Atmospheric
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Discussions

Interactive comment on "Aerosol absorption retrieval at ultraviolet wavelengths in a complex environment" *by* Stelios Kazadzis et al.

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Review: S. Kazadzis et al., Aerosol absorption retrieval at ultraviolet wavelengths in a complex environment. (Plain text version. Full version - see PDF attachment.)

General comments The paper presents first long-term retrievals of the column average aerosol Single scattering Albedo (SSA) at two UVA wavelengths (332nm and 368nm) using UVMFR measurements of diffuse, global and direct surface irradiance in Athens, Greece. The methodology retrieving SSA using UVMFR direct to global irradiance Ratio (DGR) measurements has been previously developed and authors appropriately reference previous works. The paper would benefit from adding more details of UVMFR operations (e.g., picture of the site, procedures for cleaning Teflon diffuser, checking diffuser alignment, night-time bias correction, aureole correction).

The paper presents new results that will be of interest to broad aerosol, atmospheric composition and air quality communities. This paper first analyzes long-term (5 year) UVMFR SSA retrievals that allows for statistically robust analysis of the absorbing aerosol climatology in Athens. Long-term comparison with standard AERONET SSA inversions at 440nm is also new. The paper further uses standard AERONET retrievals of the column effective imaginary refractive index in VIS – NIR wavelengths and methodology of Schuster et al., (2016) to calculate volume fractions of absorbing aerosol components (e.g., black and brown carbon, hematite, goethite) in Athens. This is new and interesting development. The main result (Fig.13) is that Brown Carbon dominates column aerosol absorption. October peak of BrC volume fraction is interesting new result. Authors should try to explain their result, e.g., using in-situ measurements. The possible enhancement of this new approach would be extending Schuster et al., (2016) methodology by using UVMFR retrieved imaginary refractive indices at 332nm and 368nm [Krotkov et al., 2005b].

The paper is appropriate for publishing in AMT after important technical corrections, such as adding missing references (e.g., IPCC 2007, 2013, 2014) and missing figure (p22,L10-11: "Figure 13 shows the temporal variability of AAE(440-870) and AAE(332-440)."). The manuscript also needs language and punctuation improvements and will benefit from editing by native English speaker.

I recommend publishing the paper after corrections and implementing additional technical suggestions described below.

Use consistently italic or regular font in references and examples. Use dot and comma after abbreviations, such as "e.g., et al." Do not use apostrophes in SSAs , AEs and AODs All references need consistent formatting according to AMT style.

Specific suggestions: 1,18: properties -> property (Only one absorption property, i.e., SSA is retrieved). 1,19 5-years period 1,22: and study absorption spectral behavior of the [retrieved] SSA values 1,24: towards lower shorter UV wavelengths 1,25: High

Strong SSA wavelength dependence ... 1,27: "SSA decrease with decreasing extinction optical depth, suggesting an effect of the different aerosol composition" – this could be due to in increased SSA uncertainties at lower AODs

2,2: "were investigated to understand seasonal variability of the results" – to explain? 2.6. e.g., 2.7: IPCC references are missing 2,8: "... as it appears that climate change is accelerating with aerosols impacting ..." - This sentence needs clarification and reference: how aerosol and climate changes are related? 2,13: "significant aerosol absorption uncertainties in [modeled?] global Single Scattering Albedo (SSA)," 2,16: mixture [mixing state?] 2,20: 50% change [decrease?] in the [surface] erythermal irradiance 2,21: et al., 2,22: e.g., (add comma) 2,24 "a fixed irradiance path" – please, re-word. Surface irradiance is a result of averaging different direct and scattered photons arriving at the surface via different paths through the atmosphere. 2,27: Do not use italic font in references. 2,28: VIS-SSA -> column average SSA retrievals at the visible and near IR wavelengths (i.e., 440nm, 670nm, 870nm, 1020nm).

2.29-2.32: "In addition, Goering et al. (2005), Taylor et al (2008) and Kudo et al. (2008) have proposed estimation techniques for the retrieval of spectral aerosol optical properties by combining multi-wavelength measurements using a priori constraints that are applied differently than in the single wavelength methods."

Suggest replacing this sentence with:

In addition, surface direct and diffuse irradiances had been used to derive spectral AOD and SSA at visible and UV wavelengths (King and Herman 1979; King 1979; Petters et al., 2003; Eck et al., 1998; Krotkov et al., 2005b; Bais et al., 2005; Goering et al., 2005; Taylor et al., 2008; Kudo et al., 2008; Corr et al., 2009).

2,32: "SSA retrieval in the ultraviolet (UV) part of the spectrum is weaker with large uncertainties." – I suggest removing this sentence.

3,10: " ...like organic, nitrate and aromatic aerosols are still poorly known" add ref-

erences, e.g., Jacobson, M. Z. (1999), Isolating nitrated and aromatic aerosols and nitrated aromatic gases as sources of ultraviolet light absorption, J. Geophys. Res., 104, 3527–3542.

3,16-18: Barnard et al. (2008) [and Corr et al., (2009)] ...in a case [field] study ... found that, in the near-UV spectral range (250 300 to 400 nm) 3,28: "in at 870 [nm] " "could be a hint reason for ..." 3,32: " using Brewer [direct sun and global irradiance spectral UV] measurements ..."

4,2: and They used imaginary refractive index ... and found ... 4,10: (e.g. Zerefos et al., 2012; - add more references 4,11: "comparable in magnitude [or exceeding] with those caused by the decline in stratospheric ozone [depending on wavelength] 4,13 "reduction of 7% of AOD ..." - at what wavelength? 4,17: "tropospheric photochemistry[, causing:]

5,5: "Solar irradiance satellite retrieval algorithms are directly affected " -> satellite retrieval algorithms of the surface UV irradiance are directly affected ... 5,8: absent from [current] satellite (e.g., OMI) retrieval algorithms 5,12: "Uncertainty on [in] commonly used .." 5,13: "... fall short in precision due to large uncertainties in the input parameters ..." -> The model accuracy of the surface UV irradiance is limited by large uncertainties in the input parameters ...

5,24- 6,10 – suggest deleting this paragraph as common knowledge. Move Equation (2) to the beginning of section 2.2

6,11-27 – this paragraph looks repetitive and could be blended with the earlier part of introduction. 7,25: constructing [manufacturing] company 7,29: irradiance[s]

8,5: "...in conjunction with radiative transfer model (RTM) calculations that have been performed using the Libradtran code (Mayer and Kylling, 2005). " 8,12: "SSA is a key aerosol optical property and describes the portion of solar irradiance that is scattered from the main direct beam passing through the atmosphere." – Equation (2) can be

moved after this sentence.

9,4: "raw voltage measurements [corrected for night-time voltages and non-ideal angular response] could be used." 9,19: "dt." -> time interval

10,fig 2 caption "...for a day with variable cloudiness [in the afternoon] 10, 9 : "... for performing [determining] extraterrestrial Langley calibration constant (ETC) determination ... 10,10: "the Beer-Lambert law for to the UVMFR direct [voltage] measurements" 10,13-14: "extrapolated AOD at UVMFR wavelengths" – Clarify how AOD was extrapolated? 11,6: AOD's at 332 nm and 368 were ... 11, 15: extrapolation [using ln(AOD) versus ln(wavelength)]? 12, Fig 4 caption: "Comparison of CIMEL and[extrapolated] and ... UVMFR retrieved AODs for ... 332 nm (up top panel) and 368 nm (down bottom panel)." 12,9: "as a function of SZA (figure 5 4)." 12,12: "...are included [found] ..." 12,18: " ...due to instrumental (filter related) changes ..." – Most likely reason for ETC change is UVMFR Teflon diffusor contamination. Explain how often the UVMFR diffusor was cleaned and what cleaning procedures applied?

12, 19 AODs 's deviations ... errors on in SSA calculations .. 13,6: were deployed for the use of [used for construction of] the LUT ... 14, Fig 6. Caption Figure 6 LUT of direct to global ratio at 368nm ... color bar represents assumed SSA values.

14,13: average annual monthly AODs 's 15,12: SSAs 's 15,15: role on [in] the uncertainty 15,21: a [close] match between 15,22: "This range broadens at low SZA and low aerosol level cases" – Please, clarify this sentence and refer to Figs 5 and 6.

15, 26: "AOD uncertainty is considered as $\pm$2% for 368nm and $\pm$4% for 332nm," Should it be absolute AOD uncertainties: 0.02 at 368nm and 0.04 at 332nm ? 16,3 AODs 16,5: In the same figure, 16,6: mean AODs 's [for each SZA bin] and 1σ, the error bars equal to one standard deviation are shown 16,16: Figure 8 Daily Mean daily SSAs ... 16,20: for 332nm (368nm)" - Should it be reversed, i.e. , at 368nm (332nm) (SSA at 332 nm is generally lower than at 368nm) – fig 7. ?

17, Fig. 9 : Include X axis title. Are spectral differences between SSA at 368nm and at 3232nm between November and March statistically significant? Apply standard statistical significance tests. 17,1: for the specific area," – correct reference Pareskevopoulou et al (2014) -> Paraskevopoulou, et al., 17,2: at in February and November 17, 3 at in a 5 year (2008-2013) ... 17,4: have similar behavior SSAs ... 17,16-17: "for different SSA uncertainty bins according to analysis of in the previous section ...

18, Figure 10: Suggest local time or SZA as X axis 18, Figure 10 caption: Mean values per hour plotted [with error bars showing one standard deviation] at 1σ 18,8: are [also] linked ... 18,8: derived at AERONET calibration site in Greenbelt, Maryland USA Washington

19,15: link between [SSA] wavelength dependence and

20, 5 The results of in figure 12 20, 9 ...relatively higher than SSA440) tend [to occur] towards high AEs ... 20,10 attributed in [to] polluted ... 21,6: Schuster et al., (2016) 21,7: method separates [contributions from] black carbon, organic carbon, hematite and goethite, using [to the retrieved] refractive index ... 21,9: 8-9: "Figure 12 shows the fractions of total aerosol [column] volume attributed to these components in both fine and coarse mode ..." – This should be Fig. 13. There are no fine and coarse mode fraction data in Fig.13.

21,14: "... higher at [in October] OCTOBER " – It will be interesting to explain BrC peak in October compared to other months. Are there in-situ measurements in Athens that could support this finding?

21,17: Figure 13 caption: "Volume fraction[s] (in the lower plot) of absorbing aerosol components ..."

22,4: " for atmospheric aerosol [mixtures] scattering varies ..." 22,10-11 "Figure 13 shows the temporal variability of AAE(440-870) and AAE(332-440)." – This figure (14?) is missing.

23, Figure 14 – This should be figure 15 23, 7. I suggest adding new reference, which shows previously measured AERONET-UVMFR SSA spectral dependence in Thessaloniki, Greece:

N. Krotkov ; G. Labow ; J. Herman ; J. Slusser ; R. Tree ; G. Janson ; B. Durham ; T. Eck ; B. Holben; Aerosol column absorption measurements using co-located UV-MFRSR and AERONET CIMEL instruments. Proc. SPIE 7462, Ultraviolet and Visible Ground- and Space-based Measurements, Trace Gases, Aerosols and Effects VI, 746205 (August 20, 2009); doi:10.1117/12.826880.

23,19: for all SZA[s]

24,10: "We have also [used] the produced dataset to investigate . . . 24,18: "We expect a possible decrease in specific days/cases of regional O3 due to the enhanced aerosol absorption" - This conclusion is not supported in the main text. Add Chemical model results to support this.

Please also note the supplement to this comment:
http://www.atmos-meas-tech-discuss.net/amt-2016-273/amt-2016-273-RC2-supplement.pdf