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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Insight into Global Trends in Aerosol Composition over 2005–2015 Inferred from the OMI Ultraviolet Aerosol Index" *by* Melanie S. Hammer et al.

Anonymous Referee #1

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In the study presented in this manuscript, the authors attempt to identify the aerosol components responsible for trends observed in monthly averaged UVAI. Aerosol fields from a global model were combined with radiative transfer calculations to obtain modeled UVAI which were compared with OMI UVAI. Whereas the topic is interesting and the approach promising, the study was not well performed. In particular, I have three major issues with the current manuscript:

1. The interpretation of UVAI and UVAI trends is not sufficiently addressed (as mentioned in the first round of review). The authors have the tools (GCM + RTM) to study the effects of changing aerosol amount, composition, and altitude on UVAI; if they

would perform a detailed systematic study for several selected regions (and, possibly, seasons) this would greatly aid the interpretation of observed trends.

2. The authors attempt to explain observed UVAI trends by comparing with model data that shows rather different trends. It is unclear how this could work.

3. There appears to be no important conclusion. The attribution of various trends to certain changes in aerosol amount or composition remains rather speculative.

Specific comments

Section 5:

a. The presented trends are tiny, at most 0.02 (seen in North Africa) over the whole decade of OMI observations. Although the analysis reports that these trends are statistically significant (as the authors state), I would like to apply to common sense and a critical look at the data: the uncertainty of UVAI is at least on the order of 0.1, and the variability (due to aerosols, clouds, and surface) is much larger. If you insist on discussing these trends, I strongly suggest adding a figure so that the reliability of the trend analysis can be estimated.

b. Why is the rather obvious modeled negative trend over the Sahara in SON not addressed in the text? In the conclusions it is mentioned that it is "erroneous", but I'd rather say it is not in agreement with observations (maybe the same trend is present in the observations, but compensated by, e.g., changes in surface reflection).

c. If the dust UVAI over Mongolia decreased due to changes in wind speed, why is this not reproduced by the model?

d. As you mention in ll. 321-325, changes in surface reflectance affect the UVAI. This will strongly affect your trend analysis, so it may be important to estimate he contribution of changing surface reflectance to the UVAI trend using VLIDORT.

Section 6:

e. As mentioned in issue 2 above, I fail to see how the comparison of observed trends with model trends can aid the understanding of the observed trends, as there is only little agreement between both data sets. A better set-up for this study would have been a focus on 3-5 regions of interest, for which extensive tests with varying aerosol amounts and composition should have been performed.

f. Similarly, it remains unclear what we learn from an analysis as that shown in Fig. 8.

Section 7:

g. In general, there should be more coupling of the model results to observations. E.g., instead of the inaccurate statement (ll. 408-409) that "The simulation attributed the negative trend over South Asia to increasing scattering secondary inorganic aerosols (...)", I would argue that by comparing Figs. 5, 6, and 9, it can be seen that over India the modeled trend is possibly too small because either the UVAI increase due to more dust is overestimated or the UVAI decrease due to more secondary aerosols is underestimated. The observed trend is negative.

h. The fact that over the eastern US you find the strongest negative UVAI trend in summer is a strong indication that this is caused by secondary organic aerosols of biogenic origin - not, as stated in the manuscript, SIA. SOA are notoriously badly reproduced by GCMs. (see, e.g., Penning de Vries et al., ACP 2015 (doi: 10.5194/acp-15-10597-2015))

Minor comments:

p.2, l.42: The indirect effect is only mentioned in passing here, but it is actually at least as great as the direct effect. Please add a line or two about the indirect effect.

p.3, l.64: Please add a reference to the study by Fioletov et al., ACP 2016 (doi: 10.5194/acp-16-11497-2016)

p.4, l.102: Please add a reference to Penning de Vries et al., ACP 2009 after "absorption and scattering." (doi: 10.5194/acp-9-9555-2009)

p.4, ll.102-105: "Prior interpretation (...) through simulation." This is rather vague. A few more words on UVAI's "dependence on other parameters" would be helpful here, preferably with a figure showing the AOD and ALH dependences of the different aerosol components (SIA, Dust, OA, BrC, BC, and Salt).

p.5, l.128: Add references to Herman et al., 1997, and Torres et al., 1998.

p.5, ll.133-134: "Negative UVAI values due to aerosol scattering are often weak and buried in noise (Torres et al., 2007)." But certainly not always! In fact, you interpret the negative UVAI values in this study, so please rephrase and cite Penning de Vries et al., ACP 2015 (doi: 10.5194/acp-15-10597-2015)

p.5, 134-137: "Because UVAI (...) the absorption signal." This statement is too simple; did you test it using your RTM? In fact, multiple scattering (particularly if a layer of scattering aerosols is located below an absorbing aerosol layer) may increase absorption and UVAI.

p.5, ll.139-145: Why do you mention the other OMAERUV products if you don't use them? Consider removing the paragraph. If you do decide to keep it, insert "by incorporating" between "observed radiances" and "aerosol type selection" on line 140.

p.5, l.151: Which "cloud fraction" do you mean? Effective? Radiative? The cloud fraction from the UVAI algorithm? How much is 0.05? And a related question: why does so little data over ocean remain - is your cloud filter too restrictive there?

p.6, l.157: "insensitive" -> "less sensitive"

p.6, l.159: "deseasonalized": this is not mentioned in the results section. Why would you want to do this? More importantly: can you deseasonalize by subtracting a mean value? The UVAI is not additive!

p.6, ll.159-160: "A minimum temporal coverage of 60%": What, exactly, do you mean?

p.6, ll.177-181: It is unclear to me how the radiances were calculated, can you present

an example? E.g., the UVAI of an OMI pixel with 4% cloudiness and cloud optical depth equal to 30 is simulated by summing the radiances from a cloudy pixel with optical depth 30 (Rcloudy) and those from the aerosol scene (Raerosol) by using the independent pixel approximation: Rpixel = 0.04*Rcloudy+0.96*Raerosol But is that approach correct? The cloud fraction determined within the UVAI algorithm is not a real cloud fraction in the sense that it represents cloudiness. It is used by the algorithm to adapt the RT-modeled radiance to the observed radiance. More appropriate would be the use of an independent cloud fraction, e.g. OMI's official cloud product or even MODIS cloud fraction and optical depth (which are available at OMI resolution).

p.9, ll.260-261: "missing sources of anthropogenic dust": I doubt that, as UVAI is only sensitive to elevated dust layers, see Torres et al., 1998.

p.9, l.274: "OMI,)": spurious comma

p.9, l.274: "GLS": acronym not explained in the text.

p. 14, l.418: Please explain the acronym TROPOMI and add a literature reference

p. 14, l.420: Please explain the acronym MAIA and add a literature reference

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