Reply to comments of Referee #3.

The authors would like to thank Referee #1 for careful reading of the manuscript and valuable comments.

Questions:

1. - What is the dominant aerosol component in Rexburg, Mongu, Mezaira, and GSFC, and how is it justified? Any supporting information to describe the air condition (particularly the aerosol composition) for selected cases. For example, readers do not know if BrC is really dominant for the cases treated in Fig. 1.

Unfortunately, detailed information on aerosol composition at selected sites is rarely exits since it would require many instrument types co-located for a full optical and chemical characterization of the aerosol. Nevertheless, we tried our best to add more information and also included three more references. One is for Mongu aerosol composition:
Eck, T.F., Holben, B.N., Ward, D.E., Mukelabai, M.M., Dubovik, O., Smirnov, A., Schafer, J.S., Hsu, N.C., Piketh, S.J., Queface, A. and Roux, J.L., 2003. Variability of biomass burning aerosol optical characteristics in southern Africa during the SAFARI 2000 dry season campaign and a comparison of single scattering albedo estimates from radiometric measurements. *Journal of Geophysical Research: Atmospheres*, *108*(D13).
Two others are related to Mezaira site which is dominated by desert dust: Reid, J.S., Reid, E.A., Walker, A., Piketh, S., Cliff, S., Al Mandoos, A., Tsay, S.C. and Eck, T.F., 2008. Dynamics of southwest Asian dust particle size characteristics with implications for global dust research. *Journal of Geophysical Research: Atmospheres*, *113*(D14),
Eck, T.F., Holben, B.N., Reid, J.S., Sinyuk, A., Dubovik, O., Smirnov, A., Giles, D., O'Neill, N.T., Tsay, S.C., Ji, Q. and Al Mandoos, A., 2008. Spatial and temporal variability of column-integrated aerosol optical properties in the southern Arabian Gulf and United Arab Emirates in summer. *Journal of Geophysical Research: Atmospheres*, *113*(D1).
Also, a sentence was added to clarify aerosol types for Rexburg and Rimrock: High aerosol optical depth events dominated by fine mode particles (high AE) in the US northern Rocky Mountain region in June-October are dominated by biomass burning emissions.

2. Can we generalize the finding and lessons in this study based on this four sites only?

The main idea of the manuscript is that no strong smoothness constraints are imposed on spectral dependence of the imaginary part of refractive index of aerosols of any type for the new REL retrievals. Therefore, there is no artificial suppression of spectral dependence of aerosol absorption. We think this is general statement applicable to all aerosol types including mixtures of aerosols of different types. The analysis for four sites dominated by four main aerosol types (BC, BrC, dust, industrial) demonstrated successful performance of inversions employing the new relaxed smoothness constraints over a wide range of aerosol characteristics .

3. Why this new REL only make some change for the BrC-dominated biomass burning (BB) aerosols, not the mineral dust and BC-dominated BB aerosols, which are other radiative absorbing aerosols?

This is because in V3 the constraints on the spectral IPRI were already relaxed for coarse mode mineral dust. In V3 the Lagrange multiplier for spectral variation of the imaginary part was interpolated from dust (low AE, low value of Lagrange multiplier, relaxed smoothness constraints) to fine mode (high AE, high value of Lagrange multiplier, strong smoothness constraints). For BC dominated aerosols strong smoothness constraints in V3 for the imaginary part of refractive index worked due to the flat spectral dependence of IPRI of BC. This is described in Introduction and Section 2.

4. This new REL can help the retrieval of qualified SSA data in 340 nm channel?

We think it possibly can, depending on the data quality. In AERONET, we started looking at 340 nm to estimate if the sky radiance signal is strong enough above the noise level and if we can accurately calibrate this channel for sky radiance measurement.

5. This new REL enable us to have SSA data more under the low AOD case; Usually the SSA analysis relates to the polluted case, at least AOD > 0.4 due to the uncertainty issue. It is curious to see if this REL can lower the uncertainty of SSA retrieval in less-polluted case. In other words, application of new REL is only helpful for the retrieval in the polluted condition (often related to the high AE because of the general contribution of find mode particle to the large air pollution), or it is also useful to improve the retrieval in the lower AOD case of polluted (urban) area where the brown carbon is dominant.

Employing REL does not increase sensitivity to aerosol absorption, therefore the AOD>0.4 condition still stands in order to have the same uncertainty in retrieved SSA. The REL constraint can lower the sky residual error in all the cases where STD were suppressing spectral dependence of the imaginary part of refractive index and therefore result in more retrievals that meet the L2 sky error criteria. The possibility of lowering the AOD threshold for SSA retrieved in polluted areas depends on the accuracy of the sky radiance calibration. Alternative calibration methods with higher accuracy will be investigated after enough statistics of REL inversions will be available.

Minor and specific comments:

1. - Nowadays, there are so many AERONET stations and really long-term measurement data have been accumulated. In this situation, it is curious if we really can apply the analysis result only based on a several cases to the general situation (only some days were selected for the analysis even in only 'four' sites). The analysis in this study looks qualified with reliable cases showing clear dominance of a target aerosol composition, which can be a representative example for the meaningful discussion for new REL impact under the certain situation. But still, it seem the limited discussion because now we have so abundant information of AERONET measurement for several hundreds of local stations. Thus, the statistical analysis using the large number of dataset will be more expected for the generalization of findings in this work. In my opinion, this manuscript can be a good paper as a case study to show the 'possibility' for the usage of new REL for better expression of BrC optical properties. But it may be better to prepare another manuscript for the 'generalization of finding in this study'. In the second manuscript, the statistical analysis looks much required.

We absolutely agreed. REL constraints will be the part of upcoming Version 4 of AERONET aerosol retrieval algorithm. All the data will be reprocessed with REL and statistics of the difference between STD (V3) and REL (V4) for all the sites will be produced, analyzed, and published.

2. Abstract seems too long, so the key point of this study is not well transferred to readers. The word number in this abstract is > 800, which looks too much compared to the general criteria (~ 200 to 300 words. I do not know the limitation of word number in ACP/AMT).

   We do not find any abstract limitations for AMT publication. However, we reduced the abstract by ~20% which we think makes it more concise and clearer.

3. Line 46-50: Two sentences are not connected well (First one mentioned DRAGON campaign, but second on mentioned the DISCOVER-AQ campaign. How to connect the story here?)

In new, shorter version of the abstract these sentences were removed.

4. Line 56-57: How to understand this sentence? (What is the relationship between the insigficant impact for the mineral dust and relatively small impact for the BC-dominated biomass burning aerosol?)

This sentence was removed in new version of abstract.

5. Line 104-106: The reference or clues are required to raise this issue about the BrC. Now there is no surporting information associated with this statement, which looks very essential for the motivation of this study.

The following reference is added to this sentence: Kirchstetter, T., W., and Thatcher, T., L.: Contribution of organic carbon to wood smoke particulate matter absorption of solar radiation, Atmos. Chem. Phys., 12, 6067-6072, 2012.

6. - Line 214: => For example,

Corrected.

7. - Line 242: BrC carbon => BrC

Corrected.

8. Line 252-255: I am not sure if this kind of discussion is possible without any fire or humidity information in this case.

This is a discussion on the possible range of conditions and relative phase of combustion from biomass burning and the resulting variation in composition of the aerosols. It is pertinent to the differences in spectral SSA that are being discussed. References to support this discussion are also given.

9. - Line 270-307: I am not sure the existence of BrC in GSFC, but It may be also possible to see high amount of BrC in the urban region in the urban region, and the BrC pattern (e.g., hygroscopic growth related to the extent of aging) can be regionally different: (e.g., Zhang et al., GRL, 2011, https://doi.org/10.1029/2011GL049385). It will be useful to see if there is difference of the BrC optical pattern between the biomass burning and urban area in the further study.

   We agreed. As soon as a large number of REL inversion will be available this will be investigated. Actually, separate and detailed future research on absorbing aerosols is planned.

10. - Line 360-363: This manuscript does not have the chapter of 'data description' or 'methodology'. So there is no information of SSA from in-situ measurement in DRAGON-MD campaign. A short phrase to mention Schafer et al. (2014) may not be enough because the SSA estimation using in-situ measurement itself can make the large difference from the optically measured SSA (e.g, surface representative vs. column information). so at least several statements about the data/methodology of in-situ SSA calculation looks needed.

The following modification was done including additional sentences:

The SSA values are derived from in situ measurements made during aircraft vertical profiles of scattering and absorption coefficients at 550 nm. For each profile, 1 s sampled values of scattering coefficient measurements at 450, 550, and 700 nm from the nephelometer and absorption coefficient measurements at 470, 532, and 660 nm from the Particle Soot/Absorption Photometer were provided, both from dried air samples. At 550 nm, an additional scattering measurement at ambient relative humidity allowed for the calculation of an ambient SSA (rather than dried aerosol) that is more suitable for comparison with the SSAs derived from AERONET radiance measurements. In order to produce a column SSA value to compare with AERONET, the 1 s SSA aircraft measurements were averaged for the duration of the profile sampling after weighting the values according to aerosol loading (Schafer et al., 2014).