Reply to the comments from Referee #4

Dear reviewer,

We appreciate your comments and suggestions on our manuscript very much, which are valuable in improving the quality of our manuscript. With reference to the issues you raised about our paper, we give the following point-by-point reply.

Comment:

This paper compares retrievals of aerosol optical depth (AOD) from polarised radiance measurements by PARASOL with AERONET data at Beijing and Kanpur. It attempts to reconcile the relative biases in the satellite data with the partition between fine and coarse mode AOD as determined in the AERONET product, as the PARASOL dataset used is primarily sensitive to fine-mode particles.

Reply:

The work in the article is the pre-research work for the aerosol retrieval algorithm from PARASOL measurements in Asia urban areas with the aerosol type of fine/ coarse mode mixtures (The article introduced the algorithm is currently under review). In the new algorithm, the preliminary aerosol properties are retrieved from the multi-angle total (unpolarized) radiance of PARASOL. Possible AODs and aerosol models are achieved. The final decision of the correct AOD and aerosol model is made according to the retrieved fine mode AOD from polarization measurements of PARASOL based on the algorithm developed by Deuzé et al. [2001]. With the knowledge of the threshold radius of aerosol particles contributed to the polarized measurements, the decision can be made by comparing the AOD contributed by the particles within the sensitive radius which can be calculated using Mie or T-matrix with the retrieval from polarized measurements. According to the results in this work, the retrieval from polarized measurements has a good agreement with the AERONET AOD recomputed for the particles within the sensitive radius. And the new algorithm has also achieved a satisfactory accuracy in case studies. But the aerosol retrieval result even within the sensitive radius can have non-ignorable error caused by the depolarization effect of coarse particles or the uncertainty of the surface polarized reflectance model. In order to achieve better retrieval accuracy of the aerosol properties within the sensitive radius in the area of interest, the influence on the aerosol retrieval accuracy from polarized measurements produced by the pre-assumed aerosol model and the surface polarized reflectance model are evaluated to give proper suggestions.

I did not find the paper particularly informative, as the analysis was not very thorough, and the conclusions were not really new. These aspects of the PARASOL dataset used have been presented in previous studies, several of which are cited by the authors, and from my perspective the strengths and weaknesses of the Deuzé et al. [2001] approach are quite well-known. This is, for example, discussed in the AMT paper by Tanré et al. submitted to the same special issue of AMT, in which they point out the weaknesses of this decade-old algorithm can be largely overcome by the new AERONET-like full inversion of the PARASOL measurements presented by Dubovik et al. [2011] (which the authors of this manuscript even reference). The original algorithm paper also mentions some of the limitations. Further, the title does not reflect the paper's contents very well, as it is largely a comment on the AERONET validation of PARASOL at two sites (which I am not convinced the authors have performed in the most meaningful way; see specific comments).

Reply:

There are few studies discussing the sensitive radius of polarized aerosol retrieval, and the uncertainties in PARASOL aerosol retrievals caused by the algorithm-assumed aerosol and surface properties, respectively, in Beijing and Kanpur. The Deuzé et al. [2001] approach provides little information on the total AOD over these regions; however, the retrievals are highly correlated with the fine mode AOD, especially with the AOD for particles with radius less than 0.35 μm, which can be used to constrain the possible aerosol models and loads derived from the multi-angle measurements of PARASOL based on the spectral surface reflectance shape invariance principle (Flowerdew & Haigh, 1995; Diner et al., 2005). The Dubovik et al. [2011] algorithm may achieve good AOD estimates over Beijing and Kanpur, which require further study in our future work. The title do require further consideration to cover the paper's contents well, which include the validation work as well as the sensitivity study on the influence of aerosol model and surface properties, respectively.

I do not favour publication of this manuscript, as I do not believe it contributes meaningfully to the literature on aerosol remote sensing. Some specific comments are below, in case the authors decide to extend the analysis and submit a revised manuscript here or elsewhere in the future.

Reply:

The comments are helpful for the improvement of the article. The authors have discussed a lot about the content of the articles and feel sorry for some confusing expressions or descriptions. But we still believe there are useful materials for the development of the polarized remote sensing of aerosols in this work. We hope the article can be accepted after being revised.

Section 1

If you want to evaluate the performance of PARASOL for detecting the anthropogenic contribution of AOD, it is not obvious to me that Beijing and Kanpur are the best sites to do it. Although they are large cities with a significant pollutant contribution to the AOD, as the authors mention in the study, they also have significant seasonal input from e.g. transported mineral dust, and seasons of very high cloud cover. Depending on the objectives of the study, it could be better to consider additional or alternative sites. Looking at the AERONET websites, some urban/suburban sites without significant coarse-mode contributions, and with a long time series of measurements, include NASA GSFC, MD Science Center, Rome Tor Vergata, Fresno (as noted on the AERONET page for this site, The Central Valley of California regularly experiences some of the worst air quality in North America), Mexico City, or Lille. There may be more, and there could be issues limiting the *applicability of some of those sites listed. But I do not think the authors have justified their choices well enough.*

Reply:

The two sites chosen in this paper are both located in urban areas with large population, and both have similar aerosol type of fine/coarse mode aerosol mixtures. It helps us to develop improved or original retrieval algorithm for such aerosol mixtures. The study on the performance of PARASOL/POLDER over regions with the dominance of the anthropogenic fine aerosols have been carried out by several studies (Kokhanovsky et al., 2007; Vachon et al., 2004).

Section 2.1

The AERONET QA process is thorough and designed to minimise the chances of poor retrievals. So if the authors found that using only the highest QA AERONET data (level 2.0) gave a small sample size, while allowing level 1.5 improved sampling, then they should certainly evaluate in some form the differences between the level 1.5 and level 2.0 data at these sites. If AERONET inversions were not raised to level 2.0 then there was some reason for it, and the authors should be aware of, and attempt to account for, the possible implications of this for their analysis.

Reply:

This issue is also what we worried. There may be poor retrievals in the Level 1.5 dataset; however, as you figured it out, we chose the Level 1.5 dataset in the validation work, which allows more match-ups, to ensure the generality and effectiveness of the analysis. In the revised manuscript, we will address to the readers the possible deficiency arising from such a way. Besides, we've conducted the comparison of PARASOL AOD estimates against the Level 2.0 AERONET fine mode AODs. For example, in Beijing, the resulted linear regression slope and intercept, obtained from 64 match-ups, show nearly no change from those against the Level 1.5 dataset. However, slightly higher correlation (R^2=0.81) is found as compared to that of 0.79.

If the objective was more to evaluate to what extent the Deuzé et al. [2001] algorithm retrieves fine-mode AOD, then a possibly more appropriate approach would be to use the AERONET spectral deconvolution algorithm (SDA) dataset, which uses the spectral behaviour of AOD to partition between fine and coarse modes, without a size-cutoff radius of the type used by the Dubovik and King [2000] inversion algorithm. I would argue that, if you are primarily interested in fine vs. coarse AOD, the SDA data are more meaningful for this purpose. Additionally, as SDA data are derived from the direct-Sun observations rather than almucantar scans, the dataset is significantly larger, and it may be that the authors would find level 2.0 provided sufficient sampling in this respect.

Reply:

We did consider using the AERONET SDA dataset which would give significantly more match-ups. However, we do have the rationale why we finally gave up on it. We need to use the reflective indices and size distributions retrieved from almucantar scans, based on Dubovik and King [2000], to determine the sensitive radius of polarized detection of aerosols. The fine mode AODs with different size-cutoff radiuses were recomputed from the retrieved reflective indices and size distributions using Mie theory. The computation method was also used in Fan et al., [2008] and Su et al., [2010], in which a size-cutoff radius of 0.3 μm was used for Asia.

Section 3

It is not useful to state percentage over/underestimates in AOD, as these are only meaningful in the context of what the absolute AOD is, and how this compares with the stated and expected uncertainties of the data being compared. The authors do not provide this information. The stated correlations between AERONET and PARASOL data found by the authors are very high, which leads me to suspect that the offset between them is probably within the expected uncertainties, although without the authors providing this information it is not possible to say.

Reply:

This comment is very helpful. In the revised manuscript, we will provide more statistics, such as the Gfrac (fraction of accurate retrievals), the average AOD of the samples, as well as the RMSE (Root-Mean-Square Error), to generate more meaningful evaluation of the PARASOL AOD. The retrievals do have high accuracy when compared with the fine mode AOD truncated at 0.35 μm. The Gfrac for Beijing and Kanpur are 87% and 97%, respectively.

Section 4

I do not find significant utility in this section. I suspect much of the discrepancy arises from the way the authors performed their comparison with AERONET. It is not clear to me how the information presented here could be useful for algorithm developers or dataset users. The improvement in correlation you get by tweaking the cutoff radius for the fine/coarse partition is just saying to me that the method you are using for your validation is not particularly robust (in these cases it is not clear what the true unknown fine-mode AOD is, or how the discrepancies relate to the uncertainties on the data, only that by changing some assumptions made you are able to improve agreement between two retrievals). If the authors have a recommendation to make here, it should be clearly stated and justified. Additionally, it is not clear exactly what is being simulated in this section (one solar and viewing angle, for presumably a range of relative azimuth angles, and the two bands used by Deuzé et al. [2001]–why not consider the range of geometries sampled at the AERONET sites, which would give much more useful/representative results)?

Reply:

The comments of the reviewer on this section can be generalized as:

1) How can the work contribute to the improvement of the retrieval algorithm and the usage of the datasets?
2) The analysis method is not precise (The correlation is improved by changing some assumptions).
3) What is exactly being simulated and why not choose the geometries closer to the reality?

It is our carelessness to miss the statement on the contribution of the analysis result to the improvement of the retrieval algorithm. It will be given in the revised manuscript. This part of the work focuses on the factors introducing errors to the retrieved AOD within the sensitive radius. Since the aerosol model and the surface type are different for different seasons at the selected cities, the errors are different. The influences on the retrieval accuracy produced by the aerosol model and the surface polarized reflectance model are not equal. The analysis of the influence arising from the two factors on the retrieval accuracy for different seasons is helpful to understand the main reason for the different accuracies among seasons. And the conclusion of the analysis can provide proper suggestions on improving the retrieval accuracy.

Since the difference between the true and the pre-assumed aerosol polarized phase function leads to the retrieval error, the two kinds of polarized phase functions are simulated based on the season average aerosol model and that assumed in the algorithm, respectively. Since the polarized phase function is only related to the scattering angles for a certain aerosol model, one group of the typical solar zenith angle and view zenith angle is selected with a series of relative azimuth angles to provide enough scattering angles.

The analysis method is based on the average of AERONET data. Some necessary assumptions and approximation are made in the process of simulation(such as the selection of typical imaging geometry instead of a precise sample of the real geometries, taking the fine mode size distribution as the assumed size distribution, etc). In fact, some of the assumptions or approximations are inevitable, and the description of the applicable condition guarantees some rationality of the assumptions. But it is uncertain whether the accuracy of the simulation result can match the result from situ measurements. Considering the general rationality of the assumptions and the coherence of condition for different seasons, it is more meaningful for the evaluation of the relative difference among different seasons as compared with the evaluation of the absolute errors. For example, in the analysis of the simulation results, the depolarization effect caused by the coarse mode particles in spring for both sites leads to a significant underestimate of the AOD within the sensitive radius. A correction of the depolarization effect is suggested to obtain better accuracy. However, the correction in autumn and winter can be neglected. The error of the surface reflectance model is obviously higher for winter when compared with those of the other two seasons. An improvement in AOD retrieval accuracy can be expected if the surface simulation error in winter can be decreased.

The key point in this article is to give a thorough analysis on the factors impacting on the retrieval accuracy of the AOD within the sensitive radius. The uncertainties in the total AOD retrieval introduced by the assumed aerosol model shown in Table 5 are used in the later evaluation of the uncertainties in the surface reflectance model. Maybe it is not proper to show them here, as it tends to give readers a wrong feeling about the accuracy of the algorithm. In the revised manuscript, this confusing part will be deleted.

More precise evaluation work need situ measurements, maybe in later work this goal will be achieved.

Table 1

This would be much more useful if some information about the absolute AODs and absolute uncertainties were included.

Reply:

We will revise it according to your advice.

Table 2 (also text and Figure 2)

I am not convinced that seasonal means are useful here. I would imagine there would be a link between the AOD and the refractive index, as presumably elevated fine-mode AODs will be associated with more fine absorbing aerosol, and elevated coarse-mode

AODs will be associated with more dust. So by taking a mean you may end up with a situation which does not correspond to a condition which you are seeing in reality.

Reply:

Table 2 lists the seasonal mean refractive indices and fine/coarse mode volume concentration to explain the differences in the TOA polarized reflectance of different seasons, as shown in Fig. 3 and Fig.5. They are found to be largely correlated with the fine/coarse mode volume concentration. At both locations, the higher fine mode volume concentration C_{fine} and lower coarse mode volume concentration C_{coarse} is the primary reason that leads to higher TOA polarized reflectance in winter. The higher imaginary refractive index in winter may has some relation with a higher fine mode AOD comparing with those in other seasons. But the TOA polarized reflectance which is mainly contributed by fine mode particles is obviously higher in winter as compared to that in autumn with little difference between the imaginary refractive indices of the two seasons.

Additionally, as you are using level 1.5 AERONET inversions, and the sample size is small in some reasons, a single poor-quality inversion could affect your results dramatically. As mentioned before, it is important to examine the level 2.0 data as well, and quantify how your sampling decisions are affecting the conclusions.

Reply:

The possible influence of using the Level 1.5 AERONET dataset is addressed above.

Tables 5, 6

If you wanted to emphasise the differences when you alter the cutoff radius, it would be better to combine these tables in some way.

Reply:

We will revise it according to your advice.

Table 7

It is not clear what the numbers in this table refer to, or how significant they are to the *retrieval. For example, 107 % uncertainty at Beijing in winter for a surface of reflectance 0.005 still corresponds to a fairly small uncertainty in reflectance in absolute terms.*

Reply:

As mentioned above, the analysis is based on some assumption and approximation; the absolute accuracy may have a deficiency when compared with that obtained from the situ measurements. The average error of the surface reflectance model in different seasons and the comparison of the average errors in different seasons are much more meaningful. It shows the general accuracy of the reflectance model in different seasons. As shown in the Table 7, in Beijing the surface reflectance model overestimates by 46% in winter, while the aerosol models in this season introduce little error to the retrieved AOD (see Table 6). The two factors lead to an underestimate in AOD within sensitive radius (~24%). The surface reflectance model is more accurate for spring and autumn.

Figures 3-6

It is not clear to me what the particular information of interest to glean from these figures is. The flattening around 0.35 μm? Also, what is the reason for the non-monotonic behaviour from 0.22-0.23 μm? The authors should mention this (I don't think they do). They should also comment on how these changes in reflectance compare with the absolute and random radiometric uncertainty of the PARASOL measurements (if within the calibration uncertainty, it's not clear whether there is much we can do about it).

Reply:

Fig. 3 and Fig. 5 show the computed seasonal TOA polarized reflectance at 670 nm and 865 nm as a function of the size-cutoff radius for Beijing and Kanpur, respectively. Fig. 4 and Fig. 6 illustrate the corresponding TOA polarized reflectance difference defined in Eqs. (1), which express the difference between the TOA polarized reflectance contributed by the particles with radii less than the size-cutoff radius and that contributed by the entire size distribution (0~15 μm). And the sensitive radius in this work is obtained from Fig. 4 and Fig. 6. We got the conclusion of 0.35 μm because the TOA polarized reflectance difference reaches the minimum at r_{cutoff} of about 0.35 μm, after then $\Delta\rho_{\mathrm{p}\_r}$ do not change much with the increase of r_{cutoff}.

The non-monotonic behaviour at 0.22-0.23 μm possibly arises from the chosen scattering angle, as it appears for each season at both locations. The detailed analysis will be given in the revised manuscript.

The polarization calibration uncertainty of PARASOL is reported to be about 0.5%, which is larger than the TOA polarized reflectance differences as shown in Fig. 4 and Fig.6. However, the TOA polarized reflectances are calculated at a scattering angle of 100 degree. As shown in Fig. 1 and Fig.2 in Deuzé et al. [2001], the scattering angle of 100 degree does not corresponds to those that have larger aerosol polarized phase function values. The TOA polarized reflectance differences would probably increase obviously if calculated at a scattering angle between about 45° and 90°. However, the variation tendency of the TOA polarized reflectance differences as a function of the size-cutoff radius would not change.