

Interactive
Comment

***Interactive comment on* “Satellite-based evidence of wavelength-dependent aerosol absorption in biomass burning smoke inferred from ozone monitoring instrument” by H. Jethva and O. Torres**

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Response to the Anonymous Reviewer # 2:

We are thankful to the anonymous Reviewer #2 for his/her constructive and critical comments on our manuscript. Below, we have addressed the issues and concerns

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raised by him/her. Let us know if you are satisfied with our response.

Restructuring of the content of the manuscript Pg 7295: line 10-26: Here we provide a short description of the standard OMAERUV aerosol algorithm. This is well-documented by Torres et al. (2007) and also accessible in the Algorithm Theoretical Basis Document (ATBD) available on

http://eosps0.gsfc.nasa.gov/eos_homepage/for_scientists/atbd/docs/OMI/ATBD-OMI-03.pdf

Pg 7297, line 9-22: Here we describe the procedure of OMI-AERONET collocation which we think is appropriate and therefore left unchanged.

Section 4: Section 4 has been re-organized in the revised text. It has two subsections, first that discusses the results of near-UV sensitivity analysis to the spectral absorption, and second that provides ground-based evidences on the spectral aerosol absorption based on which a most reasonable value AAE was selected for the new OMAERUV retrieval.

P. 7303, lines 10-18 : The content have been re-written as “A close examination of the look-up-tables generated using gray and colored aerosol models tells that the difference between them is marginal at lower AOD (<0.5 at 388 nm) values, which becomes larger at moderate to higher AODs (>1.0 at 388 nm). This results in a marginal change in the retrieval of AOD at lower aerosol loading with the use of colored aerosol models. The change in the magnitude of AOD, however, is significant at larger values of AOD. In this condition, the assumption of wavelength-dependent aerosol absorption in the near-UV region produces retrieval of AOD and SSA that are in better agreement with the AERONET data.”

Lines 19-28: Modified in the revised manuscript. P. 7306, lines 2-5: These sentences have been re-written in the revised manuscript. Pg 7292: l. 19-21: "present study (...) integrated measurements." This statement implies that OMI coupled with

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AERONET can determine aerosol type (i.e., black (BC) or brown carbon (BrC)) on a single-measurement basis. In the presented results, the improvement in the OMI retrieval when using the BrC absorption w.r.t the use of the BC absorption becomes apparent thanks to statistics (more OMI AOD fall within the uncertainty range in Fig. 4 than in Fig. 2). In short, I think the statement is too optimistic: I would not say that OMI (+AERONET) can distinguish BrC from BC based on the results presented here. OMI (+AERONET) does, however, provide evidence that BrC is present in biomass burning smoke. This is aside from the fact that smoke particles are probably complex internal and/or external mixtures of BC, BrC, secondary aerosols, and numerous other compounds. A statement mentioning the complexity of (smoke) aerosol composition should be added to the paper.

We agree with the reviewer here. Since the spectral dependence of aerosol absorption of OC differs significantly from that of pure BC in the near-UV region, the OMI measurements can identify the presence of OC in the biomass burning aerosols.

The last sentence of the abstract has been modified as: "Together with suggesting a significant improvement in the retrieval of aerosol properties from OMI, the present study demonstrates the sensitivity of near-UV measurements (or Aerosol Index) to the chemical composition of aerosols. This capability can be further explored for the identification of BC and OC in biomass burning aerosols from OMI measurements."

Pg 7294: line 5: Is (Torres et al., 2007) the only relevant reference here? Herman et al. (1997) has been added.

Line 8: "SA" Change to "South America" Changed to South America.

Pg 7295 Line 2-4 : "Secondly, (...) cloudy conditions." This is an intriguing statement that requires more explanation and references to literature Explanation on the near-UV radiative interactions: In the clear-sky condition, the radiative interactions between the aerosol layer and Rayleigh scattering under that layer produce the TOA near-UV signal (or Aerosol index) which is sensitive to the aerosol absorption (single-scattering

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albedo), aerosol optical depth, and height of aerosol layer. In cloudy conditions, where an aerosol layer is elevated above the low-level cloud deck, the higher reflectance reflectance emanate from cloud top results in further enhancement in the near-UV signal which leads to larger Aerosol Index. Torres et al. (1998, 2007) has been cited here.

Line 6: Please cite the spatial resolution of OMI here (numbers). The spatial resolution of OMI is 13x24 km² at nadir. It's added here in the manuscript.

Line 6: "SCLamachy" Should be written "SCIAMACHY" Corrected.

Line 7: I. 7. OMI is a satellite instrument; it does not "derive". Please rephrase the sentence The statement has been corrected to "OMI retrieves..."

I. 11. Please add a reference to the OMI or TOMS AI algorithm here Reference to the OMI and TOMS AI has been cited before in the text.

Pg 7296: I. 13-14: "the OMAERUV-AERONET comparison was reasonable." What is reasonable? Please be more quantitative. This sentence is re-written as: "Torres et al. (2007) compared the OMI-derived AOD with that of AERONET direct sun measurements and found that under minimum cloud interference the OMAERUV-AERONET comparison was reasonable (root-mean-square error=0.26 and correlation=0.74 in the AOD comparison; 73% SSA retrievals within ± 0.05 of AERONET inversions)."

I. 24: "Cimel sunphotometers that measures" Change to "Cimel sun-photometers that measure" I. 26: "over worldwide locations" Change to "at many locations worldwide" Changes have been made in the revised text.

Pg 7297: I. 16: "in a given pass" Change to "in a given satellite overpass" Line 16: Corrected.

I. 23-26: Mention here that AERONET AOD was interpolated to 388 nm The following sentence has been added in the revised text "Also, note that the AERONET AOD measurements are available at 340 nm and 380 nm in the near-UV and 440 nm in the

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visible. Therefore, using the Extinction Angstrom Exponent in this range the AERONET measurements were interpolated to 388 nm to match with the wavelength of OMI retrieval.”

I. 23-26: Some more details on the OMI quality flags would be very useful here or earlier on in the text. The current OMAERUV flagging scheme includes all retrievals with conditions, $\Delta R \leq 0.04$ for dust and sulphate; $\Delta R \leq 0.08$ for biomass burning, and $UVAI \geq 1.0$; where ΔR is the TOA reflectivity-surface reflectivity, in to the flag 0 category. In this condition, we expect that these retrievals are cloud-free since the LER hardly goes beyond 10%. For the flag 1 category, the LER is allowed to go up to 30%, however, with $AI > 1.2$. Due to higher reflectivity in such cases, it is expected that the OMI pixel may be contaminated with cloud. However, larger magnitude of aerosol index for these measurements is a strong indicator of the presence of absorbing aerosols over these pixels. It has been re-written in section 2, 3rd paragraph

Pg 7298: I. 20: Why do you not use the OMI surface albedo database compiled by Kleipool et al.? Kleipool et al. (2008) uses three years of OMI data for estimating the surface albedo, whereas the current OMAERUV algorithm uses climatology of the surface reflectance product derived from the 14-years of TOMS record. Owing to its longer record, the TOMS surface albedo product is expected to represent the true surface condition. Therefore, we believe that the TOMS surface reflectivity database is more reliable.

Also, please refer to next comment.

I. 21-23: "The surface albedo in the UV (...) the error in the aerosol retrieval produced by the uncertainty associated with surface albedo is expected to be very low." Are there sensitivity studies that prove this? A small error in a small number (i.e. UV surface albedo) may lead to large errors in retrieved products? Might the reason also be the strong contribution from Rayleigh scattering in this wavelength range, which makes the contribution of radiation reflected by the surface less important? Torres et al. (1998)

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paper (Table 3) presents the analysis of errors in the retrieved aerosol optical depth. For a surface reflectivity uncertainty of ± 0.01 , the accuracy of the retrieved optical depth is ± 0.1 for non-absorbing and weakly absorbing aerosols. As the aerosol becomes more absorbing, the sensitivity to surface albedo decreases, and the accuracy of the retrieval is better than ± 0.05 . A higher level of absorption in the near-UV indicated by the range of AOD measured by AERONET sunphotometers (AOD up to 4.0) and higher magnitudes of UV-AI over South America region would likely lessen the impact of uncertainty in the assumed surface albedo on the retrievals.

Pg 7299: l. 8-9: "This suggests (...) is likely to be correct" No, it does not (at least not in general, but only for this case). But it does say that the error is not (only) caused by layer height uncertainty. Does a validation study of OMI aerosol height exist?

For carbonaceous aerosols, the AOD is overestimated by 10% per 1 km underestimation [Torres et al, 1998]. Thus, to explain errors of 100% and larger in terms of the prescribed aerosol height, the 'actual' location of the aerosol layer would need to be at least 13 km which clearly unrealistic. The assumed vertical distribution is actually consistent with direct measurements by the CALIOP lidar. l. 11-14: "The use (...) near-UV radiation." Remove this sentence. It appears to be incorrect and is not of importance. We think that this is true. The gray aerosols such as pure black carbon has no spectral dependence in absorption which yields AAE of 1.0. The urban/industrial aerosol mass which is dominated by BC serves as a good example for this case. l. 16: Please explain the AAE in more detail, as it is an important parameter for this study. Also add a reference to: "Spectral absorption properties of atmospheric aerosols R. W. Bergstrom, P. Pilewskie, P. B. Russell, J. Redemann, T. C. Bond, P. K. Quinn, and B. Sierau Atmos. Chem. Phys., 7, 5937-5943, 2007" A short description of AAE is added here. The AAOD is related to the wavelength by power law fit given by, $AAOD = k \cdot \lambda^{-AAE}$ where, AAOD is the absorption aerosol optical depth, λ is the wavelength, and AAE is the absorption angstrom exponent which is the slope of AAOD-wavelength relation in log-log space. Thus, AAE describes the spectral dependence of aerosol

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absorption with larger (smaller) values of AAE indicate steeper decrease (increase) in absorption with wavelength

Pg 7300: I. 3: "RSD" What does this abbreviation mean? Please do not use too many different terms for the spectral dependence of absorption, as it confuses the reader. Use only AAE, or, if you must use another quantity, mention the AAE for that particular case. RSD is the relative spectral dependence of the imaginary index between the 354 and 388 nm retrieval channels. The term "RSD" has been replaced with spectral dependence of imaginary index and AAE values in the revised text.

I. 12: Add a reference to (M. de Graaf, P. Stammes, O. Torres, and R. B. A. Koelemeijer, J. Geophys. Res., 110, D01201, doi:10.1029/2004JD005178, 2005) here A sentence with de Graff et al (2005) has been added here. "A similar result was also found by de Graff et al. (2005) where they found Absorbing AI to be sensitive to the wavelength-dependence refractive indices and elevated absorbing aerosols"

Pg 7301: I. 26-29 (and on next page): What kind of samples were collected here? How were they measured, e.g., at what relative humidity? What is the relevance of those results to the OMI retrieval? We realize that among the other related references to the present study, Martins et al. (2009) may not be the best reference to include here as this study presents the results which are valid for the urban region (Sao Paulo, Brazil), whereas our focus in this study was to retrieve aerosols over the biomass burning region. Therefore, we have omitted Martins et al. (2009) in our manuscript.

Pg 7302: I. 1-10: Also compare the results discussed in (Bergstrom, 2007) (full reference can be found above) "Bergstrom et al. (2007) presents the results on the spectral absorption properties of aerosols derived during several major field campaigns (TARFOX 1996, SAFARI 2000, PRIDE 2000, ACE Asia 2001, ICARTT 2004) conducted over different regions of the world to sample distinct aerosol mass. It was found that the AAE was near one for urban pollution (TARFOX and ICARTT) and nearer two for biomass smoke (SAFARI 2000). " Above description has been added in the last paragraph of

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section 4.0

I. 8 / I. 19: "The AAE in the range 440-670 nm was ~ 1.5 " But this is in contradiction with the values cited later: "AAE in the range 2.5 \sim 3.0" Why is 2.5 assumed for the retrieval, and not 1.5? The AAE value of 1.5 was derived by Russell et al. (2010) using the AERONET absorption retrievals in the visible wavelength range 440-670 nm, whereas AAE of 2.5 to 3.0 was derived from the assumed spectral absorption properties in the near-UV (354-388 nm) region for the new OMI smoke models.

The AAE value of 1.5 is valid for the visible region as shown by Russell et al. (2010), whereas our method employs near-UV channels for the aerosol retrieval. Based on the Kirchstetter et al. (2004) (AAE \sim 2.8 in the near-UV) and the sensitivity analysis (section 3.2, last paragraph), we found that the 20% RSD (corresponding AAE in the range 2.5-3.0 depending on the smoke model) is the appropriate assumption for the retrieval.

I. 23-24: "Using a (...) was carried out." Rephrase this sentence, it does not make much sense in the current form. We use a research version of OMAERUV aerosol algorithm software for the retrievals. This code has greater flexibility to carry out sensitivity analysis and retrieval simultaneously. It is equivalent to the operational OMAERUV algorithm.

I. 26: "observations at the AERONET sites used in the study." Change to "observations at four AERONET sites." Changed to "observations for the seven AERONET sites considered in the study".

Pg 7303: I. 2: What do you mean by $\sim 100\%$ improvement? Improvement in what quantity and how is this percentage determined? The sentence has been re-written as "The improvement is significant for the previously labeled flag 1 for which the new AOD is much better aligned with the ground-truth." Few sentences have been re-written: "Note that this comparison includes all flag 0 and flag 1 retrievals for which the measured reflectivity was less than 20%." "About 70% of total retrievals with quality flag 0 and 1

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now fall within the pre-estimated uncertainty against 32% retrievals within uncertainty noted in the standard OMI-AERONET comparison (see Figure 2).”

I. 19-28: Fig. 5 has three panels, not two (although the far right panel, which is not mentioned in the text, is not essential to the paper and can be left out). Please refer to my second comment from last below.

Pg 7304: I. 5: "facilitate the comparison" Should be something like "make a fair comparison possible" This sentence has been re-written as: “OMAERUV-retrieved values have been converted to 441 nm by performing interpolation between OMI reported 388 and 500 nm values to compare them with AERONET inversions at the same wavelength.”

I. 14: "carbonaceous aerosol models" Change to "colored aerosol models", for BC is also carbonaceous. Changed to “new colored smoke aerosol models”.

I. 20: "The African burning season" Change this to something like "The burning season in Central Africa", since there are two major biomass burning seasons in Africa. Changed to “The biomass burning season in the central Africa starts. . .”

Pg 7305: I. 2: "associated with (...) (EAE>1)." Did you infer this from AERONET measurements of SSA? No. EAE is not derived from AERONET inversion of SSA. EAE is derived from spectral aerosol extinction optical depth. Our experience with AERONET data over the Kanpur region and previously published papers show that EAE is mostly greater than 1 during the winter months over this region.

I. 5: "MODIS-derived fine mode fraction (...) persistence layer of smoke" How does MODIS FMF show this? Smoke particles tend to be small, like pollution particles. Our new retrievals of MODIS fine mode fraction (Jethva et al., 2010, JGR) over the Indian region show the presence of smoke (FMF>0.7) over the Indo-Gangetic Plain. Please refer to the paper: Jethva, H., S. K. Satheesh, J. Srinivasan, and R. C. Levy (2010), Improved retrieval of aerosol size-resolved properties from moderate resolution imaging spectroradiometer over India: Role of aerosol model and surface reflectance,

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I. 13: "OMI algorithm identifies aerosol type as smoke" How does the OMI algorithm do this? Based on climatology and/or AI value? Do you know how reliable this is? The standard OMAERUV aerosol algorithm uses the aerosol-related change in the LER (TOA Reflectivity – Surface Reflectivity) and UV-AI in conjunction with the pre-determined geographic distribution of aerosol type for the selection of aerosol model in the retrievals (Torres et al., 2007). However, in the retrievals presented in this manuscript, we assume a set of new carbonaceous models that account for the organics and black carbon in performing the retrievals over South America, central Africa, and northern India. This selection is based on the fact that the aerosol type observed over these regions is primarily of carbonaceous type during biomass burning season.

Pg 7306: I. 5: "we attempted to evaluate" Change to "we evaluated" Corrected.

I. 7: "central Africa, and northern India" This statement is too bold: you only investigated the agreement at two AERONET sites in Africa and India, this does not mean that the good agreement is found for the whole region. Can you add some AERONET stations in these regions to your comparison? Mongu and Kanpur AERONET stations have relatively longer aerosol record than the other AERONET sites in these regions. Gandhi College-AERONET site is located near to Kanpur station and exhibits similar aerosol properties as Kanpur (personal experience with the AERONET data). Also, Kanpur-AERONET measurements have been used in many investigations for characterizing aerosols over the Indo-Gangetic Plain. The evaluation of OMAERUV aerosol products over these regions is an ongoing process and will be carried out once additional measurements are available.

I. 13: "fresh" Change to "new" Corrected.

I. 19: "therefore should be considered as good quality data." You are probably referring to the quality flag here. Do you mean to say that for these cases the flag should be changed from 1 to 0? Yes. A close examination of OMI reflectivity (LER) along with

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its near-time MODIS true color RGB images of smoke events observed over South America and northern India suggests that the smoke events often produce reflectivity in the range 10-20%. In some cases, the reflectivity of 25% has also been observed. The dramatic response of the AOD retrieval with the use of new carbonaceous models for these retrievals provides the justification to treat them as good quality retrievals.

I. 20-24: "This paper strongly suggests (...) absorption properties." Not only for OMAERUV, but also in other remote sensing aerosol retrievals! This is a matter of investigation. Since the impact of absorption by organics is much stronger in the near-UV than in the visible, it affected the OMAERUV retrieval severely. Based on our analysis presented in this paper, we suggest the inclusion of organics in the carbonaceous aerosol models for the operational OMAERUV aerosol retrieval over the biomass burning environment. A similar kind of analysis is needed to analyze the impact on the aerosol retrieval in the visible region, such as carried out by MODIS and MISR.

Pg 7307: I. 1: "of biomass burning analyzed here." Add: "but this needs to be corroborated with further evidence from other AERONET stations in those regions." The sentences have been re-written: "The use of limited ground-based information on the spectral dependence of aerosol absorption and a significant improvement in the retrieval of OMAERUV AOD and SSA at near-UV wavelengths achieved with the new smoke models over South America, central Africa, and northern India, indicate the presence of organics in biomass burning aerosols over these regions."

I. 6: "AAE as inferred from OMI" To my understanding, AAE is not inferred (i.e., retrieved), but assumed in the retrieval. Yes, the AAE is currently assumed in the OMAERUV aerosol algorithm. These sentences have been modified in the manuscript.

I. 9-17: Although I think the presented results are quite sensational, these conclusions are taking it one step too far. First, because the OMI retrieval needs input from AERONET to distinguish between BC and BrC; and second, because this can hardly be done on a single-measurement basis, as I mentioned in my first comment (to P.

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7292, I. 19-21). It would be wonderful if OMI-AERONET measurements could be used for the "identification of aerosol composition", but it seems that we are still very far from achieving that goal. We agree with the reviewer on his comments. Based on our theoretical analysis and ground-based evidences, we assume a fixed spectral dependence for the retrieval of aerosol properties from OMI measurements. We anticipate that the use of AERONET AOD measurements in OMI retrieval will allow the OMAERUV algorithm to derive spectral dependence of imaginary index and hence AAE. This should help in identifying the "presence of organics" in the aerosol mass. This work is under investigation.

The last paragraph of the conclusion has been re-written as: "In addition to suggesting a significant improvement in the retrieval of aerosol properties from OMI, the present study also demonstrates the sensitivity of near-UV measurements to the chemical composition of biomass burning aerosols. This opens an opportunity to exploit the near-UV capabilities of OMI to identify the presence of organic carbon in the biomass burning aerosols. When an increasing number of studies reveals the association between aerosol types and its spectral aerosol absorption (or AAE), the near-UV inference of spectral absorption through the integration of OMI-AERONET measurements can be explored further to infer the aerosol composition."

Figure 1. "JiParna" is corrected to "JiParana"

Fig. 3: Add the AAE to the figure legend The AAE for the three simulated cases have been added to the plot. AAE = 1.07 for 'gray' aerosols (only BC) AAE = 2.67 for 'colored' aerosols with 20% spectral dependence in the imaginary index in the near-UV AAE = 3.37 for 'colored' aerosols with 30% spectral dependence in the imaginary index in the near-UV

Fig. 5: The far right panel can be removed, see also my comment to P. 7303, I. 19-28. Though we have presented the spatial distribution of AOD for standard (left panel) and new (middle panel) OMAERUV retrieval separately in this plot, we think that the

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difference between them can be clearly shown in a separate panel to aid the reader for ease of comparison.

Fig. 7: Can you make two panels out of this, one for Kanpur and one for Mongu? The OMI Vs AERONET comparison plots for Kanpur and Mongu are attached with this response. While the improvement in the AOD retrieval over both sites is clearly evident from the increased number of data points within expected uncertainty, the impact of new carbonaceous models is pronounced in the case of Kanpur measurements. To save space in the manuscript, both plots are merged in to a single plot.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 7291, 2011.

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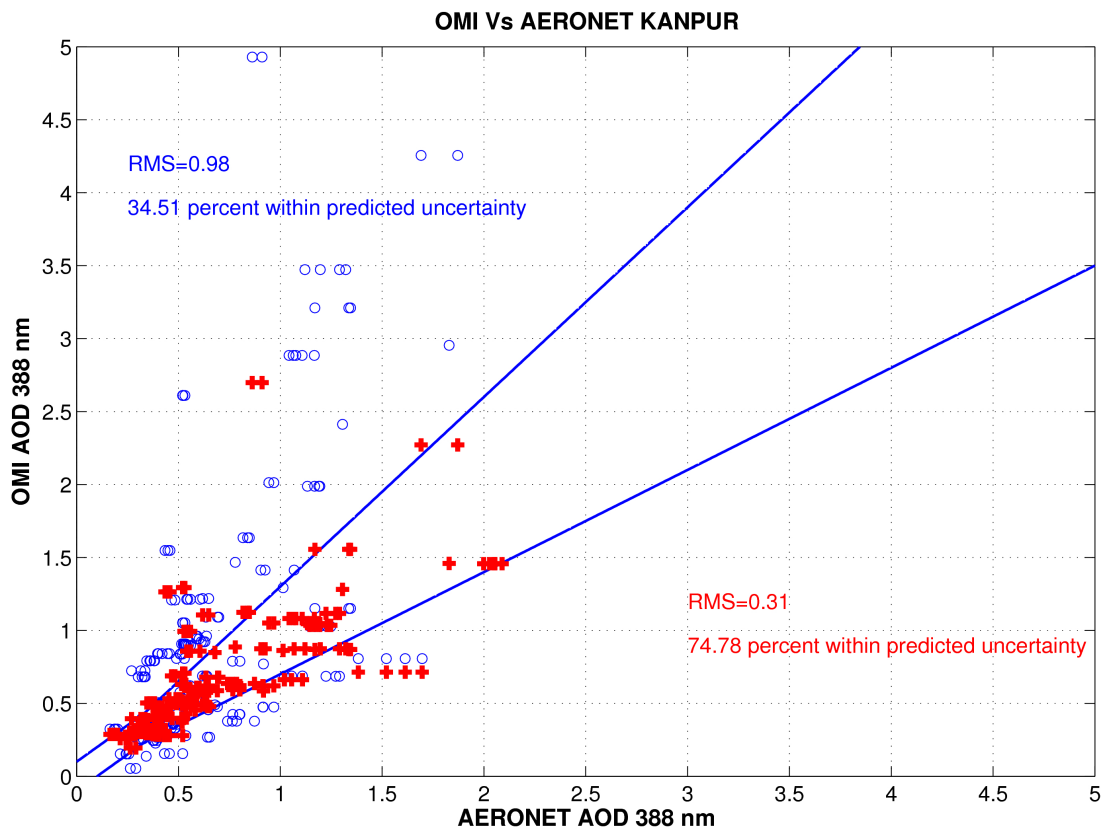


Fig. 1.

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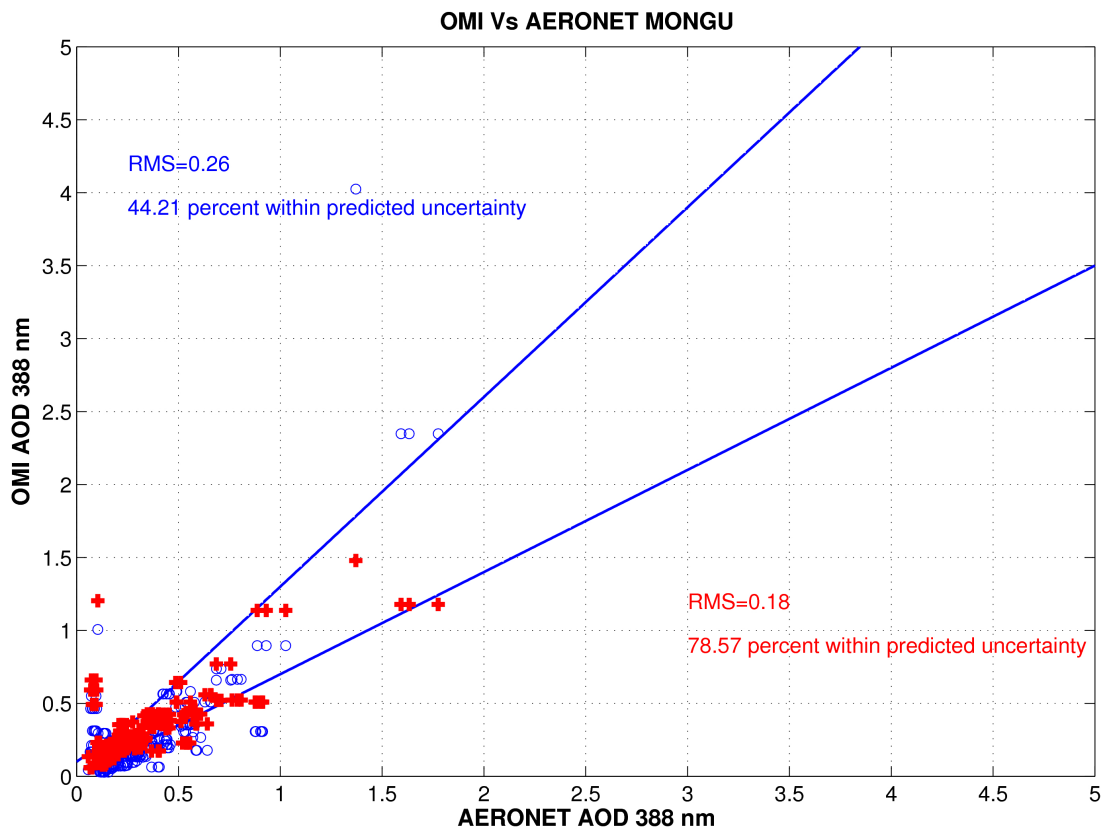
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Fig. 2.

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