We would like to thank both referees for their comments regarding our manuscript. Below we have included our responses. The referee comments are in black text while our responses are in blue.

Referee Comment #1

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.

We have previously conducted a detailed, systematic study of our simulated UVAI compared to the OMI UVAI for several selected regions and seasons in our previous publication (Hammer et al., 2016; https://doi.org/10.5194/acp-16-2507-2016). We expand on the reference to this previous publication in our introduction:

Page 4 lines 110-112: "In this work, we apply a simulation of the UVAI, which was developed and evaluated regionally and seasonally in Hammer et al. (2016) …"

We also elaborate on our studies of the effects of changing aerosol amount and composition in this manuscript:

Page 12 lines 353-356: "Figure 8 shows the change in annual mean UVAI due to doubling the concentration of individual aerosol species. This information facilitates interpretation of the observed UVAI trends by identifying the chemical components that could explain the observed trends."

and we add new sensitivity simulations of the effects of aerosol altitude:

Page 6 lines 183-186: "We also calculated the change in UVAI due to changes in simulated aerosol altitude, but found that the trends in aerosol altitude were negligible (order 10^{-5} hPa yr^{-1}). Therefore we focus our analysis on trends in aerosol composition which have a larger effect on the UVAI as demonstrated below."

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.

Indeed we are learning a great deal from the comparison of observed and modelled trends. Our study does not require that our modeled trends match the observed OMI UVAI trends. Rather, in our UVAI simulation we have known aerosol composition. Therefore we can directly interpret how trends in modelled aerosol composition relate to trends in simulated UVAI values. We can then use the trends in simulated UVAI values with known aerosol composition to interpret either the similarities or differences in the trends in observed OMI UVAI values. This comparison thus offers insight into the observed OMI UVAI values that can be explained by the simulation, and insight into possible explanations of remaining discrepancies.

We write in our introduction:

Page 4 lines 115-118: "Comparison of trends in observed OMI UVAI values to the trends in simulated UVAI values, which are calculated using known aerosol composition, enables qualification of how changes in aerosol absorption and scattering could influence the observed UVAI trends and identification of model development needs."

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

We have emphasized that interpretation of the OMI UVAI with a quantitative simulation of the UVAI offers information about trends in aerosol composition. For example, we found that global trends in the UVAI were largely explained by trends in absorption of mineral dust, absorption by brown carbon, and scattering by secondary inorganic aerosols. We also identified areas for model development, such as dust emissions from the desiccating Aral Sea.

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.

Indeed, we have taken great care to address the errors in the UVAI to enable quantitative interpretation of observed trends as now described in line 143. In particular we have uased a recently reprocessed version of the UVAI algorithm which treats clouds with a Mie-scattering based water cloud model. We have used a UVAI algorithm that more accurately accounted for scattering by mineral dust and by clouds, reducing systematic artifacts and scan angle bias. Furthermore, we focus on 10-years of observations so that multiple observations can reduce the random error of UVAI observations.

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).

We have added a comment about the modeled negative trend over the Sahara in SON:

Page 10 lines 308-312: "The simulation underestimates the observed UVAI trend over North Africa in SON, perhaps related to an underestimate in trends in mineral dust emissions in the simulation during this season. He et al. (2014) examined the 2000-2010 trends in global surface albedo using the Global Land Surface Satellites (GLASS) dataset and found no significant trends over this region during SON."

We have rephrased the conclusion:

Page 14 lines 406-408: "The simulated UVAI attributes the positive trends over North Africa to increasing mineral dust, despite an underestimated simulated trend in fall (SON) that deserves further attention."

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

Our simulation is for a different time period (2005-2015) than the Guan et al. (2017) study over 1960-2007. We have rephrased to emphasize the differences:

Page 11, lines 313-319: "The OMI UVAI trend over Mongolia/Inner Mongolia may be part of a longer term trend. Guan et al. (2017) examined dust storm data over northern China (including Inner Mongolia) for the period 1960-2007, and found that dust storm frequency has been declining over the region due to a gradual decrease in wind speed."

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 the contribution of changing surface reflectance to the UVAI trend using VLIDORT.

We have added to the manuscript:

Page 6 lines 179-183: "For the UVAI calculation we use the surface reflectance fields provided by OMI. We calculated the 2005-2015 trends in these surface reflectance fields, and found that they were statistically insignificant globally and on the order of 10^{-5}. We calculated the change in UVAI due to a change in surface reflectance of this order of magnitude, and found that the change in UVAI was negligible. Therefore we focus our analysis on trends in aerosol composition which have a larger effect on the UVAI as demonstrated below."

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.

Thank you for your suggestion. We prefer to share the full global analysis with the reader for transparency, so the reader can see both areas of agreement and disagreement. We learn from areas of disagreement as much as from areas of agreement as evidenced by the discussion of the feature near the Aral Sea. We elaborate on this in the manuscript:

Page 4 lines 115-118: "Comparison of trends in observed OMI UVAI values to the trends in simulated UVAI values, which are calculated using known aerosol composition, enables qualification of how changes in aerosol absorption and scattering could influence the observed UVAI trends and identification of model development needs."

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

We added:

Page 12 lines 353-356: "Figure 8 shows the change in annual mean UVAI due to doubling the concentration of individual aerosol species. This information facilitates interpretation of the observed UVAI trends by identifying the chemical components that could explain the observed trends."

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.

In this revision we have tried to emphasize the coupling of model results to observations. For example, by using scene-dependent OMI viewing geometry together with scene-dependent modelled atmospheric composition we enable quantitative comparison of model results with observations.

Regarding the specific comment for India, the trend in both simulated and observed UVAI values is negative over South Asia. Figure 9 shows that the largest change in our simulated UVAI values over India is a decrease due to the positive trends in scattering SIA over the region. Hence we respectfully retain the statement: "The simulation attributed the negative trend over South Asia to increasing scattering secondary inorganic aerosols, a trend that the observations imply could be even larger."

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))

Good point that OA may play a role. We revised the sentence:

Page 14 lines 415-417: “We found the positive trends in the UVAI over the eastern United States that were strongest in summer (JJA) in both the observations and the simulation were driven by negative trends in scattering secondary inorganic aerosol and organic aerosol.”

We also note that the GEOS-Chem aerosol simulation we use has been extensively evaluated against ground-based PM2.5 composition measurements (Li et al., (2016); DOI: 10.1021/acs.est.7b02530). The simulated aerosol trends were found to well reproduce the summer trends in OA over the southeastern U.S., mainly due to the inclusion of an aqueous formation mechanism of isoprene (Marais et al., 2016; DOI: 10.5194/acp-16-1603-2016).

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.

We now further emphasize the indirect effect:

Page 2 lines 38-39: “Atmospheric aerosols have significant climate impacts due to their ability to scatter and absorb solar radiation and to their indirect effect through modification of cloud properties.”

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

We have added this reference to the revised text:

Page 3 lines 63-66: “Due to the wide implementation of flue-gas desulfurization equipment on most power plants in China, emissions of sulfur dioxide (SO_2) in some regions have been decreasing since about 2006-2008 (Lu et al., 2011; Wang et al., 2015; Fioletov et al., 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)

Done.

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).

We have rephrased this sentence and have added references to the introduction about these dependencies:

Page 4 lines 105-107: “Prior interpretation of the UVAI has been complicated by its dependence on geophysical parameters, such as aerosol layer height (Herman et al., 1997; Torres et al., 1998; de Graaf et al., 2005).”

In terms of our calculated trends, we calculated the trend in ALH over 2005-2015, and found the trends were ~10^{-5} hPa yr^{-1} in magnitude. We conducted a sensitivity test by calculating the change in UVAI associated with a 1 hPa increase in ALH, and found that gave a negligible change in UVAI (~10^{-6}). As described in the manuscript:

Page 6 lines 183-186: "We also calculated the change in UVAI due to changes in simulated aerosol altitude, but found that the trends in aerosol altitude were negligible (order 10^{-5} hPa yr^{-1}). Therefore we focus our analysis on trends in aerosol composition which have a larger effect on the UVAI as demonstrated below."

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

Done.

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)

We have rephrased the sentence and added the citation:

Page 5 lines 137-138: "Negative UVAI values due to aerosol scattering are often weak and have historically been affected by noise in previous datasets (Torres et al., 2007; Penning de Vries et al., 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.

We have rephrased the sentence:

Page 5 lines 138-141: "Because UVAI values are calculated from top of atmosphere (TOA) radiance which contains total aerosol effects, the presence (or lack) of scattering aerosol along with absorbing aerosol can either weaken (or strengthen) the absorption signal."

We have tested this in the RTM, as shown in Figures 8 and 9.

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.

We have removed the paragraph.

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?

We have clarified in the text that the radiative cloud fraction is from the OMI UVAI algorithm, and 0.05 is 5%. We purposely use a very restrictive cloud filter to reduce error associated with clouds. We clarify this on lines 147-148.

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

We have made this correction in the revised text.

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!

We clarified our objective to focus on the long-term trends. Deseasonalization is a standard method for removing temporal variation from a dataset in order to accurately calculate a long-term trend in a time series. There is no need for the data to be "additive", subtracting the annual mean value from the monthly values allows for the removal of the monthly variation in the UVAI dataset which is not actually due to a trend in the data. This allows focus on long-term UVAI changes over time.

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

The sentence is rephrased to clarify that a particular pixel must contain data for at least 60% of the time period in order for regression to be performed, to avoid conducting a trend analysis on a pixel where there is only a few years of data.

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).

Page 6 lines 175-177: "Following Torres et al. (2018), we compute the radiances used in the UVAI calculation as a combination of clear and cloudy sky conditions. We use the same cloud fractions and cloud optical depths used in the OMI UVAI algorithm for coincident OMI pixels."

An in-depth description of the calculation of the radiances used in the UVAI calculation is provided in Torres et al. (2018) (https://doi.org/10.5194/amt-2017-429).

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.

The sentence has been rephrased:

Page 9 lines 261-264: "The simulation underestimates some of the smaller dust features captured by OMI, such as over western North America, South America, Australia, and parts of Asia, perhaps reflecting an underestimate in the simulated mineral dust lifetime (Ridley et al. 2012) and missing dust sources (Ginoux et al., 2012; Guan et al., 2016; Huang et al., 2015; Philip et al., 2017)."

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

We have corrected this in the revised text.

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

It is explained in the text:

Page 6 lines 158-160: "We perform trend analysis on monthly mean time series data for the years 2005-2015 using Generalized Least Squares (GLS) regression, as described by Boys et al. (2014)."

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

We have added this to the revised text:

Page 14 lines 423-429: "The recent launch of the TROPOspheric Monitoring Instrument (TROPOMI; Veefkind et al., 2012) and the forthcoming geostationary constellation offer UVAI observations at finer spatial and temporal resolution. The forthcoming Multi-Angle Imager for Aerosols (MAIA; Diner et al., 2018) satellite instrument offers an exciting opportunity to derive even more information about aerosol composition by combining measurements at ultraviolet wavelengths with multi-angle observations and polarization sensitivity."

Referee Comment #2

The paper is appropriate for ACP. I have almost nothing to comment. However, the paper is rather lengthy, like a final report of a project. I can imagine that the information is very useful for many aerosol researchers, especially modelers.

However, if possible the results should be presented in a much more condensed form. Positive as well as negative trends are reported, and a large number of trends are found, at the end the reader is left a bit alone what the main conclusions, what the key messages are.

Thank you for the positive review.

We have emphasized that interpretation of the OMI UVAI with a quantitative simulation of the UVAI offers information about trends in aerosol composition. We write in our introduction:

Page 4 lines 115-118: "Comparison of trends in observed OMI UVAI values to the trends in simulated UVAI values, which are calculated using known aerosol composition, enables qualification of how changes in aerosol absorption and scattering could influence the observed UVAI trends."

We also identified areas for model development, such as dust emissions from the desiccating Aral Sea.

We have reformatted and condensed some of the text in sections 3 and 5 in a manner which we believe makes the reporting of the UVAI trends easier to follow for the reader.