspond to IAP or our AOD sites, both closely located in the North part of the city. Meanwhile, the PM_{2.5} sites with very high values from 10 to 12 Jan 2013 are mainly located in the South part of the city (e.g. YiZhuang PM_{2.5} site). The significant difference of haze spatial distribution in the 2013 event is clearly illustrated by the high spatial resolution satellite images shown below. Therefore, if the AOD and PM_{2.5} data are not related to similar haze pollution conditions, they should not be compared directly. It is the case of the 2013 haze event and not the cases of 2011 and 2012 haze events in our paper (example on 27 Feb 2012 is shown below), which could be checked by satellite images.

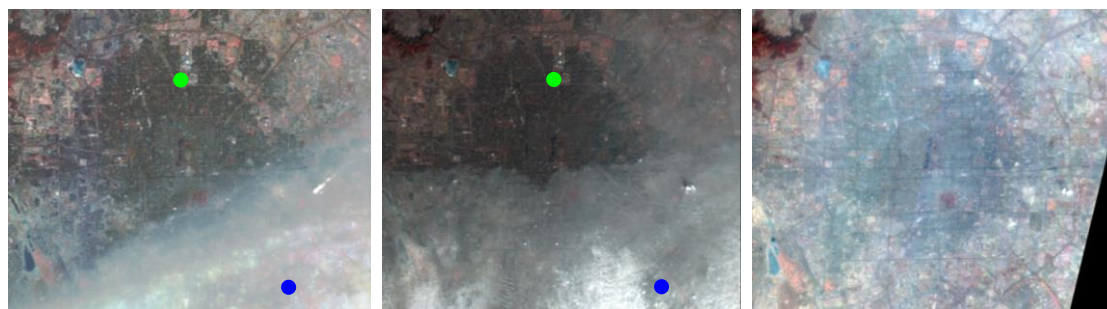


Fig S3. The high spatial resolution images of Beijing city from HJ-1 satellite (two days revisiting and passing time about 10:30 AM) on 10 Jan 2013 (left), 12 Jan 2013 (middle) and 27 Feb 2012 (right). The AOD site (green) and PM_{2.5} site (blue) locations in the 2013 haze cases are also marked on the graph.

We are very grateful for reviewer's general formatting and minor comments. All points are carefully corrected in the revised manuscript. Below are authors' responses:

Page 5094, line 12: AERONET direct sun measurements are made every 15 minutes, not 10 minutes as you say in this sentence.

Response: Corrected.

Page 5094, Section 2.1: Please state whether the data you present in this paper is AERONET Level 1.5 or Level 2.0 data. Additionally you discuss the direct sun calibration (lines 19-25) of the Cimel sun-sky radiometer but do not provide enough

specifics. For instance the Litang mountain Langley site is not considered a standard mountain Langley site for AERONET (Mauna Loa and Izana are). Therefore please provide some specifics on the AOD levels at this site during the Langley measurements and the repeatability of the V_0 values obtained from the Litang site. Also give an estimate of the AOD uncertainty that results from this calibration and compare it to the ~ 0.01 to 0.02 uncertainty for field Cimels, and 0.002 to 0.009 for Langley calibrated reference Cimels in AERONET (Eck et al., 1999). Regarding the vicarious calibration for sky radiances, please give an estimate of the calibration uncertainty since the vicarious method is not discussed in Holben et al. (1998) as you suggest.

Response: Litang's AOD levels are stable and as low as 0.02 - 0.03 at 870nm as shown in figure below. The AOD uncertainty by applying Langley V_0 calibration is estimated preliminarily to be less than 0.01 (the maximum difference between curves in Fig S4 can be a proxy) which is better than that of AERONET filed calibration (0.01 - 0.02) and less accurate than that of AERONET maser calibration at Mauna-loa or Izana (0.002 - 0.009). For the sky measurements, we supplement also descriptions on the radiance calibration uncertainties (3 - 5%) and polarization uncertainty (about 0.01 in degree of linear polarization) in the text.

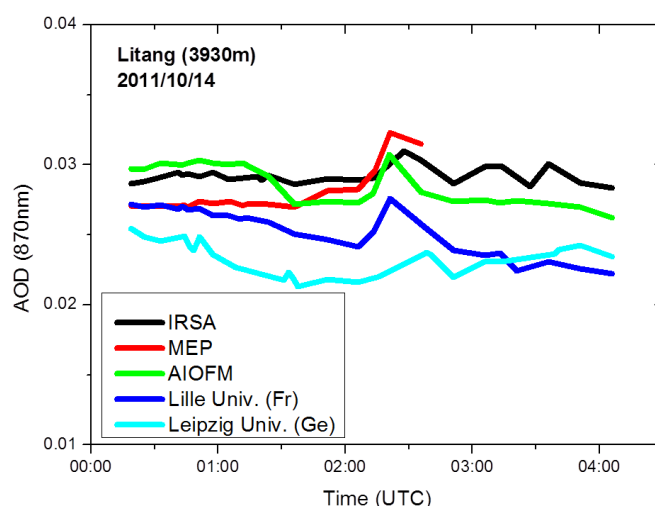


Fig S4. AOD comparison of five CIMEL sunphotometers by applying Langley calibration method performed at Litang (3930m) site.

Page 5095, lines 2-3: This sentence is confusing since sky dimming (irradiance reduction) is more closely related to AOD and not visibility, since visibility is weighted by near surface concentrations and not total column concentrations.

Response: We deleted "Haze can cause sky dimming".

Page 5095, lines 5: Please explain why $RH < 90\%$ was selected as your threshold since fog has 100% RH, so why not use a higher RH threshold, say 95% ?

Response: This value is proposed by a literature of Deng et al (2008). We supplemented this article in the reference:

Deng, X. J., Tie, X. X., Wu, D., Zhou, X. J., Bi, X. Y., Tan, H. B., Li, F., Hang, C. L.:

Long-term trend of visibility and its characterizations in the Pearl River Delta (PRD) region, China, *Atmospheric Environment*, 42(7): 1424-1435, 2008.

Page 5095, lines 15-16: Please revise this sentence to explain that higher Angstrom exponent can mean either more fine mode particles relative to the coarse mode particles or it could also be due to smaller radius fine mode particles (fine mode radius can vary substantially due to coagulation, humidification, and/or cloud processing).

Response: Thank you for this suggestion and corresponding changes have been made in the reversed manuscript.

Page 5095, lines 17-19: Please reference Sinyuk et al. (2012) regarding the measurement limitation of AERONET Cimels, which is 10 counts at 440 nm in AERONET and that this threshold is related to the issue of diffuse scattering into the instrument FOV. Also state in the text whether the 440 nm raw counts actually approached the 10 count cutoff value for the observations presented in this paper.

Response: We added reference to Sinyuk et al. (2012) in the reversed paper. It should be mentioned that we have stated previously in the paper that “AOD at 440nm may associate with much larger uncertainties than other three wavelengths due to reaching the measurement limitation of the CE318 instrument in these extremely heavy haze events”. Moreover, we showed here AOD (440nm) associated with $DN \leq 10$ (green cross symbols) in Fig S5 below. It can be seen that these points are acceptable in our cases compared with other 440nm points. This is explained by the reason that the 10 DN limitation suggest by Sinyuk et al. (2012) is mainly for the condition of dust (dominated by large aerosol particles) while our measurements in this work corresponding to haze aerosol composed mainly of small particles. So we decided to keep figure 1 no changed and supplemented reference to Sinyuk et al. (2012) in the paper text.

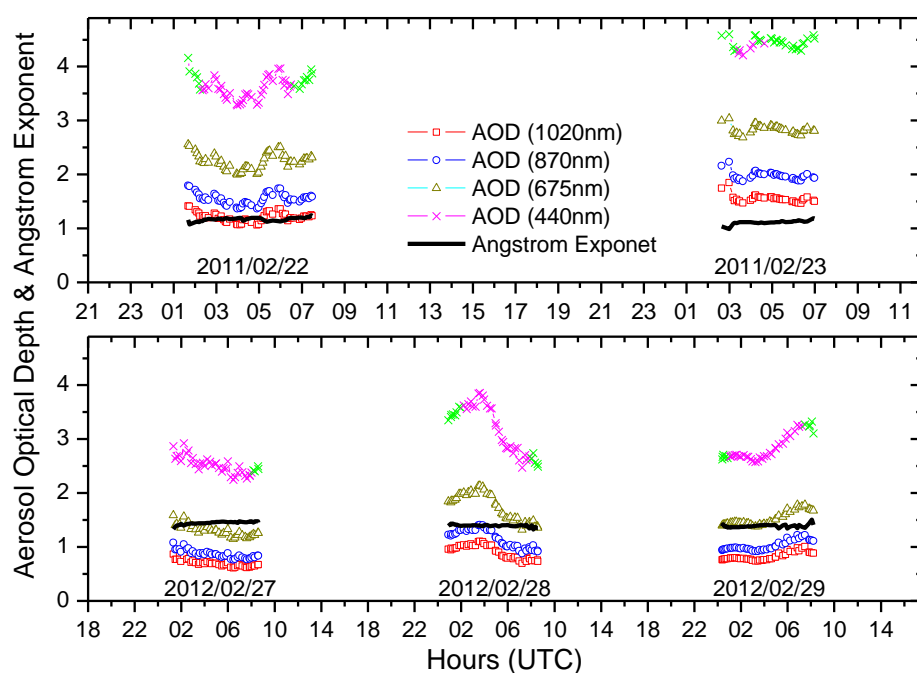


Fig S5. The same with Figure 1 of the paper except that AOD(440nm) points associated with DN \leq 10 are marked with “green cross symbols”.

Page 5096, lines 1-3: In section 2.1 you suggest that the data you present in this paper are AERONET data, yet it seems that the ground reflectance that you used is not consistent with AERONET. You used MODIS surface albedo from Li et al (2006) while AERONET uses the MODIS derived data base described by Moody et al. (2005) in conjunction with ecosystem based BRDF models (also from MODIS) to account for the large variation in albedo as a function of solar zenith angle (see Eck et al. (2008)). Please explain why the data processing you applied is not consistent with AERONET and make it clear that there are differences in the way you have processed this data.

Response: The observation data we used in this study come from one of test instrument (#350) of AERONET which has additional polarization measurements at all spectral wavelength compared with standard AERONET radiometers. The most important reason to choose this instrument is the advantage of coincident in situ measurements, i.e. aethalometer measurements used in part 4.1 to compare with BC fraction retrievals. Another reason is the advantage of polarization measurements which provides additional validation of retrieval results. These additional polarization data are not yet officially processed in AERONET but treated by the test procedure of Li et al. (JQSRT, 2009). The new process are based on the same core inversion part (Dubovik et al., 2000; 2006) and thus it can produce very comparable results with AERONET standard products (Li et al., 2009), even if small difference in some secondary procedures like surface model. Moreover, in the case of this paper, the retrieval results of optical and microphysical properties have been compared case by case with AERONET standard products and we found negligible difference in the level of quantities focused in this work.

Page 5096, lines 3-5: The Smirnov et al. (2000) cloud screening describes the use of triplet stability and also the 2nd derivative of the AOD in time to filter spikes, for cloud screening of the AOD data. The almucantar asymmetry check (not mentioned at all in Smirnov et al. (2000)) is done for additional cloud screening of the sky radiances prior to input to the Dubovik retrieval algorithm and is described by Holben et al. (2006; SPIE).

Response: Thanks for your suggestion. We have added the missed reference to Holben et al. (2006).

Page 5097, lines 17-20: Note that not all coarse mode particles in China are dust particles, some are fly ash emitted during coal combustion.

Response: Thanks for your suggestion. We have supplemented “It should also be noted that not all coarse mode particles in Beijing are dust particles and some are fly ash emitted during coal combustion.”

Page 5097, lines 24-26: It should be noted that the spectral variation in SSA as a

function of Angstrom or Fine Mode Fraction was shown in much more detail from the AERONET Beijing site climatology in Eck et al (2010).

Response: We added reference to Eck et al. (2010).

Page 5099, lines 20-23: In addition to volume percentage please also give the AOD of each mode, as the fine mode AOD and coarse mode AOD are also output computations of the Dubovik AERONET retrievals. Volume is somewhat misleading for these haze cases since a relatively large volume of coarse mode particles yields a relatively low value of AOD.

Response: Thanks for your suggestion. $AOD_f(440\text{nm})$ and $AOD_c(440\text{nm})$ are supplemented in Table 1 to provide AOD contributions of fine and coarse modes respectively.

Page 5100, lines 20-22: The values of the real part of the refractive index at 675 nm are 1.50 in 2011 and 1.48 in 2012, which are almost equal given the uncertainty of the retrieval for this parameter. Therefore I don't think you can state with much confidence that there is more water in the 2012 event.

Response: We agree with reviewer's comments and deleted "which indicates more water content in 2012 event than 2011" in the paper.

Page 5101, lines 19-23: This discussion of the high dust loading in winter is very misleading since for the cases presented in this paper the coarse mode fraction of the AOD is only ~ 3 to 10% of the total AOD at 440 nm. In other words the Fine Mode Fraction (FMF) of these cases range from 0.90 to 0.97 at 440 nm, thereby these are pollution-dominated events with likely a significant amount of the coarse mode composed of fly ash and not desert dust. FMF of optical depth is more important than fine or coarse volume ratio in the context of aerosol radiative effects.

Response: Thanks for your comments and suggestion. Following the supplied AOD_f and AOD_c in Table 1, the Fine Mode Fraction (FMF) is calculated to be 0.93 at 440nm. So we replaced the text with "Even DU occupy such a large volume fraction in the aerosol during the haze events, it should be noted that this is mainly caused by their large particle size and the Fine Mode Fraction ($FMF = AOD_f / AOD$) is more important indicator of the dominated component. The FMF is 0.93 in our case which means pollution-dominated events with likely a significant amount of the coarse mode composed of fly ash related to coal burning (Yang et al., 2009) or dust from local arid land emissions".