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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Extremely large anthropogenic aerosol component over the Bay of Bengal during winter season" *by* D. G. Kaskaoutis et al.

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Reviewer 2

General comments

The paper by Kaskaoutis et al. deals with microtop measurements made on a cruise during a campaign W-ICARB organized by Indian Space Agency. Angström exponent (alpha) and curvature (a2) derived from polynomial fit to aerosol optical depth spectra have been analyzed as well an attempt has been made to understand the dynamics of aerosol size distribution (growth, coagulation etc.) by deriving a relationship between alpha and spectral curvature of alpha (dalpha). Contribution of various aerosol species

to columnar optical depth has been inferred by constraining the measured spectral optical depths to those obtained from Optical Properties of Aerosol and Cloud (OPAC). However, there are serious pitfalls in all of the above analyses which I list one by one. That said there is no significant advancement of the present knowledge of BoB aerosol size properties already published (e.g. Nair et al., 2008; Kedia and Ramachandarn, 2009 and many more) based on radiometery therefore I recommend the paper be rejected for ACP.

We strongly disagree and surprised with the Reviewer comments that the present results are similar to those published by previous studies over BoB and that the present work has no significant advantage of the scientific knowledge of BoB aerosols. In the last part of the Introduction section we highlight the goals of the present study as well as the advantages of the W-ICARB cruise campaign against previous campaigns conducted over BoB. Except of this, the studies referred by the Reviewer (Nair et al., 2008; Kedia and Ramachandran 2009) deal with aerosol properties over BoB and Arabian Sea regarding a previous campaign (ICARB) conducted in March-May 2006 and NOT the present one (W-ICARB). Furthermore, none of these or other studies conducted over BoB throughout the years deals with identification of the aerosol types, not using the Gobbi's et al. identification scheme for monitoring of the aerosol properties and modification processes over BoB. Moreover, the present work is the first one that analyses the spectral aerosol properties obtained from OPAC based on the measured spectral AODs. As far as W-ICARB is concerned, there are some published articles (Kumar et al., 2010; Moorthy et al., 2010; Raghavendra Kumar et al., 2011; Sinha et al., 2011) that are cited in the manuscript, which do not attempt to identify the aerosol types or modification processes over BoB as done by us in the present study. We will take care of some of the missing references and citations of work carried out by others in the revised manuscript. We strongly believe that the present work contributes significantly in advancing the scientific knowledge of the aerosol optical properties and types over BoB during winter season.

Specific Comments

1. The authors used the method presented in detail by Schuster et al. (2006) for alpha and a2 deduction however they should have noted that Schuster et al. (2006) clearly mention the cutoff size as 0.6 micron (effective radius) which is the mode of separation for coarse and ïňĄne aerosol, as recommended by aeronet team. Strangely enough the authors have not even cared to mention this cut-off size. Schuster et al. (2006) were careful not to mention about the source of aerosols except restricting to size distribution analysis. Authors should know from India aerosol climatology that a significant amount of anthropogenic aerosols may be found in coarse mode aerosol with effective radius > 0.6 micron due to mixing and aging processes. In addition to that the direct usage of the classification developed by Kalapureddy et al. (2009) for Arabian Sea is highly questionable given the striking differences between BoB and AS aerosols.

The initial use of a2 or the second derivative of AOD in logarithmic co-ordinates for the classification of aerosols was performed by Eck et al (1999) and in his next publications Eck et al. (2001, 2005), which are cited by us. Furthermore, the same technique for identification of aerosol types has been already used over Indian mainland and the Arabian Sea (Kalapureddy et al., 2009; Kaskaoutis et al., 2009, 2010; Kedia and Ramachandran, 2009). So, such application is absolutely acceptable from the international scientific literature. In our study, we initially discriminate the aerosol types using the AOD versus alpha relationship and not any method by Schuster et al. (2006). We are rather surprised how the Referee mentioned that we have used the method presented by Schuster! After discriminating the aerosol types, we used the relationships between AOD and a2 or alpha and a2 in order to further insight in to the aerosol properties as Schuster et al. suggested, i.e. “the use of curvature is an additional tool for the discrimination of different aerosol properties”. We do not refer any threshold value for fine and coarse aerosols, since we do not use size distribution analysis. We do agree with the Reviewer that the anthropogenic aerosols over India can be found in sizes larger than 0.6 microns and, for this reason, Moorthy et al. (2010) and Raghavendra

Kumar et al. (2011) used the threshold of 1.0 micron for the discrimination between accumulation and coarse aerosols over the BoB during W-ICARB. However, in their studies they used aerosol size distribution measurements, which is not the case in our study and for this reason such a threshold is not referred. Nevertheless, Eck et al. (1999, 2001, 2005) do not refer that the use of a2 is limited only for a specific aerosol range, i.e. fine or coarse, but they used this technique over all spectrum and for all aerosol sizes. The same technique was followed by us. On the other hand, we used the same thresholds for AOD and alpha for the discrimination of the different aerosol types as those used over the Arabian Sea (Kalapureddy et al., 2009) in order to compare the results over the BoB to those obtained over the Arabian Sea. The results show significant differences in the aerosol optical properties and types between the two oceanic regions, as Reviewer pointed out, but these differences may be revealed if the same technique and thresholds are used.

2. The methodology used in second part of the paper based on a visual analysis approach of Gobbi et al. (2006) is also erroneous. First, the authors should note that definition of dalpha proposed by Gobbi et al. (2007) is valid for AERONET measurements, which has well known uncertainties for various channels. It was expected from the authors to perform a rigorous analysis before using same set of wavelengths as used previously. Secondly, the details of computations performed are missing. How is the use of low and uniform absorbing refractory index justified when the authors subsequently found highly absorbing aerosols with significant spatial variation in their single scattering properties in BoB?

Initially, we performed a detailed analysis for data screening taking special care about the validity of our spectral measurements using the method by Cachorro et al. (2004) for avoiding diurnal artifacts in the spectral AODs and further showing the reliability of our dataset by means of the a2-a1 vs alpha relationship. Nevertheless, in the revised version we have now discussed in detail the errors and uncertainties, which are very low, in AODs, alpha and a2 retrievals. Furthermore, the errors in a1 and a2 parameters using the second order polynomial fit are lower than those computed in a similar study conducted over the Arabian Sea (Kaskaoutis et al., 2010). The Gobbi's identification scheme can be performed only qualitatively and not quantitatively. We do not use this scheme to retrieve any measurement of fine-mode fraction or fine-mode radii, but only to qualitatively monitor the aerosol modification processes for increasing AOD. The same has been done in the previous studies, i.e. Gobbi et al. (2007), Basart et al. (2009). Furthermore, this scheme is not restricted only to AERONET data, since it has already been used over the marine environment of the Arabian Sea (published paper in ACP by Kaskaoutis et al. 2010) using Microtops-II data and over the urban environment of Athens (published paper in ACP by Gerasopoulos et al. 2011) using MFR data. Especially in the studies of Gobbi and Basart, the same scheme was used for the description of aerosol properties over very differentiate environments, including high-polluted urban areas, desert environments, locations affected by biomass burning, etc always using the same refraction indices. In the paper by Gobbi et al. (2007) there is an analysis of the same scheme by using very different values of refractive index and the results are very similar to each other. For these reasons, we strongly believe that the application of this identification scheme is absolutely valid over BoB, since initially we took special care, as pointed out in the manuscript, for excluding any perturb data from the analysis in order to reduce the errors in the spectral AOD, alpha and dalpha retrievals. The fact that the whole set of the observations are within the identification scheme shows strong reliability and quality of our dataset. Furthermore, the close agreement of the results obtained from this identification scheme with the obtained aerosol types as well as with the OPAC retrievals gives credit to the whole of our analysis.

Through a personal communication with Dr. Gian Paolo Gobbi about the use of his scheme he responded to us that “the definition of dalpha is valid for the wavelengths it was computed for (i.e. also for non Aeronet sunphotometers). Indeed AOD measurement uncertainties need to be known to evaluate the propagation of errors affecting alpha and dalpha computations. In this respect, our 2007 ACP paper indicated in sec-

tion 2: "To avoid errors larger than 30%, this work employs only observations of AOT> 0.15." Indeed highly absorbing conditions were not considered in that paper. I would then add that application of the scheme is valid for average aerosol conditions (that is most of the time) as the ones used in the paper (obtained from extended Aeronet climatologies). In fact we specify (comment on Fig. 2): Figure 2 illustrates the sensitivity of the classification scheme to refractive index. Computations indicate some clockwise rotation about the origin of the constant radius curves for increasing refractive index. The effect is much weaker in the case of the curves. For a given point, maximum Rf indetermination is of the order of $\pm$25% for refractive index varying between m=1.33–0.0i and m=1.53–0.003i. At the same time, the fine mode extinction fraction spans a range of the order of $\pm$10%. Within this level of indetermination, the scheme is robust enough to provide an operational classification of the aerosol properties.” The aerosols over BoB are NOT so much absorbing as pointed out by the Reviewer, since the SSA value range from $\sim$0.84 to 0.95 that is comparable with the values obtained over urban environments and locations affected by biomass burning in the Gobbi's et al. (2007) work. Furthermore, in our study the AOD500 values are above the threshold of 0.15 in the 97% of the cases and, therefore, the large errors in the computations are avoided. From all the above, it becomes obvious that the use of this scheme is absolutely valid over BoB even using Microtops-II sun photometer in case of great accuracy in the retrievals.

3. Use of OPAC model to infer the various aerosol types contributing to the columnar burden without having any validation against a concurrent in situ surface measurement is something stretched too far. I seriously doubt the validity of entire section 4.4.

The OPAC spectral AODs as well as the derived alpha values for each BoB sub-region have been compared with the measured spectral AODs (Fig. 14) and the correlations are very good for all the cases. The soot and water-vapor components obtained from OPAC as well as the SSA values are in excellent agreement with in situ observations; we have now revised the manuscript to make our points more clear. Furthermore, the qualitative agreement of the OPAC results, i.e. more absorbing aerosols over northwestern and eastern BoB having a significant soot component, with the aerosol type identification (e.g. large anthropogenic or fine-mode component over these regions) based on AOD-alpha relationship and with the results obtained by the Gobbi's identification scheme gives credit to our analysis. Furthermore, OPAC has been used over the BoB for the computation of aerosol properties in order to estimate the aerosol radiative forcing over the area (Raghavendra Kumar et al., 2011) and their results and discussions are in close agreement with our own results. More specifically their mean SSA value over entire BoB of ~0.88 is in close agreement with our results as well as with the SSA values from previous studies over the region that are cited by Raghavendra Kumar et al. (2011).

The validation of OPAC results with the concurrent measurement is the best suited. Fortunately, the direct measurements of spatial distribution of spectral AOD, scattering coefficient (σsca), absorption coefficient (σabs) and SSA are available during the same cruise campaign discussed elsewhere and, therefore, we would only provide the intercomparison study between the model simulated and the direct measured parameters in order to avoid the repetition. Figure 14 shows the validation of spectral AOD with the simulated one and the satisfactory agreement between them is obvious (authors already explained). The root mean square (RMS) difference between the measured and simulated AODs (at 500nm) was found to be:

Sub region: West BoB North BoB Central East South

RMS 0.021 0.010 0.004 0.012 0.014

In order to validate the other simulated aerosol optical parameters namely σsca, σabs and SSA, we refer to the results presented by Gogoi et al. (2010) in AOGS (2010) annual meeting and proceeding of ISRO-ARFI review report. They found higher values of scattering coefficient over northwestern and south eastern BoB, which are in close agreement with our results. Furthermore, they have observed the higher and

lower values of absorption coefficient across the coastal and central BoB, respectively, which are in excellent agreement with the present results from OPAC simulation (see Table 2). They computed mean values of SSA and asymmetry parameter over the entire BoB of ~0.88±0.05 and 0.71±0.03, respectively, which are again in excellent agreement with our results (0.89±0.05 and 0.70±0.02, respectively). Therefore, the satisfactory agreement between the OPAC simulated (our results) and the measured (Gogoi's results) parameters ensures the validity of the OPAC used in our study.

In order to further validate the composition of aerosols constrained by using the OPAC simulation, we refer the results by Sarin et al. (submitted manuscript) from the same cruise campaign. They report that the mean contribution of water soluble components (nss-SO42-, NO3-, NH4+,) and EC(soot) was 58% and 4%, respectively for the PM10, which is in excellent agreement with our results (59.1±15.54% and 4.77±2.92%, respectively). From all the above we do not agree with the Reviewer about his objection in the validity of section 4.4. In the revised version we discuss in detail the consistency of the OPAC retrievals with other results obtained over BoB during W-ICARB.

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