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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Effects of absorbing aerosols in cloudy skies: a satellite study over the Atlantic Ocean" *by* K. Peters et al.

K. Peters et al.

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We thank the reviewer for the specific remarks, which helped us considerably improving the manuscript.

Specific remarks:

Pg 20854 Line 13, 16. Should not use symbols in abstract, use the name of the parameter.
Thank you, we follow the remark of the reviewer.

Pg 20854 Line 17: Increasing amount of absorbing aerosols? How is the amount of absorbing aerosols defined?
Indeed, this was just an imprecise wording in the previous version of our manuscript. We do not directly sample for the amount of absorbing aerosols, but for the absorption effect of the observed aerosol(mixture). We have modified this in the revised manuscript.

Pg 20855 Line 13: Provide reference for statement quoting 0.8 as SSA lower limit.
Thank you for hinting at this. As this was just seen as an illustrative example, it can just as well be removed from the manuscript. We adopted the manuscript in this regard.

Pg 20855, Line23: '...surface albedo and aerosol single scattering albedo.'
We have included this clarification in the modified manuscript.

Pg 20856 Line6: insert 'of the aerosol layer in the absence of clouds' after 'negative TOD DRF'
Thank you, the previous formulation does not allow for immediate understanding. We have adopted the formulation of the reviewer in the revised manuscript.

Pg 20856 Line19: use 'long timescales', instead of 'long temporal timescales' (redundant)
Indeed, this is redundant. We have modified the manuscript accordingly.

Pg 20856 Line23, what is the reference wavelength for alpha ?
Alpha is computed from the broadband shortwave TOA fluxes from CERES. We have included a short note in the revised manuscript.

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Pg 20859 Line13: What determines the variability of the spatial resolution ? Is the spatial resolution identical for the three (UV1, UV2, and VIS) OMI sensors ?
Thank you very much for this comment. Indeed, the description of OMI was too short in the original manuscript. Referring to the literature describing OMI (especially Levelt et al., 2006), we added additional information to the revised version of the manuscript. The variability in the spatial resolution happens by instrument design (this is due to pixel distortion at the edge of the swath). OMI has two channels (UV and VIS), with the UV channel divided into two subchannels (UV-1 and UV-2). The spatial resolution of UV-1 is reduced by a factor of two with respect to UV-2, which has the same spatial resolution as the VIS channel. OMI also offers different viewing modes (global observation, spatial zoom-in and spectral zoom-in), but this is not relevant for our study, where we only use data from the "global observation" mode. We have modified the manuscript accordingly and have added the reference to Levelt et al. (2006).

Pg 20859 Line 14: What aerosol properties are derived from OMI in cloudy scenes ? The Aerosol Index is not an aerosol property.
The reviewer is right – our formulation was misleading. As far as we are aware of, OMI does not retrieve aerosol properties in cloudy scenes. The UV-AI can be seen merely as an indicator of bulk aerosol absorption in cloud scenes. We have adopted the manuscript accordingly.

Pg 20859 Line 24: Provide an adequate reference for the actual OMAERUV product used in the analysis.
Indeed, a correct reference is missing here. We have included reference to Torres et al. (2007) in the revised manuscript.

Pg 20859 Line 25: Which AI product is used in the analysis ? According to the wavelength definition the authors seem to be referring to the OMAERUV product.

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Mention the full product. The correct wavelengths are 354 and 388 and the most appropriate reference is Torres et al, [2007].
Thank you very much for this important comment. The description of the AI product used has been included in the revised manuscript from the previous comment. In the reference we provided (the OMI ATBD Document), it is stated that the UV-AI is calculated from 342.5 and 388 nm. Checking of the reference provided by the reviewer and other references, we have now changed the manuscript in accordance with the reviewer's suggestions.

Pg 20860 Line7: Does OMI detect aerosols in clouds ? If true provide reference for this statement. This is stated in Torres et al., 2007, section 3.1 "Aerosol cloud interaction". "...A unique UVAI property is the capability of detecting aerosol absorption regardless of the overall scene brightness. The UVAI clearly identifies absorbing aerosols even when intermingled with or above clouds. No other passive remote sensing technique can detect aerosol-cloud..."
The correct reference is now given in the revised manuscript. We have removed the reference to the OMI-ATBD.

Pg 20860 Line 17: According to what theory should the UV-AI be close to unity for very low AOD values ? Provide reference for this statement.
Thank you very much for highlighting this unfortunate mis-wording. We did not mean to say "unity", but "zero". According to conceptual theory, the UV-AI should be close to zero for low AOD values, since the radiative properties of an atmosphere containing low amounts of aerosol come close to those of a pure Rayleigh atmosphere. This results in low UV-AI values.
We have corrected the manuscript in this sense.

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Pg 20861 Line 25: which two years ?
Indeed, this information is missing at this point. The time frame is 2005 - 2006 and we have added this information in the manuscript.

Pg 20862 Line 4 : In the context of this analysis one cannot use 'absorbing aerosol' in a generic way to include desert dust absorption and black carbon absorption. Aerosol absorption is a wavelength dependent process that has a very different effect on the overall energy content in the range 0.3 - 5 microns depending on the aerosoltype. For biomass burning aerosols the broadband absorption may be significant, but for dust it is not.
Thank you for this detailed comment on aerosol absorption characteristics. We are very well aware of the fact, that biomass-burning aerosol has a very different spectral-absorption signature from dust. Nevertheless, we are interested in characterising the bulk absorption effect of an observed aerosol mixture in the broadband shortwave part of the solar spectrum. This implies derivation of the radiative effect of dust aerosol in the visible part of the spectrum, even though it's radiative effect is much larger in the UV.
We included some phrases on the spectral absorption characteristics in the manuscript. Further justification for our method is given in the reply to the next but one comment.

Pg 20862 Line 18: What MODIS cloud product is used to characterize a scene as overcast ? MODIS cloud properties are affected by the presence of absorbing aerosols above the clouds (Wilcox et, al 2009). The effect is different for dust aerosols (reduced absorption in the vis-near IR) than for biomass burning aerosols (strong absorption in the entire vis near-IR). How does the wavelength dependent effect of

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aerosol absorption above clouds affect the criteria for the threshold used to identify overcast scenes ?

Thank you for this valuable comment. The information on the data used is given in section 2.1 describing the MODIS dataset. But indeed, a reference to this section should be given in this part of the text.
We have already referenced Wilcox et al., 2009, but in a different context (usage of microwave LWP from AMSR-E instead of LWP retrieved from the visible by MODIS). To our knowledge, Wilcox et al. do not address the effect of absorbing aerosols with respect to cloud masking. Steven Platnick (pers. comm.) states, that there should not be a problem identifying heavy smoke as cloud. Please note that precisely because of the influence of aerosol absorption on MODIS retrievals of cloud microphysical properties, we do not use any such product from MODIS, but just the cloud fraction, which can be considered largely unaffected by the aerosol absorption. For the liquid water path, we rather use the microwave retrieval from AMSR-E, which is much less sensitive to aerosol absorption than the vis-near IR retrievals by MODIS.
Nevertheless, we have put a considerable amount of effort into checking our analysis again. We found, that the data going into the analysis did not fulfill the requirements. The analysis was repeated, this time utilizing the Quality Assurance and Cloudmask flags. Looking at single cases, we found that pixels being flagged as "Uncertain" by the cloudmask are put to 100% cloud fraction. This is an obvious error source. In the repeated analysis, we then used only MODIS pixels which are determined to being "Cloudy" by the "Cloud_Mask" and "single layer liquid water cloud" by the "Cloud_Quality_Assurance" flag. We are confident, that this resolves any issues concerning cloud fraction retrieval. We have adopted the manuscript accordingly.

Pg 20862 Line22: The assumption that aerosol characteristics are similar on a similar

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time scale is hard to justify. The area TNEA is affected by the presence of both dust and biomass burning aerosols from November through early march. The rest of the year the aerosol load is predominantly dust. The most northern part of TSEA is also affect by the presence of dust and biomass burning mixtures.

Indeed, this is an important point. To address this issue, we have investigated the distribution of anthropogenic and natural aerosol AOD for the region and time frame of interest. This is done using the analysis of anthropogenic versus natural fractions of AOD as described in Bellouin et al., 2005, using MODIS collection 5 data from EOS-Aqua.

This dataset mainly uses (over oceans, but in this study we are anyway not interested in land areas) the MODIS retrieved accumulation mode fraction (particles smaller than 1 micron) to discriminate between natural and anthropogenic aerosols. Particles contained in the accumulation mode are treated as anthropogenic (e.g. biomass burning aerosols) and others as natural particles (e.g. mineral dust aerosols). If mixtures of biomass burning and mineral dust aerosols are identified, the accumulation mode fraction of the retrieved aerosol optical depth is treated as anthropogenic.

By using this dataset, it is thus possible to discriminate between large and small particles. For the region of interest, for absorbing aerosol, this mainly leads to a discrimination between mineral dust and biomass burning aerosols.

To analyse the distribution of aerosol types (anthropogenic(small) and natural(large)) with respect to region and season, we divide the dataset in half: one half contains all measurements with an anthropogenic fraction larger (HIGH) and the other half all measurements with an anthropogenic fraction smaller (LOW) than 50%. We are aware that this is rather crude, but it gives a good impression of the observed distributions of aerosol types in cloudy scenes. Four different fractions were defined and analysed (region- and seasonwise):

(1)**Fraction of cloudy scenes with HIGH anthropogenic fraction.** (This is the ratio of the amount of HIGH-scenes to the sum of all (HIGH and LOW) scenes identified.) Fig. 1

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(2)**Fraction of anthropogenic-absorbing-aerosol identifications in cloudy scenes with respect to all absorbing-aerosol identifications in cloudy scenes.** (This is the amount of HIGH-scenes with UV-AI $>$ 0.7 divided by the sum of all scenes with UV-AI $>$ 0.7.) Fig. 2
(3)**Fraction of absorbing-aerosol identifications (UV-AI $>$ 0.7) in cloudy scenes with HIGH anthropogenic fraction.** (This is similar to Table 2 in the manuscript.) Fig. 3
(4)**Fraction of absorbing-aerosol identifications (UV-AI $>$ 0.7) in cloudy scenes with LOW anthropogenic fraction.** (This is similar to Table 2 in the manuscript.) Fig. 4

Conclusions from the four different studies (special interest is given to regions TNEA and TSEA, as these are of greatest importance with respect to absorbing aerosol occurence):

(1)Here we find, that region TSEA is the only one being subject to significant anthropogenic aerosol pollution, with the anthropogenic fraction being about 10% from December through May, about 48% from June - August and about 35% from September - November. For the other regions, the anthropogenic fraction rarely exceeds 10%: for region TNWA from March-May (≈12%) and from September-November (≈15%) and for region TSWA from September-November (≈17%). Averaging over all seasons shows the anthropogenic fraction being about 6% for region TNEA, 10% for regions TNWA and TSWA and about 33% for region TSEA.

(2)For region TNEA, we find that absorbing aerosols retrieved in this region are mostly natural. There is no season where the anthropogenic fraction of absorbing aerosols is above ≈7%. This leads to the conclusion, that the vast majority of absorbing aerosols consists of large particles (such as mineral dust) throughout the year.
For region TSEA we find a distinct seasonal cycle in absorbing aerosol type: from De-

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cember through May, mostly large absorbing aerosol particles occur and from June through November, the absorbing aerosol occurrence is dominated by small particles. This is in accordance with expectations, because the biomass burning season in middle to southern Africa takes places during this time of the year.
Region TNWA shows elevated levels of small particle absorbing aerosols from March - May. This is most likely due to intensified biomass burning in the insular Caribbean during the local dry season (Robbins et al., 2008).
Region TSWA is found to have elevated levels of small particle absorbing aerosols from June through November. This is most likely due to biomass burning aerosol being advected from Africa.
Averaging over all seasons shows, that the anthropogenic fraction of absorbing aerosols is lowest in region TNEA ($\approx$3%), about 10% for TNWA, $\approx$18% for region TSWA and $\approx$60% for region TSEA.

(3)For region TNEA, we find that small size particles are found to be mostly scattering (UV-AI $<$ 0.7) for all seasons. It is only from December through February where the absorbing fraction is above 15%.
On the contrary, anthropogenic aerosol is found to be mostly absorbing (UV-AI $>$ 0.7) in region TSEA for all seasons except from December through February. From June - August, absorbing aerosols make up almost 90% of all measured anthropogenic aerosols. Thus, the distinct seasonal cycle of biomass burning can be clearly depicted for this region.
For the region TSWA, we find elevated absorbing aerosol fractions from June through November. This again most likely due to biomass burning aerosol advection from Africa.
In region TNWA, the absorbing aerosol fraction of anthropogenic aerosols is elevated from December through May (December-February: $\approx$18%, March-May: $\approx$30%). This is again most likely due to intensified biomass burning activity during the local dry season.

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Averaged over all seasons, we find that anthropogenic aerosols are mainly absorbing in region TSEA (≈80%) and constitute a considerable fraction in region TSWA (≈45%). Region TNWA and TNEA show fractions of ≈23% and ≈18%, repectively.

(4)Here, the regions under consideration show a constant partitioning between mostly scattering (UV-AI < 0.7) and mostly absorbing (UV-AI > 0.7) aerosols from December - May (≈15 - 20% of observed aerosol is found to be absorbing). From June - August, elevated fractions of natural absorbing aerosol is found for regions TNWA, TNEA and TSEA.
For the regions TNWA and TNEA, this is mostly due to dust-aerosol transport from northern Africa. For the region TSEA, this elevated fraction of natural absorbing aerosol might be due to the algorithm design in Bellouin et al., 2005. Biomass burning aerosol may also have a significant fraction being larger than 1 micron (e.g. Remer et al., 1998). So by design, the algorithm described in Bellouin et al., 2005 defines this aerosol as natural, although it is of anthropogenic origin.
For the time period from September through November, the only region showing elevated fractions of natural absorbing aerosol is TNEA.
Evaluation of annual averages illustrates, that region TNEA is the one being subject to the highest fraction of natural absorbing aerosols.

Additionally, we computed the multiple regression (Eq. 1 in the manuscript) for HIGH and LOW anthropogenic aerosol fractions (see Figs. 5 and 6).
We find different results for HIGH and LOW anthropogenic aerosol fractions. For all seasons, the change of local planetary albedo with AOD is less positive or negative for scenes with an UV-AI > 0.7 than for scenes with an UV-AI < 0.7. The results for scenes with an UV-AI > 0.7 are associated with a higher uncertainty. This is due to the low number of measurements. (Compare to the results of point (1) above). Comparing the results obtained when performing the regression for all measurements (Fig. 3 in the revised manuscript), it is evident that the results for the coefficient a_2 are dominated

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by the results for LOW anthropogenic AOD fractions in all regions except for region TSEA. Here, the coefficients represent more or less a mean value between the results obtained for LOW and HIGH anthropogenic AOD fractions. This is due to the few number of measurements of anthropogenic aerosols in regions TNWA, TNEA and TSWA, where the effect of natural aerosols outweighs the effect of anthropogenic aerosols.

From this analysis of the measurements, we conclude that our assumption of approximately similar aerosol characteristics in one region over the time frame under consideration (multiple seasons) is justified. Although it is evident that the calculated radiative effects of natural and anthropogenic aerosols are somewhat different for the regions TNWA, TNEA and TSWA, it is sufficient to characterise the radiative effects of absorbing aerosols by the mean value over all measurements. This holds, because the fractional amount of anthropogenic aerosols is negligible compared to natural aerosols, making the radiative properties of anthropogenic aerosols negligible with respect to the bulk radiative effects of the aerosol mixture.
The same holds for the region TSEA, although the results between LOW and HIGH anthropogenic fractions do not differ so much from each other than for the other regions. This is most likely due to the higher anthropogenic fraction in region TSEA compared to that of the other regions.
We have added a somewhat shorter summary of these findings to the revised version of the manuscript.

Pg 20862 Line24 (19): The UV-AI can certainly be used to identify the presence of UV-absorbing aerosols. In terms of what parameter is the aerosol characterized using the UV-AI ? The magnitude of the UV-AI depends on multiple parameters (AOD, height, particle size) as shown in Herman et al (1997). The magnitude of the AI is also affected when aerosols are above a highly reflective background as clouds, and the effect is different depending on the aerosol types [Torres et al,1998].
Thank you very much for this important comment. The observed aerosol is charac-

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terised with respect to it's absorptive properties in the UV, with this being an indication whether an observed aerosol mixture absorbs incident solar radiation in the broadband spectral region or not. With the available information on an observed scene, this is the only way the UV-AI can be used to (qualitatively) characterise aerosol properties. We by no means attempt to perform a quantitative derivation of aerosol properties from the UV-AI, since this is almost impossible, like the reviewer mentions. We also already mention this in section 3 of the revised manuscript.
We have added a few words to the manuscript to make this clearer.
Thank you very much for pointing to the work of Herman et al., 1997, which we did not know before. We have added reference to this work where appropriate.

Pg 20862 Line 25: How is the absorbing aerosol mass defined ? The MODIS AOD is probably related to the absorbing aerosol mass in the case of biomass burning aerosols since BC absorbs in the visible. In the case of dust one cannot assume that the MODIS AOD is related to aerosol absorption mass, dust absorption is significantly lower in the visible.
The reviewer completely correct, and this was merely an imprecise wording in the previous manuscript. We are not able to quantify absorbing aerosol mass, but derive a statistical relationship between the local planetary albedo, cloud liquid water path and AOD.
We have changed this in the manuscript.

Pg 20863 Line 10: The 0.7 threshold is too simplistic. The 0.7 value is adequate for cloud-free scenes. For aerosol above clouds a different threshold is needed. During the analysis, we have tried several thresholds for the UV-AI (0, 0.7 and 1) and have found the value of 0.7 as being the best fit between being able to sample for aerosol absorption (from conceptual theory everything above 0 should be absorbing, although scenes with an UV-AI below 0.5 may contain a ground signal, nonabsorbing aerosols

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or noise (Herman et al., 1997)) and having a sufficient amount of measurements available for the statistical analysis. We therefore state the threshold of 0.7 being sufficient for this analysis. A short phrase is added to the manuscript to underline this.

Pg 20864 Line 9: should read 'TSEA and TNEA'
Thank you very much. We have corrected this in the revised manuscript.

Comment on the restructuring of some parts of the paper

From February 22-26, the first author took part in the workshop "Advanced Scientific Writing", held by Dallas Murphy and Jochem Marotzke, at the Max Planck Institute for Meteorology in Hamburg. The input received there lead to the need of rewriting and restructuring some parts of the paper in order to improve readibility. These modifications included (1) a shortening of the "abstract", (2) restructuring of the "introduction", (3) giving the description of the UV-AI it's own section, (4) restructuring of the "methods", (5) restructuring and refinement of the "summary and conclusions" as well as (6) rewriting of several sentences to enhance clarity.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 20853, 2009.

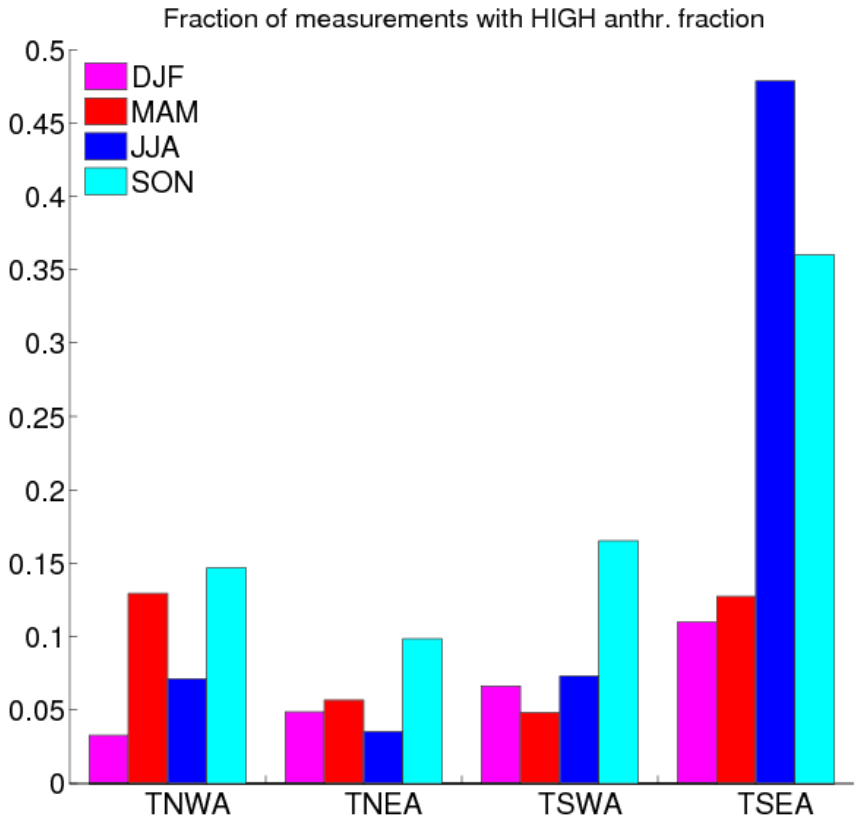

Fig. 1. Fraction of cloudy scenes with HIGH anthropogenic fraction. This is the ratio of the amount of HIGH-scenes to the sum of all (HIGH and LOW) scenes used.

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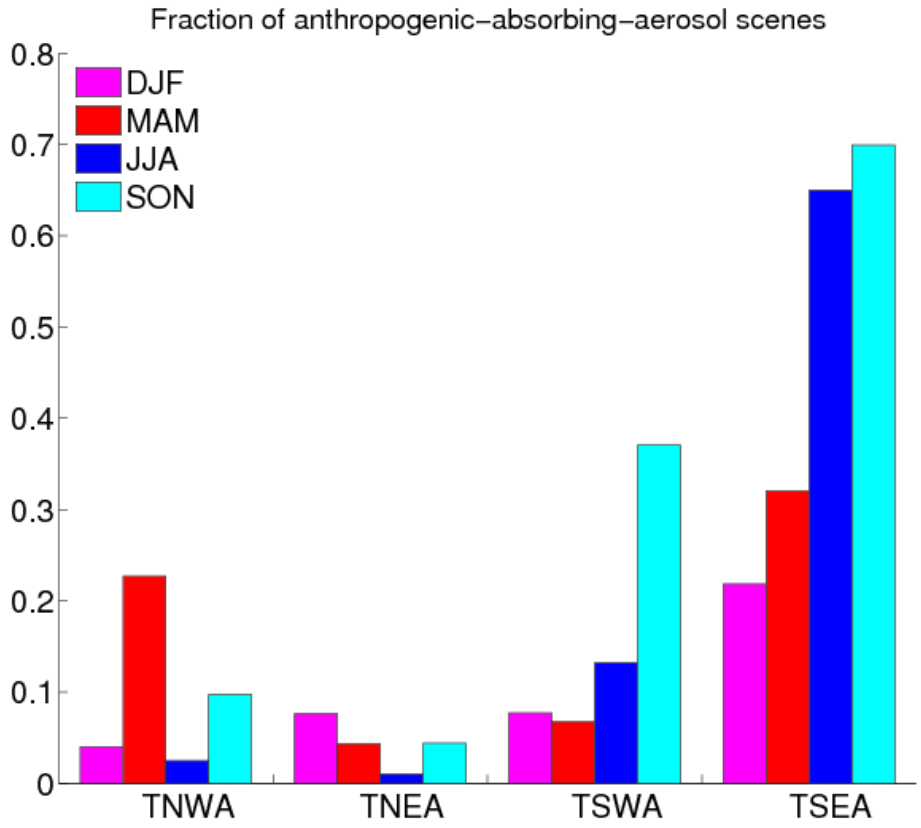

Fig. 2. Fraction of anthropogenic-absorbing-aerosol identifications in cloudy scenes with respect to all absorbing-aerosol identifications in cloudy scenes. This is the amount of HIGH-scenes with UV-AI > 0.7

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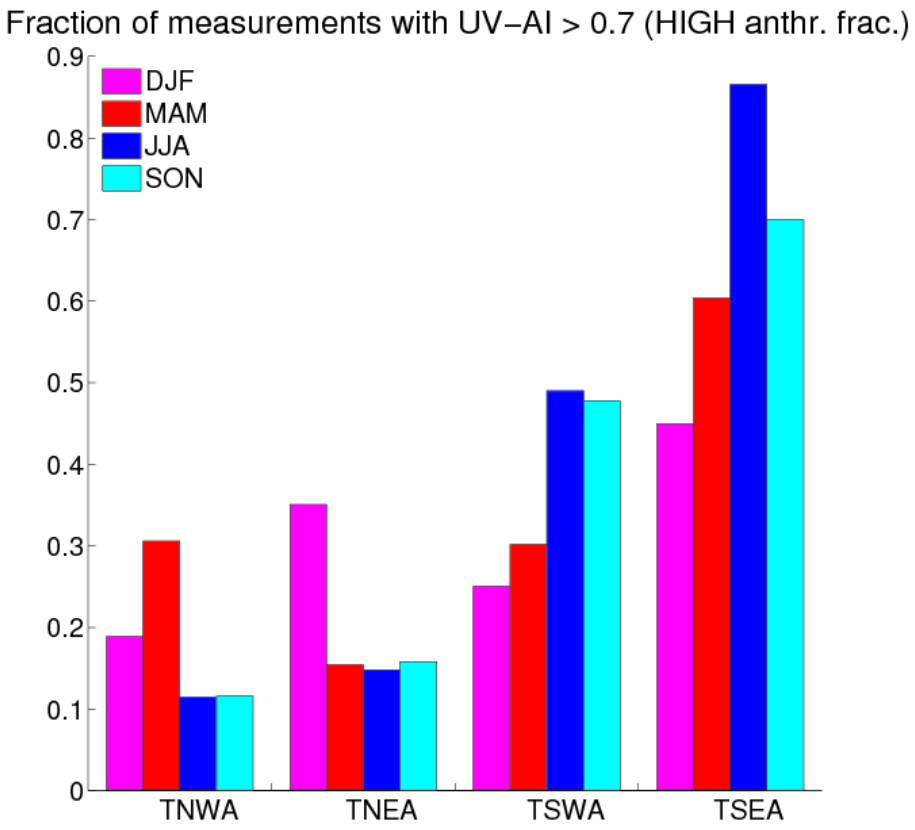

Fig. 3. Fraction of absorbing-aerosol identifications (UV-AI > 0.7) in cloudy scenes with HIGH anthropogenic fraction. This is similar to Table 2 in the manuscript.

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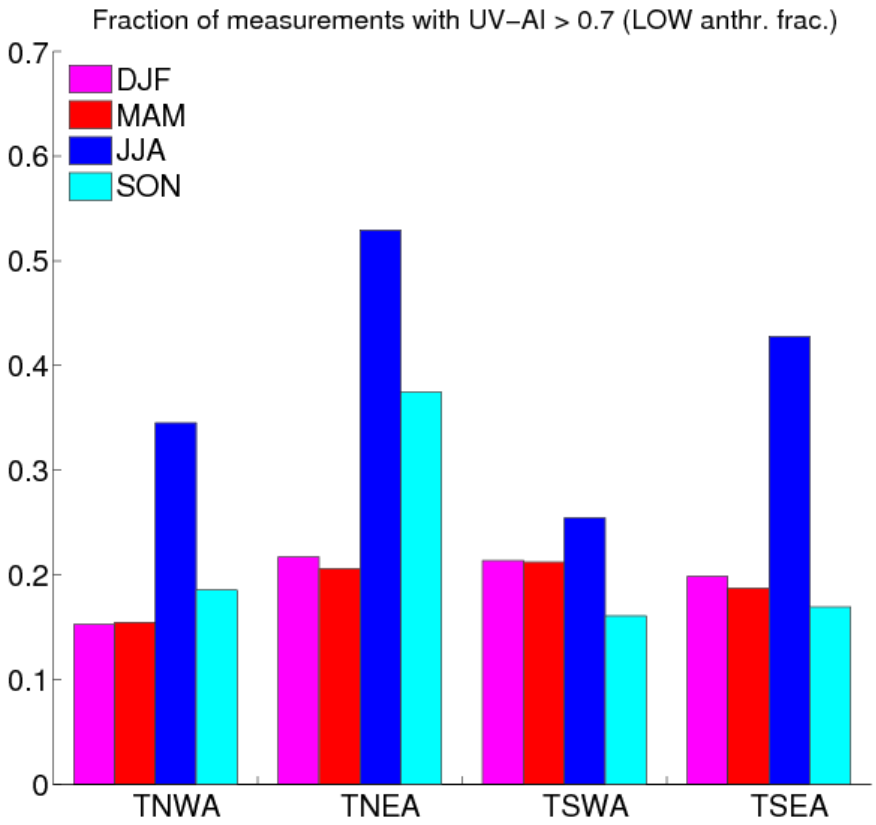

Fig. 4. Fraction of absorbing-aerosol identifications (UV-AI > 0.7) in cloudy scenes with LOW anthropogenic fraction. This is similar to Table 2 in the manuscript.

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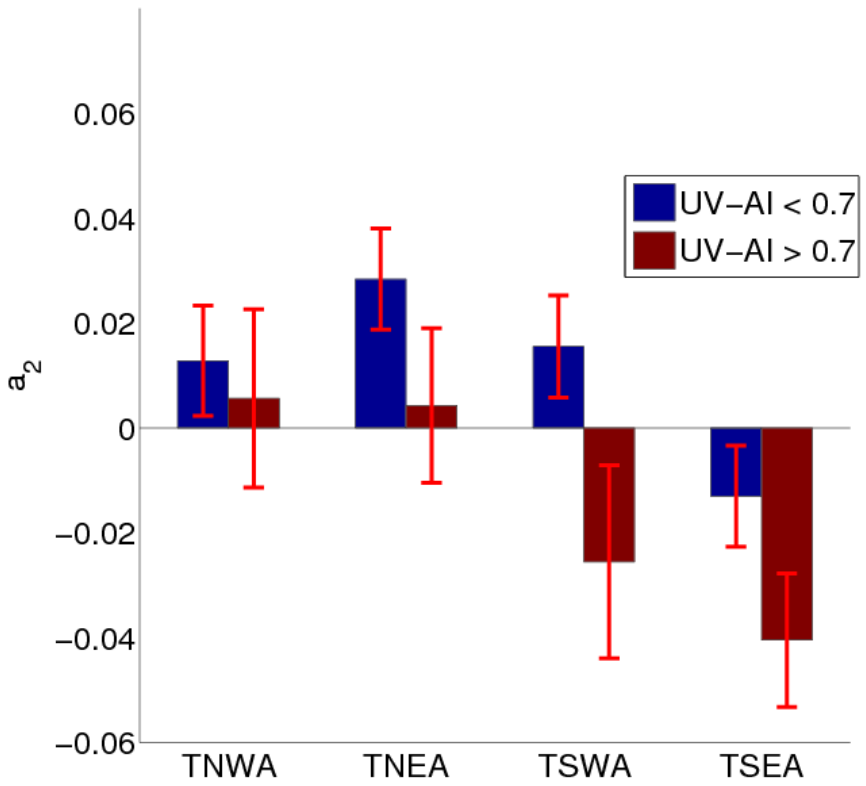

Fig. 5. Result from the regression analysis performed with scenes yielding HIGH anthropogenic AOD fractions: coefficients a2 and respective standard deviations for the selected regions.

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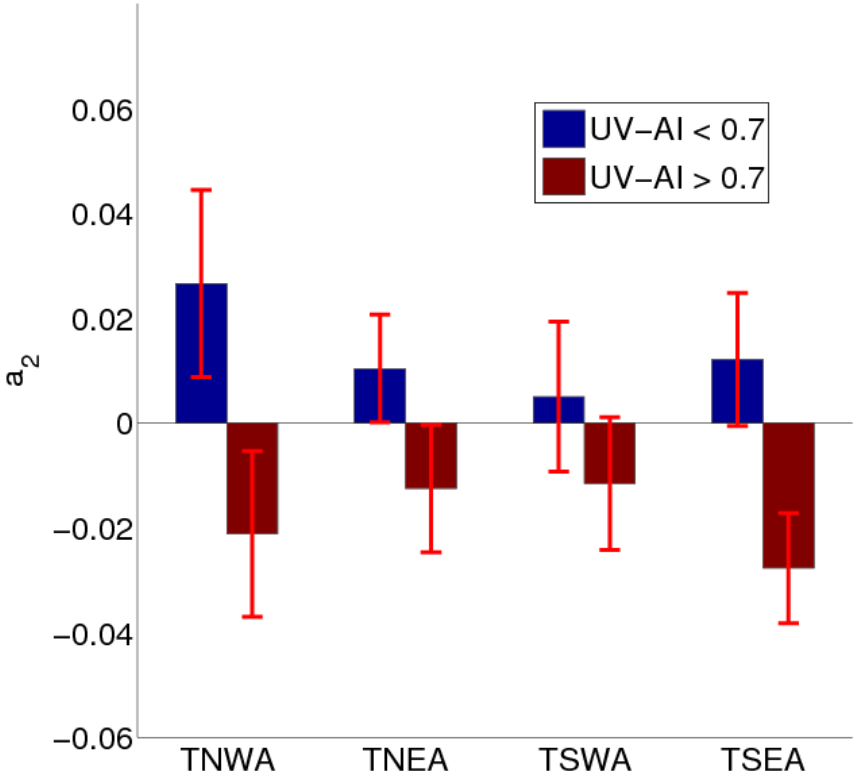

Fig. 6. Result from the regression analysis performed with scenes yielding LOW anthropogenic AOD fractions: coefficients a2 and respective standard deviations for the selected regions.

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