

Replies to Referee comments

On “Interactive comment on “Spatio-temporal aerosol optical characteristics over the Arabian Sea during the pre monsoon season” by D. G. Kaskaoutis et al.

For Referee A Saha’s short comments/Referee #4

.....found that the polynomial fit is more accurate for the spectral range 340–1020 nm. Other than this result, I don’t see anything new being reported in this study. Most of the results presented in this study have already been published by the authors (details given in the comments below). I disagree with the opinion of other reviewers that this paper is a useful contribution to enhance our understanding of aerosols over Arabian Sea.

Reply: The polynomial fit and its accuracies are the prime focus of this paper. In order to establish this we have tried various exercises with AOD, alpha and coefficients a_1 and a_2 . In order to make the discussions holistic and to help the reader in understanding the heterogeneities in the aerosol field over AS, we presented the AOD and alpha distribution. Furthermore, these graphs are discussed with the view of comparison with previous studies over the region, and more care has been taken in explaining the relative high alpha values, since the previous studies did not highlight it.

(1) The Figures 12 & 13 (in page 22266-22267) are already published in the Journal of Earth System Sci. (Vol. 117, pages: 243-262, 2008) and Atmospheric Environment (Vol. 42, pages: 6816-6827, 2008) by the authors of this study (Kalapureddy and Devara, 2008, Fig. 4a; Moorthy et al. 2008, Fig. 4 & Fig. 7a). So what new scientific information is conveyed by again publishing these two figures?

Reply: See our above response.

Was the same sunphotometer used in all these studies? If yes, then what is the need to show these plots again (they can be referred and cited)? However, if different sunphotometers were used, then did the authors ever attempted to inter-compare the results from the other instruments used during this campaign?

Reply: Yes, the same sunphotometers (two) were used, one for AOD at 340, 440, 500, 675, 870 nm and other for AOD at 1020 nm, column ozone and water vapor (using three UV band and two IR band, one out of them is 1020nm). Unfortunately, this was not reported in the initial submission but it is referred in the revised. We have attempted inter comparison of sun photometers during the campaign and those can be seen here as a Fig. A.

(2) The plots of air mass back trajectories (shown in Fig.11, page 22265 and discussed in section 6.2, pages: 22241-22243) has already been presented by the authors (Kalapureddy and Devara, 2008, Fig. 3a) which also uses the same database. The only difference is that in this study, the authors have examined the

back trajectories for all the days of the cruise, whereas in their previous article, they have presented the back trajectories for representative days only. So what additional information does these mass plots of back trajectories convey?

Reply: In the revised version it is now referred that despite previous publications (e.g. Kedia and Ramachandran, 2008; George and Nair 2008; Kalapureddy and Devara, 2008), the present study provides the air-mass trajectories at 3 distinct altitudes, 500 m, 1500 m and 3000 m. The figure reveals great differences in the air-mass pathways depending on height. For example, on 29 April, although the 500 m air mass presented a marine origin, the 1500 m and 3000 m air masses came from arid areas in Arabia, Iran and Pakistan justifying the coarse-mode (dust) aerosols found on this day. For this reason, the air-mass trajectory figure is given for each day and for three altitudes.

(3) In Page 22227, lines 10-12, the authors have mentioned that the typical error in the AOD estimation is ± 0.03 . Is this absolute error or relative error? How did they arrive at this value? Which wavelength does this error correspond to? A positive bias (+0.03) at 340 nm and a negative bias (-0.03) at 1020 nm can effect the results and conclusions. So, it is very important to know the errors in AOD at each wavelength of the spectral range. Cachorro et al. (2004) pointed out that an inaccurate calibration can lead to a diurnal cycle of the AOD and would result in significant AOD errors at the miscalibrated wavelength. The authors should attempt to perform a detailed error analysis on the measured optical parameters and examine the results on the retrieved parameters (Angstrom exponent, a_1 , a_2) in light of this.

Reply: We have checked the error in AOD estimation with all the wavelengths of our sunphotometers individually and found them over all for all the existing wavelengths to be below or equivalent to 0.03. The larger errors were found in the UV. These errors are the absolute ones. The reviewer well suggested the Cachorro et al. (2004) work, which is very relevant to the present study. In fact we have applied this technique of correction to each wavelength for eliminating diurnal artifact and hence we used the error free data set for the present study. It was found that the application of this technique affects more the AODs at shorter wavelengths (e.g. 380 nm), further strongly affecting the a_1 and a_2 values, but not so much the alpha ones. In the revised version it is noted that only the data where the polynomial fit was simulated better than $R^2 > 0.92$ were used for the analysis.

(4) In page 22227 (lines 8-10), the authors have stated that "This sunphotometer provides the AOD at 6 channels using internal calibration". What does the internal calibration mean? Was the sunphotometer calibrated (direct sun calibration using Langley technique or radiance sphere calibration) before and after the campaign? As it is not mentioned explicitly, one can assume that the

calibration was not done. If it was done, then it would be worth mentioning the results of the calibration (change/drift if any, in the calibration values before and after the campaign) and how these issues has been taken care of.

Reply: We understood that we have not clearly mentioned about using of two sun photometers for 6 AOD and WV and ozone. These instruments are Microtops-II and are calibrated at JPL. The calibration constants are stored in the MT to evaluate AODs for each time of observations based on Langley technique. Frankly, we haven't calibrated our sun photometers just before the cruise but inter comparison results with other sunphotometers were found to be well within the mean variation of other recently calibrated sunphotometers present on board the ship. With the intercomparison results, we won't find any drastic deviation of the calibration values after the cruise. All these issues are noted in the revised version.

(5) In page 22227 (lines 6-10), the authors state that Microtops-II provides AOD at 6 channels, columnar water vapor and ozone. I am not aware of any such Microtops-II instrument that provides so many parameters (would need at least 10 channels). To my knowledge, a single Microtops-II can have a maximum of 5 channels. Did the authors use two different Microtops-II: one which provides AODs at 5 wavelengths (340, 440, 500, 675, 870), and the other which provides ozone retrievals (using 3 UV channels), water vapor (using 936 nm) and AOD at 1020 nm. If this is the case, then the authors should mention this point explicitly in the manuscript.

Reply: See our above response.

How did the authors ensured that the errors (pointing errors, errors due to detector temperature sensitivity, etc) in the two Microtops-II were not substantial enough that could cause a bias in the results. Since Microtops-II is a handheld instrument (manually operated), there could be pointing errors and the magnitude of the errors would be much higher on moving platforms. Were these instruments operated by the same operator during the entire campaign? Since two different Microtops-II could have been used in this study (which I assume might be the case), the pointing errors could be different, and also operator dependent. The authors should justify all these issues.

Reply: We agree with the concern of referee on the errors pointed out. All these are underlined in the revised. Of course there are problems and are aggravate when the observations are from moving platform. But the earlier works suggest that the pointing errors can be tackled some extent by quick triplet observations to pick minimum value out of three. In the text this is explained in detail and great care has been taken for reducing the pointing errors. The temperature sensitivity problem has tackled by exposing the MTs briefly towards sun for the observation and keeping them immediately in the shade but not in Air condition room to eliminate grater temperature

deviations from surrounds. Secondly, the two MT are fixed on wooden slate to take simultaneous observations by a single observer. Finally, the same observer has taken whole dataset for the entire cruise.

(6) In page 22227 (lines 26-28), the authors state that the column water vapor (PW) was observed to be 2.22 ± 0.44 cm (Kalapureddy et al., 2008) and hence the possible water vapor absorption effects at 1020 nm have not been considered. However, on going through the article by Kalapureddy et al. 2008 (Page 3162, Fig.7), it is very clear that the PW varied between 1.0 and 5.0 cm and showed large spatial variability. PW values as high as 4.0 cm occurred over coastal regions of South West India (Arabian Sea region). Values of PW >3.0 cm can influence the AODs at longer wavelengths, which has been ignored in this study (as stated in Page 2227, lines 26-28). This definitely needs some justification?

Reply: In fact, observed PW values are mostly in the range of 1.5-2.8 cm over AS region and the mean is about 2.22 cm. Moreover, only during 18 April we found maximum PW values, around 4cm. Except 18 and 21 April, the PW values are always below 3.2 cm only. These findings are conformed also with the MODIS columnar water vapor data during the cruise period over AS region. In the revised we have taken into account the water-vapor effects at 1020 nm AOD. Using the H₂O absorption coefficient from the SMARTS2.9.2 model, we subtracted the water-vapor contribution. This has a relative small effect on AOD₁₀₂₀ value, but differentiates more the a_1 and a_2 retrievals. As a consequence, these values are different from those reported in the initial version or in the previous publication (Kalapureddy et al., 2009, JGR).

(7) In Page 22227 (lines 18-24), the authors mention about detector temperature sensitivity as a possible uncertainty for the 1020 nm channel and admit that Microtops-II algorithm doesn't take the detector temperature sensitivity into account. Why didn't the authors attempt to re-analyse the data taking the temperature effects into account. Microtops-II does have a built-in temperature sensor which monitors and delivers the internal temperature for each set of measurement. In the absence of this information, the authors can still consider using the onboard meteorological data (presented in Kalapureddy and Devara, 2008). The authors should make use of the temperature information and re-estimate the AOD at 1020 nm, so as to minimize the errors in the AOD estimation.

Reply: The temperature sensitivity, although very small for the reason mentioned above, was taken into account in the revised version. We re-estimate the AOD₁₀₂₀ considering the temperature taken from the onboard meteorological observations. This is another reason for the differences in the a_1 and a_2 values.

(8) In Page 22227 (lines 23-24), it is mentioned that the detector temperature sensitivity can be a problem during very clear conditions (very low AODs), which the authors have ignored. An examination of Fig. 8 (page 22262) shows that the AODs were very low during the period April 30 - May 5. During this period, the AOD at 1020 was ~ 0.05 on most of the days and reached values as low as ~ 0.025 on few instances. So, there can be temperature artefacts in the data during the prevailing clear conditions. This definitely needs some clarification.

Reply: See our above response.

(9) In page 22227 (line 8-10), the authors state that the sunphotometer provides the AOD at 6 channels based on the internal calibration, while in page 22230 (lines 11-13), they say that the AOD values at each wavelength is obtained from the direct beam irradiance measurements via the Bouguer-Beer law and extensive analysis of the errors retrieved via this methodology is described in Kaskaoutis and Kambezidis (2006). It is not clear whether the AODs used in this study was obtained based on the internal calibration or based on the post processing of raw data obtained from the Microtops-II. The authors should clarify this point.

Reply: The AODs were obtained based on the internal calibration only, but were also tested using post processing of raw data obtained from mictorops. This has been clarified.

(10) In page 22230 (lines 13-15), the authors say that the ozone optical depth was omitted in the estimation of AOD, since its contribution to the total atmospheric optical depth can be significant under low turbidities. Why did the authors ignore ozone absorption in the AOD estimation, why they already have at their disposal column ozone values for each set of AOD measurement (they mention this point in Page 22227, lines 6-10). The authors have also published the results of ozone from this campaign in ACPD (Kalapureddy et al. 2008). Then why didn't they make use of this ozone data for the AOD estimation. Omission of the ozone optical depth can cause an underestimation or over-estimation (depending the column ozone values) of the AOD in the Chappius band (440-770 nm), thereby causing artefacts in the AOD spectra.

Reply: We took into account the ozone absorption and we did not ignore it. Unfortunately, the right word is "subtraction" and not "omission". This was corrected in the text.

(11) The authors mention in page 22230 (lines 15-17) that they didn't take into account absorption due to trace gases by assuming that it will be negligible over the oceanic regions. To reiterate this point, they also cited a reference of Kaskaoutis et al., 2007. The paper by Kaskaoutis et al. (2007) gives results of the investigation of ozone and trace gas contribution to the total optical depth over a polluted urban environment (Greece) and not over Arabian Sea. Furthermore,

Kaskaoutis et al. (2007) have clearly stated in their abstract that the corrections due to ozone and trace gases optical depths is necessary for an accurate determination of AOD in the Chappuis band. How could the authors differ in their opinion in this study.

Reply: Despite we subtracted the ozone absorption, we ignored that of NO₂ due its very low values over marine environments. The article by Kaskaoutis et al. (2007) is referred to an urban environment with relative high NO₂ concentrations. We did not differ our opinion about it. However, this sentence has now been modified. Earlier work (Tomasi et al., 1983) have shown that the error due to ignoring NO₂ absorption could be at most 0.006 in AOD wavelengths shorter than 450 nm.

(13) The authors should provide strong justification for ignoring the absorption effects due to ozone, other trace gases (NO₂) and PW in the estimation of AOD. Ignoring the absorption effects due to these species could result in significant artefacts in the AOD spectra.

Reply: All these issues have already been responded. Tomasi, C., Marani, S. and Vitale, V. Multiwavelength Sunphotometer Calibration corrected on the basis of the spectral features characterising particulate extinction and nitrogen dioxide absorption. *Appl. Opt.*, 24, 2962-2970, 1985. This reference very clearly indicates the strong reason for it.

(14) In pages 22235-22236, the authors have compared their results with the literature, however they didn't compare their results with those obtained using similar instrument during the same field campaign as reported by other investigators (Kedia and Ramachandran, 2008). Using shipborne sunphotometer measurements and MODIS satellite data during the ICARB campaign, Kedia and Ramachandran (2008) have reported an AOD of 0.24 and Angstrom exponent of 1.0 over Arabian Sea. These results are very much similar to that obtained by the authors in this study. However, it is surprising that this paper (which appeared in the ICARB special issue of the *Journal of Earth System Sci.* (Vol. 117, 2008) has not been cited (could be an oversight). Furthermore, the temporal variation of the spectral AODs shown in Fig. 8 (page 22262) is very similar to that presented by Kedia and Ramachandran (Page 379, Figure 2 right panel) and Kalapureddy and Devara (Page 6818, Fig. 2). So what additional information does the authors want to convey from Fig.8 which is already published by them and other colleagues/investigators?

Reply: The suggested references are added which were missed in our earlier manuscript. This paper introduces temporal information of high-temporal resolution Microtops-II data (~ 10 min.) of all six AODs. Kedia and Ramachandran paper used indigenous instrument in which they change

optical filters to record irradiance information and later in post process they estimate AOD values. They provided the daily mean AOD₅₀₀ values comparing them with the MODIS observations. Moreover, their instrument had a higher field of view of ~10°. The spectral AOD variation, even in high temporal resolution, is the advantage of the present work.

(15) Several instruments were used during the ICARB shipborne campaign and surely several sunphotometers were in operation during this study (from the series of papers published from this campaign). One such example (mentioned in the previous comment) is the paper by Kedia and Ramachandran, (2008), which also presents results from shipborne sunphotometer measurements from the same campaign. Did the authors ever attempted to inter-compare the results from the other onboard sunphotometers? The authors also have at their disposal a Cimel Sunphotometer (AERONET instrument) which has been in continuous operation in Pune (India) since October 2004 and one of the co-authors is the PI for this instrument. The authors could have attempted to compare the AOD retrievals from Microtops-II (used in this study) with Cimel before or after the campaign? It would be worthwhile inter-comparing the AODs obtained from various instruments before presenting the results.

Reply: Yes, inter comparison has been made and the results are given in the above figure A.

(16) The ICARB shipborne campaign was conducted over Arabian Sea (AS), Indian Ocean (IO) and Bay of Bengal (BoB) and the authors have definitely made measurements in all these oceanic regions (as reflected in their publications: Kalapureddy and Devara, 2008; Kalapureddy et al., 2008, 2009; Moorthy et al., 2008). Then why in particular, did they use the data from AS only? Why didn't they extend their analysis to BoB and IO region and compare the results with that over AS?

Reply: This is true. Initially, we used the whole data set covering IO, BOB and AS for Atmos. Environ. paper. But later, we decided to confine to the AS for two reasons: i) explore the pre-monsoon AS region vigorously due to its importance and implications on the forth coming monsoon, and ii) due to keep morals with whole team understandings on using cruise data, we confined only to AS. However, the main results, AOD, alpha, are compared with those obtained over BoB.

The authors have a very good database with several instruments at their disposal. It is highly recommended that the authors should try to inter-compare the retrievals with those obtained (i) using other instruments (other onboard sunphotometers, Cimel, etc) and (ii) over other oceanic regions (BoB, IO). Also, a detailed error analysis of the measured optical parameters (AOD) and its effect on the retrieved parameters (Angstrom exponent, a_1 , a_2) should be included and discussed in the manuscript.

Reply: For the present study we are only confine to AS region due to the above reason but the inter comparison point has been well taken. The detailed analysis of the errors is already given in a separate section of the manuscript.

It is very much important for the authors to quantify the error in the measurement of the AOD at 1020 nm with high accuracy, because the authors have found that the polynomial fit is more accurate for 340–1020 nm wavelength range, as compared to the 340–870 nm range (this being one of the major conclusions of this paper). Since the inclusion and exclusion of the 1020 nm in the data analysis significantly affects the results, it is very important to have a very accurate measurements of AOD at this wavelength. In the absence of instrument calibration and detailed error analysis in the AOD estimation, the results presented could be an artefact of the data processing.

Reply: We strongly agree with this. That is what we would like to state from whole of this work to the interested researchers who are on this line. For this reason, in the revised manuscript we have taken into account the water-vapor and temperature-sensitivity effects on the AOD1020, also applying the Cachorro et al's. (2004) method for calibrating the AODs.

Overall, this study lacks an original and compelling experimental design. Reports on the spatiotemporal characteristics of aerosols over Arabian Sea is not new (and part of the results presented in this paper are already published by the authors), and I find the reported results are too routine to warrant publication in ACP in its current form. I therefore suggest a major revision of the manuscript, taking into account all the issues discussed above.

Reply: We prefer to disagree. The study is original, though the report of spatio-temporal characteristics of aerosols have been addressed by several investigators in the past and also being investigated even today by the global aerosol scientific community. This is essentially due to the very fact that large heterogeneity is the hallmark of aerosols, and that is what makes it region and season specific. We believe and also are encouraged by the 4 earlier review reports that this study has quite a lot of scientific value. Such an exhaustive attempt covering the entire Arabian Sea, spanning from 9N to >20N, and 77E to 58E, with its inherent heterogeneity, has not been attempted in any earlier field experiments. In the modified paper we considered all the reviewer's critical comments. We have taken into account the Cachorro et al. method, the water-vapor and temperature sensitivity effects, we avoided repetitions and pointing our discussion to the new results. These are the a_1 and a_2 spatial distributions, which are discussed with the help of those for AOD and alpha, a detailed analysis of the errors, uncertainties and differences revealed in a_1 and a_2 using different spectral bands and the inclusion of the α vs $d\alpha$ figure, according

to the classification scheme of Gobbi et al. (2007). Despite the numerous publications conducted over AS during ICARB and previous campaigns (e.g. INDOEX, ARMEX) the above topics are investigated for the first time.