Thanks for your comments. Below are our responses to major comments:

(1) On the performance and sensitivity to errors in the retrieval, we added the new section 2.5:

2.5 Error estimation of aerosol component fraction retrieval

Uncertainties on the retrieved aerosol component fractions come mainly from two sources. One is the retrieval method itself, including assumption on the component mixing form, on the particle shape, on the refractive indices of representative components and so on, which is difficult to assess and might be not the dominative uncertainty source at present. Another uncertainty source is the errors in the input information, i.e. errors of the optical and physical properties from remote sensing measurements, which will be preliminarily evaluated here.

Uncertainties of the optical and physical properties obtained from sun-sky radiometer retrievals have been carefully assessed (Dubovik et al., 2000). Typical uncertainty on the real part of refractive index is about 0.04 while on the imaginary part about 40% and on SSA about 0.03. Following the algorithm description in section 2.4, the input parameter n is assumed spectrally constant and obtained from the average of real parts of refractive indices at 4 bands of sun-sky radiometer. The uncertainty of n is then estimated to be $(N_\lambda)^{-1/2} \times 0.04 = 0.02$ where N_λ equals 4. Similarly, uncertainty on k_{blue} is 40% and on k_{red} is about 23% thanks to the average of 3 bands. The uncertainty on dSSA is difficult to estimate and we can temporally set it to uncertainty of SSA, i.e. 0.03. It should be mentioned that the band difference form of dSSA (i.e. dSSA = SSA(870 nm) – SSA(675 nm)) should be able to eliminate most of systematical errors on SSA and thus uncertainty on dSSA could be less than 0.03.

We employ here the 2010 yearly averaged aerosol optical and physical properties at Beijing as the aerosol model in the error assessment. The refractive indices and SSA are shown in Fig 4 and the retrieved component fractions are 0.8%, 8%, 15%, 55.2% and 15% for BC, BrC, DU, AS and AW respectively. To test sensitivity of component fraction retrieval to errors in input information, we set the variation range and steps for n ([-0.05, 0.05], 0.01), for k_{blue} ([-50%, 50%], 10%), for k_{red} ([-50%, 50%], 10%) and for dSSA ([-0.05, 0.05], 0.01), respectively. The uncertainty estimation results are presented in Fig. 5.

It can be seen from Fig. 5(a) that absorbing components (BC, BrC and DU) are basically not sensitive to uncertainty on n, while non-absorbing components (AS and AW) shows counteracting behaviors and the maximum uncertainties are 10% corresponding to error range of n. In Fig. 5(b), we find that strong absorbing BC is nearly insensible to Δk_{blue}, while weak absorbing components (BrC and DU) are sensible. The AS shows roughly opposite variation versus that of DU and uncertainty of AW is nearly zero regardless of Δk_{blue}. In Fig. 5(c), we find that all component fractions are not affected by uncertainty of Δk_{red} in the range from -20% to 15%. Moreover, AS and DU are most affected components and their corresponding curves are symmetric. In Fig. 5(d), when ΔdSSA less than -0.01, component fraction uncertainties are larger, especially for DU and AS. This is explained by characteristics of the retrieval algorithm which employs dSSA to distinguish DU from BrC as

introduced in section 2.4. Once the sign of dSSA is changed due to ΔdSSA, the DU and BrC fractions will change significantly as well as that of AS and AW. However, sign changes of dSSA are expected to be rare in the retrieval, e.g. uncertainty assessment of SSA in Dubovik et al. (2000) shows that spectral SSA curves are usually shifted entirely when suffering from measurement uncertainties, which will keep the sign of dSSA no change. Therefore, in Fig. 5(d), we consider only ΔdSSA from 0 to 0.03 which results in smaller uncertainties on component fractions.

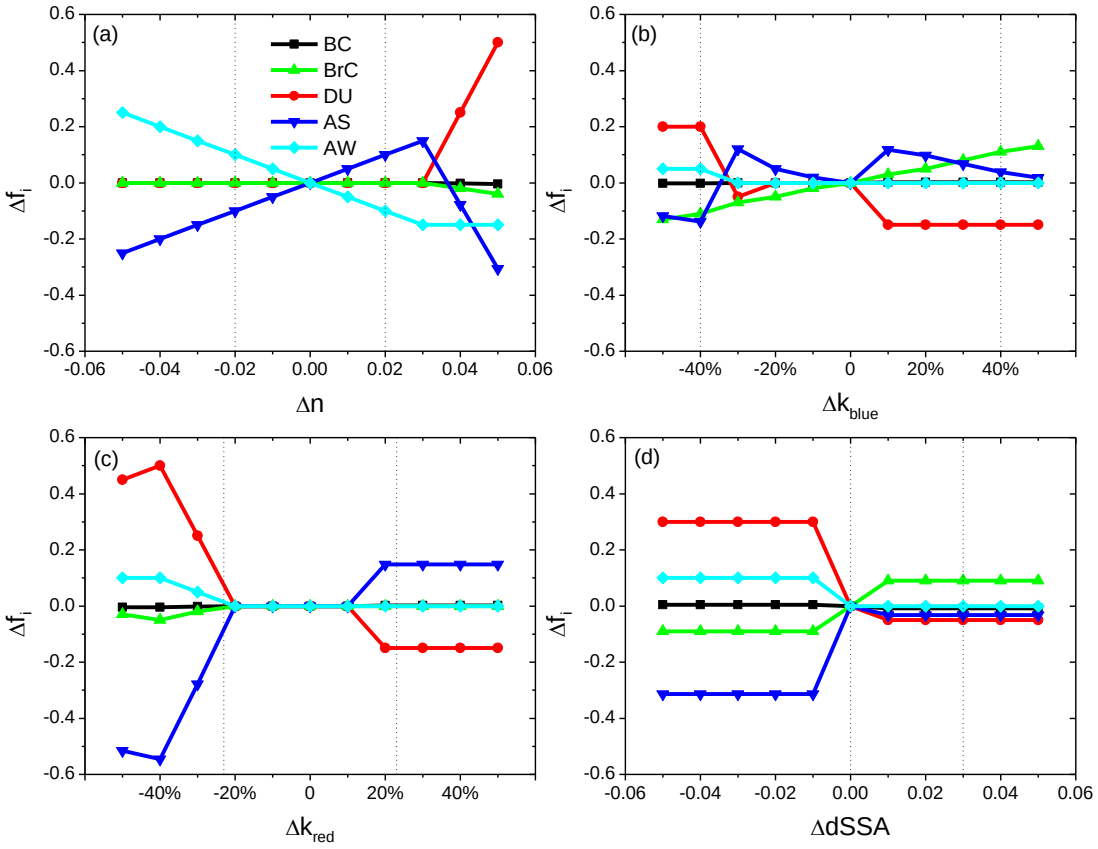

Fig 5. Uncertainty on the retrieval of aerosol component fractions. Errors in n, k_{blue}, k_{red} and dSSA are assessed with their maximum uncertainty ranges marked by dot lines in each graph.

Fig. 5 provides a case study to assess the aerosol component fraction uncertainties. Considering it is unlikely that all error sources (Δn, Δk_{blue}, Δk_{red}, ΔdSSA) reach coincidently the maximum, we assume that the possible uncertainty of each component corresponds to the maximum error caused by one of four error sources. Therefore, the maximum uncertainties on BC, BrC, DU, AS and AW are roughly estimated to be 0.008, 0.11, 0.2, 0.15 and 0.1 associated to error sources of ΔdSSA, Δk_{blue}, Δk_{blue}, Δk_{red} and Δn, respectively. Comparing with model's component fraction used in the assessment, the relative uncertainties in this case are about 100%, 140%, 140%, 30% and 70% for BC, BrC, DU, AS and AW components respectively. We should mention that aerosol component uncertainties as BC from emission inventories can be 200% or more (Schuster et al., 2005) and thus our retrieval are valuable and uncertainties can be expected to be further decreased in the future. Moreover, it

should be mentioned that in a stable atmosphere, e.g. the case of heavy haze, which can be verified by the stable AOD or other optical and physical properties, the sun-sky radiometer can provide 5-8 times retrieval results in a day and taking average can diminish significantly uncertainties. Therefore, in this study we will present mainly daily averaged component fractions.

(2) On the labeling of the aerosol groups, we thank very much for reviewer's suggestion and supplemented a new Table 1 in the paper to clarify the component labels.

Table 1. Aerosol component groups and their parameters used in the retrieval of chemical fractions from remote sensing measurements. n is the real part of refractive index, k_{blue} is the imaginary refractive index at blue band, k_{red} is the imaginary refractive index at red band, MAE is the mass absorption efficiency and ρ is density.

No.	Representative components	n	k_{blue}	k_{red}	MAE (675 nm)	MAE (870 nm)	ρ (g/cm^3)	Group Abbr.
1	Black carbon	1.95	0.66	0.66	8.14	6.32	2.0	BC
2	Brown carbon	1.53	0.063	0.005	0.067	0.05	1.8	BrC
3	Dust or fly ash	1.57	0.01	0.004	0.045	0.035	2.6	DU
4	Ammonium sulfate	1.53	1×10^{-7}	1×10^{-7}	-	-	2.3	AS
5	Aerosol water content	1.33	0	0	-	-	1.0	AW

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:

Abstract:

Line 1: replace "With the development of economy" with "With the increased economical development".

Response: corrected.

Line 10-11 : improper use of composition terms. This paper is about remote sensing and should not use these labels as they were actual composition measurements. A more proper labeling would be something like "aerosol fractions identified as BC, BrC. . .. " and so on.

Response: corrected.

Line 14: what do you mean by "stable"? not clear if mean change in time.

Response: we replaced "relatively stable" with "comparable".

Line 15: not clear the meaning of "Therefore, a parameterized heavy haze characterization was drawn to present a research" . Is the paper reporting parameterization? Is the paper parameterizing something? I do not think this is a proper use of the term.

Response: we replaced "a parameterized heavy haze characterization was drawn" with "averaged heavy haze property parameters were drawn".

Line 16-18: Please be more quantitative with respect to the size distribution. For example, you may want to add information about the fine mode fraction.

Response: we supplemented “The fine mode AOD is 2.8 corresponding to a fine mode fraction of 0.93.”

Line 21 : remove “obviously” , it makes the sentence confusing.

Response: we deleted “obviously”.

Line 23 : no proper use of the word “occupied”.

Response: we replaced “occupied” with “are”.

Introduction:

Page 5093, lines 1-4: vapor is invisible and it only contributes to visibility in the condensation phase. Please correct. Rewrite the whole sentence, the use of which is not recommended here.

Response: we rewrote the sentence “Haze can reduce severely atmospheric visibility due to increased extinction of suspended solid or liquid particles.”

Page 5093, Line 15: type “blown”.

Response: corrected.

Page 5093, Line 24: Clarify what a “new polarized sun-sky radiometer” is. Are the new Cimel detectors measuring polarization? Is this polarization used in the retrievals?

Response: we supplemented “dual wheels with an additional polarizer wheel in comparison with standard CIMEL radiometer equipped only with one filter wheel (Li et al., 2009)”.

Page 5094, Line 13-14: replace “in a triplet way within about 30 s which can be used to detect clouds” with “three times within 30 s and its variability in the period is used”.

Response: we replaced “in a triplet way within about 30 s which can be used to detect clouds” with “three times within 30 s and its variability in the period is used to detect clouds”.

Page 5094, Line 24-25: More information needs to be provided about the inherent uncertainty of the standard Aeronet retrievals. Relies on the quality of the retrievals to make inferences, the uncertainties need to be stated and not referred to a publication.

Response: we added description on uncertainties of the measurement in AOD, radiance and polarization. The retrieval uncertainties on the optical and microphysical properties are supplemented in part 2.3 as “particle size distribution (typical uncertainty of 25%) and the wavelength dependent refractive indices (typical uncertainty of 0.04 for real parts and 40% for imaginary parts) as well as other aerosol

optical parameters like SSA (typical uncertainty of 0.03)". The retrieval uncertainties on aerosol component fractions are evaluated in the supplemented section 2.5.

Page 5095, Line 15-19: I think the author should expand on this. OMI specially and probably MODIS have difficulty sensing aerosols at heavy loading conditions like these. One of the reasons are no sensitivity to the lower levels of the aerosol and it is assumed that a measurements like Aeronet from the ground would not have such problem. But if you think that Aeronet is having problems too like not been able to sense the whole column of the aerosol, please state it.

Response: The measurement difficulty of ground-based sunphotometer at very high AOD condition is mainly due to the reason of reaching the minimum detectable signal of the detector (in case of CE318, the Digital Number DN < 1). Moreover, in the case of dust aerosols when signal is low (e.g. CE318 DN less than 10), there can be forward scattering problem caused by non-zero field of view (FOV) of the photometer (Zhao et al., 2012). More information on this issue can be found in the response to comments of reviewer 1.

Page 5096, Line 21: what do you mean with "which agrees well with the calibration accuracy of the polarization measurements" ? is Aeronet measuring polarization and used in the retrieval or in the calibration only? I think you need to be more clear about this.

Response: We supplemented "where P^{meas} is the DOLP measured by CE318-DP and P^{cal} is the calculated DOLP using the retrieved aerosol property parameters."

P5097-5098: description of new algorithm. This is confusing and too brief. While it is appropriate to reference the original paper description of the algorithm, this algorithm is very new and the brief description provided is inadequate for the vast majority of readers who are not familiar with it and not necessarily will go to the original paper to learn about it. I suggest expanding this description to the point of dedicating a whole section to it (or an appendix).

Response: We expanded section 2.4 to provide a more detailed description of aerosol component fraction retrieval algorithm as below:

"Here we provide the description of algorithm when applied to sun-sky radiometer data as follows:

(i) Input. The program reads into aerosol optical and physical properties retrieved from CE318 and coverts them to parameters used in the component fraction retrieval, including n from the average of real part of aerosol refractive index at four bands (440, 675, 870, 1020 nm), k_{blue} from the imaginary part of aerosol refractive index at 440nm, k_{red} from the imaginary parts from 675 to 1020 nm, dSSA obtained by SSA(870 nm) – SSA(675 nm). Moreover, size distribution and AOD at 675 and 870nm are also read into and useful in step four.

(ii) Initialization. Refractive indices of five components (BC, BrC, DU, AS, AW), mass absorption efficiency (MAE) and density of three absorbing components (BC, BrC, DU) are set according to related literatures. Detailed numbers used in this study

are listed in Table 1.

(iii) Discretization of the solution space. We set steps of volume fraction of components to 0.2%, 1%, 5%, 5% for BC, BrC, DU and AW respectively, and the remaining fractions to AS. We notes "TN" for the total number of combination of possible solutions (each solution includes five volume fractions of aerosol components), i.e. $(f_i, i=1, 5)_j$ with j = 1, TN.

(iv) Calculate parameters of aerosol component mixture. We assume the internal mixture of all components and employ the volume average mixing rule (Heller, 1995) to calculate n^{cal}, $k_{blue}{}^{cal}$, $k_{red}{}^{cal}$ for each volume fraction solution $(f_i)_j$. Moreover, we assume spherical particles and utilize the size distribution, AOD, MAE and aerosol density to calculate $dSSA^{cal}$ following method described in Wang et al. (2013).

(v) Calculate residuals on each information. The optimization of function $\Psi(\varepsilon_n, \varepsilon_{k(blue)}, \varepsilon_{k(red)}, \varepsilon_{dSSA})$ provides the best solution for the volume fraction retrieval, where residuals on each input information defined as

$$\begin{aligned}
\varepsilon_n &= \frac{\left|n^{cal} - n\right|}{n}, \\
\varepsilon_{k(blue)} &= \frac{\left|k_{blue}^{cal} - k_{blue}\right|}{k_{blue}}, \\
\varepsilon_{k(red)} &= \frac{\left|k_{red}^{cal} - k_{red}\right|}{k_{red}}, \\
\varepsilon_{dSSA} &= \frac{\left|dSSA^{cal} - dSSA\right|}{\left|dSSA\right|}.
\end{aligned} \tag{3}$$

(vi) Find the best solution by optimization method. In the processing of optimization of function ψ, in order to avoid problems on weighting four kinds of input information (n, k_{blue}, k_{red} and dSSA), we utilize a "rank position priority (RPP)" strategy, instead of using the traditional method of absolute residual minimization. Firstly, we number an integer series $RP_j(\varepsilon_n)$ related to each member of solution space $(f_1, f_2, f_3, f_4, f_5)_j$, j = 1, TN. The value of $RP_j(\varepsilon_n)$, or the rank position, is calculated based on ε_n while $RP_j(\varepsilon_n) = 1$ for the minimum ε_n and $RP_j(\varepsilon_n) = TN$ for the maximum ε_n; Secondly, we create similarly the integer series $RP_j(\varepsilon_{k(blue)})$, $RP_j(\varepsilon_{k(red)})$ and $RP_j(\varepsilon_{dSSA})$ respectively; Thirdly, the j corresponds to $\min(RP_j(\varepsilon_n) + RP_j(\varepsilon_{k(blue)}) + RP_j(\varepsilon_{k(red)}) + RP_j(\varepsilon_{dSSA}))$ provides the optimization of function ψ and the corresponding $(f_1, f_2, f_3, f_4, f_5)_j$ is the best solution to optimize all kinds of information on n, k_{blue}, k_{red} and dSSA.

(vii) Output. The program prints fractions of BC, BrC, DU, AS, AW components corresponding to the best solution."

P 5099, 17: "the absolute value" of what? Please clarify.

Response: we replaced "the absolute value" with "the size distribution value".

P 5100, 1-5 : Can you speculate/suggest why the differences observed?

Response: we supplemented explanation "This might be explained by the hygroscopic growth of fine mode particles, i.e. with the increase of water content, fine particle radius and fine mode dV/dlnr augment simultaneously."

P5100: overall this page has too many grammar errors and poor choice of words. Here are some corrections, but by no means they are not all. Please have the text read by a native or very knowledgeable person in English. Line 1: correct with "exponent IN 2012 than IN 2011 in Fig. 1" Line 8 and 12 , replace "reflects" with "modulates" Line 13: replace "property" with "coefficient" Line 14: replace "reveal" with "indicate".

Response: corrected and we carefully revised the English throughout the paper.

P5100, line 22-23: Why do you state that water is responsible for the lower refractive index? Do you have any proof? Otherwise, it is just one of the possibilities.

Response: we deleted "which indicates more water content in 2012 event than 2011."

P5101, line 17: Any growth factor large 1, indicates that a particle will grow in the presence of water. So this sentence is not clear because it reads like if only particles with GF>1.1 will grow, which is not the case. Please clarify/modify the sentence.

Response: we replaced "When *gHGF* is larger than 1.11, the aerosol is hygroscopic (Schuster et al. 2009), indicating that the aerosols can absorb moisture from the environment." with "When *gHGF* is larger than 1, the aerosol is hygroscopic (Schuster et al. 2009), indicating that the particle will grow in the presence of water."

P5102, 20 to p5103,6: I do not see much value in comparing with the models used by 6S. The 6S aerosol models are old (1990's) and do not reflect much of the knowledge gained on aerosol optical properties since then. So, I suggest to remove these references and/or offer an alternative comparison with other algorithms/models.

Response: Thank you for this suggestion but we still want to keep the comparison with 6S aerosol models. Even the 6S models are old but they are still widely used in the satellite remote sensing community. Moreover, in China, there are none of well accepted aerosol models so that 6S is still the most popular model used in the remote sensing activities from our knowledge. That is also partly the reason for us to choose 6S models in the comparison.

P5104, line 5: replace with "2) there is a good"

Response: corrected.

P5104,7-10: this explanation is not clear. Are you referring to variability of aerosol in the atmospheric column? Is this the only explanation?

Response: we replaced "This can be explained by the fact that during heavy haze most of particles are suspended somewhat homogenous in the low level surface and no extra aerosol layers existed" with "This may be related to higher fine mode fraction (here 0.93) of haze aerosols."

P5104, 10: I am not fan of these equations. They are notably imprecise and difficult to generalize. Although I do not advocate for the removal, the authors need to be very clear under what conditions this equation is valid and can be applied. Otherwise it has little value and it could be used in the wrong set of conditions.

Response: we deleted the AOD-PM2.5 equation and supplemented more discussion like “Moreover, it is accepted that aerosol vertical distribution and ambient humidity corrections can also improve the correction between AOD and $PM_{2.5}$ (e.g. Kotchenruther et al., 1999; Tsai et al., 2011)”.