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Interactive comment on "Estimation of aerosol complex refractive indices for both fine and coarse modes simultaneously based on AERONET remote sensing products" by Ying Zhang et al.

Ying Zhang et al.

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Dear reviewer:

Thank you very much for your careful review and constructive suggestions with regard to this manuscript. We appreciate for Reviewer's work earnestly, and hope that the corrections will meet with approval. Please find below our detailed responses to reviewer's question and comments.

Referee #4 The Authors present a method for the separate estimation of the aerosol refractive index from AERONET data. First they fit AERONET aerosol size distribution to

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a multimodal log-normal distribution, then they group the modes of the fitted log-normal distribution into a "fine" and a "coarse" mode, and then they proceed to the estimate of the refractive index of each mode by an iterative fitting AERONET total and absorption AOTs to Mie forward calculations. The proposed method looks fine to me. The steps of the procedure are well identified, the underlying assumptions are clearly stated and so are the limitations of the method (e.g., not taking the possibility of nonspherical particles into account). The validation on synthetic data, instead, looks a bit shallow, because the Authors only test three configurations, in which three realistic fine mode aerosol types (water-soluble, biomass burning and dust) are combined with a "default" coarse mode with refractive index 1.53+i0.008: in this section I would have been curious to see tests with more combinations of aerosol parameters. Anyway, in the last section of the paper the Authors also make the effort of applying their method to real AERONET measurements taken at Beijing, and they show that their separate retrieval allows a reasonable physical interpretation (which is probably the best possible "validation" of a refractive index retrieval, given that independent correlative measurements of this parameter are very difficult to obtain. and even an objective definition of the refractive index of a mixture of aerosol components is problematic in itself). Furthermore, the Authors show that their multicomponent refractive index retrievals fit AERONET AOTs quite well. In view of this, I think this paper can be published with minor revisions. I would recommend, though, a proofreading by a native English speaker, because the quality of the written English looks below par in some parts of the manuscript. Below are some suggestions for the modification of some unclear statements, and some other minor comments.

MINOR COMMENTS

(1) P1, L4-5. I would suggest to change "...based on AERONET aerosol products, including" etc., with "...based on AERONET volume particle size distribution" etc.

Response: We have corrected according to the Reviewer's comments. "This paper establishes a method to separate CRIs of fine and coarse particles based on AERONET

volume particle size distribution (VPSD), aerosol optical depth (AOD) and absorbing AOD."

(2) P1, L5-6. The sentence "The method ... simultaneously" is a bit unclear. Consider removing it or rephrasing with something like "The method consists of two steps. First a multimodal log-normal distribution that best approximates the AERONET VPSD is found. Then the fine and coarse mode CRIs are found by iterative fitting of AERONET AODs to Mie calculations."

Response: We have corrected according to the Reviewer's comments. "The method consists of two steps. First a multimodal log-normal distribution that best approximates the AERONET VPSD is found. Then the fine and coarse mode CRIs are found by iterative fitting of AERONET AODs to Mie calculations."

(3) P2, L8-9. I do not understand what the last two sentences mean. Especially the last one ("Raul and Chazette etc.").

Response: We have corrected the sentences in the manuscript. "In addition, Li et al. (2006) further added the polarized sky radiance measurements to the inversion algorithm in order to better constrain AERONET CRI retrievals. The Lidar measurements are also used to obtain CRI of aerosols within planetary boundary layer (Raut and Chazette, 2007)."

(4) P2, L17. Change "There are only few studies ... attempted..." to "Only a few studies ... attempted...". Furthermore, Wu et al. (2015) does not describe retrieval from ground-based measurements. It describes retrievals from airborne measurements.

Response: We have corrected according to the Reviewer's comments. "Only few studies (e.g. Xu et al., 2015; Wu et al., 2015) attempted to retrieve CRI of both fine and coarse modes simultaneously from advanced remote sensing measurements, e.g. multi-spectral polarized sky radiance."

(5) P3, L12. σ i is the "geometric standard deviation" (not the ordinary one) of r for each

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mode. As an alternative, if you prefer not introducing the concept of geometric standard deviation, you can say that " $\ln \sigma i$ is the standard deviation of $\ln r$ for each mode".

Response: We have corrected according to the Reviewer's comments. "Ci (μ m3/ μ m2) and ri (μ m) and ln σ i are the volume modal concentration, median radius and standard deviation of ln ri for each LNM mode, respectively."

(6) P5, L1. Cite: J. A. Nelder, and R. Mead (1965), "A simplex method for function minimization". Comp. J., 7, 308-313, doi: 10.1093/comjnl/7.4.308

Response: We have cited the paper according to the Reviewer's comments. "..., an iterative procedure is performed by minimizing Chi-Square on VPSD (see Eq.5) using the NelderMead simplex algorithm (Nelder and Mead, 1965; Lagarias et al., 1998)."

(7) P9, L1. What does "In a meaning of band average" mean?

Response: "In a meaning of band average" means the RMSE is calculated in each wavelength and the largest RMSE in five wavelength appears in the dust type. We rewrite the sentence as follows: "The largest root-mean-square-error (RMSE) of fitting τ appears in the dust type, corResponse to the underestimate of nf and nc (Fig. 3)."

(8) P10, L16-17. I do not understand the meaning of the last sentence: "Either sensitivity on τ or τ a will be able to support the estimation of related sub-CRI parameters".

Response: This is an English expression problem. We replaced the sentence by "this suggest that both τ and τ a sensitivities contribute to the convergence of the iterative scheme. Given only one information (τ or τ a) is sensitive, it is still possible to constrain the scheme give its sensitivity is strong enough."

TECHNICAL CORRECTIONS

(1) P2, L16. Consider moving "inventories" before the parenthesis.

Response: We have corrected according to the Reviewer's comments. "..., aerosols radiative properties are simulated based on source emission inventories (i.e. fine and

coarse sources separately), ..."

(2) P2, L17. P2, L17. "knowledge of . . . are essential" -> ". . . is essential . . . "

Response: We have corrected according to the Reviewer's comments. "..., and thus knowledge on CRI of different aerosol modes is essential to validate model performance for the assessment of aerosol climate effects."

(3) P3, L21. "subsequence" -> "subsequent"

Response: We have corrected according to the Reviewer's comments. "The above assumed spectral properties of sub-CRIs are useful to simplify subsequent procedure and it basically fits current knowledge on aerosol properties."

(4) P5, L26, "an" -> "a"

Response: We have corrected according to the Reviewer's comments. "e). Find the optimal solution based on a Limited-memory optimization algorithm (BFGS: Broyden–Fletcher–Goldfarb–Shanno) (Zhu et al., 1997) by constraining both $\tau(\lambda)$ and $\tau a(\lambda)$ with AERONET products."

(5) P5, L28. "achieves" -> is achieved. "If yes" -> "If so"

Response: We have corrected according to the Reviewer's comments. "Check if the convergence, (fi - fi+1)/max(fi+1, fi, 1) < $\eta \times \varepsilon$, is achieved. If so, output the separated sub-CRI parameters, . . ."

(6) P9, L5. "preform" -> "perform". "access" -> "assess" ?

Response: We have corrected according to the Reviewer's comments. "In order to evaluate the overall performance of the estimation scheme, we perform numerical experiments to assess errors of output sub-mode CRI parameters related to: ..."

(7) P9, L18. "imagery" -> "imaginary"

Response: We have corrected according to the Reviewer's comments. "The errors

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caused by uncertainty in VPSD are guite different for real and imaginary parts of CRI."

(8) P10, L16. "Another saying" -> "In other words"

Response: We have corrected according to the Reviewer's comments. "In other words, this suggest that both τ and τ a sensitivities contribute to the convergence of the iterative scheme. Given only one information (τ or τ a) is sensitive, it is still possible to constrain the scheme give its sensitivity is strong enough."

(9) P11, L2-3. "relative" -> "relatively", "presents" -> "present", "high" -> "higher", "in the case" -> "for this case" ?

Response: We have corrected according to the Reviewer's comments. "Although the relatively low sensitivity to nf present in DU type, the $\delta \tau/\tau$ is still higher than the sensitivity threshold for this case."

(10) P11, L4. "sensibilities" -> "sensitivities"

Response: We have corrected according to the Reviewer's comments. "The sensitivities of nc of all three types are considerably low, . . ."

(11) P12, L13. ". . . the hygroscopicity . . . are significantly increased" -> ". . . is significantly increased"

Response: We have corrected according to the Reviewer's comments. "..., this discrepancy suggests that hygroscopicity of fine particles is significantly increased in summer under high humidity condition."

(12) P12, L15-16. Consider removing the parentheses from "(kf and kc)", and from "(kf;440 and kc;440)".

Response: We have corrected according to the Reviewer's comments. "(iii) In Fig.6c, it can be seen that characteristics of kf and kc are similar with that of kf,440 and kc,440, except for the enlarged seasonal variation amplitude (especially for kc,440)."

(13) P14, L5. What does "online" mean in this sentence?

Response: The "online" means "real-time". We rewrite this sentence as follows: "..., e.g. the joint extinction, absorption and size distribution observation obtained from measurements in real-time."

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