Authors' reply to comment of Anonymous Referee #1

We thank the referee for the extensive and critical review of our paper.

The main point that the referee makes is that the UVAI signal in the range ±1.0 contains effects of:
- Improper instrumental characterisation
- Systematic geophysical errors
- Inaccurate RT calculations

We agree with the referee that these effects influence aerosol index results, but we don't agree on the conclusion that they render our SCI insignificant. We estimate that the resulting errors in UVAI are typically in the range of ±0.3, but we note here that SCI signals significantly larger than 1.0 are regularly found in regions where large amounts of scattering aerosols occur (see Figs. 3, 4 and 5-7 in the revised manuscript), signifying that the SCI signal is not an artefact. Systematic errors due to incorrect assumptions in the aerosol index algorithm (i.e., the assumption that surface albedo does not change with wavelength in the UV range) are more serious, therefore aerosol index values in specific regions (deserts and certain parts of the oceans) should be regarded with caution, as pointed out by the referee. It is important to note that the regions where the highest SCI values are found – vegetated land surfaces – are not subject to these systematic errors.
In the following, we will discuss each of the three issues mentioned above and the resulting estimated errors.

Improper instrumental characterisation
The authors are aware of three problems with SCIAMACHY measurements of radiance: (1) the calibration of the absolute radiance is slightly off; (2) a wavelength-dependent degradation of (in particular) the UV channels has been occurring since the beginning of 2004; (3) a scan-angle and wavelength-dependent degradation has been observed from end of 2005/ beginning of 2006.
The first problem has been studied, and is currently being taken care of by multiplying the radiances at 335.5 and 376.5 nm with correction factors calculated by linear interpolation from those determined by SRON for the radiances at 340 and 380 nm (see Tilstra: SCIAMACHY Absorbing Aerosol Index ATBD, SRON 2008).
The second problem is being dealt with in the revised manuscript by applying so-called M-factors to the level-1 data. The M-factors are determined empirically and serve to correct for the gradual wavelength-dependent degradation of the SCIAMACHY radiances (Bramstedt et al., Calculation of SCIAMACHY M-Factors, IUP Bremen, 2007).
We compared daily and globally averaged UVAI and radiances (at 335.5 nm) for the time period under study (2004-2006), and found that the effects of degradation of SCIAMACHY radiance measurements are sufficiently corrected by the M-factors.

The third problem, the scan-angle dependent degradation, becomes significant from the middle of 2006 on (see Snel and Tilstra, SCIAMACHY Scan-angle Dependent Degradation, SRON 2008). The data shown in Figs. 3 and 4 in the revised manuscript (2 and 3 in the old manuscript) are from 2005 and are therefore not significantly affected. Nevertheless, the pixels potentially most affected by scan-angle dependent degradation (the three eastern-most pixels) were removed from the analysis. This did not significantly alter the results. The data shown in the comparison with AERONET AOT values are averaged over the time period 2004-2006 to improve the statistics. Also here the three eastern-most pixels were discarded, and no obvious effect of scan-angle dependent degradation could be observed in the data of 2006 in comparison to the data from 2004 or 2005.

Systematic geophysical errors

As noted by the referee, systematic geophysical errors (we prefer the term "systematic error" to noise, since these effects are not random, and occur under well-defined circumstances) can be caused by: spectral dependence of surface albedo, specular reflection of the sun on the ocean surface, clear water absorption, and ocean colour effects.

Simulations using the RTM SCIATRAN show that a small wavelength dependence of surface albedo (e.g. 0.070 at 335.5 nm and 0.073 at 376.5 nm, as for "water" in: Kleipool et al., OMI surface reflectance climatology, JGR 113, 2008) has only a minor influence on UVAI (0.2-0.3 units). In contrast, the "bare" surface type (Kleipool et al., 2008) has surface albedo = 0.080 at 335.5 nm and 0.095 at 376.5 nm. This causes UVAI errors of up to +1 unit, and is reason for concern: absorbing aerosols may be detected where there aren't any present. We have added a comment to this effect in the revised paper.

Clear water absorption is another cause for concern. It causes UVAI values of up to about +1, and is particularly obvious in the Pacific Ocean around 20°N and around 15°S (see Fig. 2 and 3 in the old manuscript, or 3 and 4 in the revised manuscript). We are currently not correcting for this effect, but have included a comment about the origin of the signal in the revised manuscript.

As pointed out by the referee, ocean colour (caused by the presence of e.g. algae or CDOM) would probably cause negative UVAI values. However, if we compare monthly averaged, cloud-cleared UVAI maps for July, August, and September 2005 with the corresponding monthly averaged MODIS ocean colour maps, no clear agreement is found. Ocean colour might thus be the cause of the SCI signal over the northern Atlantic and Pacific Ocean, but it might as well be caused by clouds or scattering aerosols. This issue needs further investigation in the future. For now, we focus on SCI over land, where these problems do not play a role. Nevertheless, the influence of ocean colour on UVAI is an important issue, and it is now mentioned in the revised manuscript.

Direct reflection of sunlight, or sun glint, is avoided by removing pixels in sun glint geometry from the dataset (see section 5 in the paper, and answer f below).

Although the discussion on albedo effects is important for the correct interpretation of UVAI in general, it is less relevant to the results we presented because our manuscript (and, in particular, the comparison with AERONET measurements) focuses on the detection of aerosols over land. For completeness, we have added a summary of the discussion above to the manuscript.

Inaccurate RT calculations
Inaccuracies in the RT calculations may occur in case of large viewing or solar zenith angles, due to the use of the plane-parallel approximation. To avoid this (and to constrict the viewing geometry dependence of UVAI itself), we only calculate UVAI for SZA below 60° and viewing zenith angle up to 35°.
The errors caused by using an incorrect ozone profile are relatively small for the areas where the highest SCI values are found (as mentioned in section 3 in the manuscript), but may be significant at higher latitudes and will be corrected in the future.
The average surface altitude is calculated for each SCIAMACHY ground pixel separately, and because the dependence of TOA radiance on surface altitude is taken into account in the UVAI algorithm, the error caused by orography was assumed to be negligible. The assumption was tested using the independent pixel approximation for several combinations of surface altitude (e.g., 50% of surface at 0 km and 50% of surface at 1 km altitude, 80% at 0.2 km and 20% at 1.5 km, etc.). The error in UVAI for all plausible tested cases was negligible (less than 0.1 UVAI units), thereby justifying our assumption.
An error is introduced by the use of interpolated look-up-tables, but our simulations have shown that for Rayleigh atmosphere, SZA between 15° and 55° and nadir viewing geometry this error is less than 0.05 UVAI units.

Specific comments:
a. Torres et al [1998] should be included in the section around line 54
The referee is right; the paper is quoted before and after the section, but it is indeed missing from line 54. We have now included the reference.

b. Signature of ocean effects on UVAI is not discussed (line 65)
We have focused on observations over land and have indeed not discussed the effects of ocean colour on UVAI. We have now included it in the section on UVAI sensitivity. We mentioned that in the UVAI calculation it is assumed that surface albedo in the UV range is constant (with respect to wavelength), but had not discussed the potential errors due to wavelength-dependent surface albedo (see above). This point is now also addressed.

c. Model calculations by Torres et al [1998] do not agree with findings that UVAI depends on clouds: where does cloud signal come from?
The referee refers to the results presented in Figure 5 in Torres et al. [1998]. According to these results, large scattering particles cause UVAI values of around -0.1 (for AOT = 1). However, the results from our own model calculations (see Fig. 1 in the manuscript) show that scattering aerosols cause UVAI of -1 to -2 (much larger than in Torres et al., [1998]). Similarly, the large scattering particles (wavelength-independent AOT of 1.0) modelled by de Graaf et al. [2005], shown in their Fig. 3, yield a negative residue of -1.2. De Graaf et al. also explain in detail why scattering aerosols cause negative residues.
It is as yet unclear to the authors where the discrepancy between the model results published in Torres et al., [1998] on the one hand, and those from de Graaf et al., [2005] and ours on the other hand come from. Clearly more detailed future investigations are urgently needed to shed some light on the issue.

The model results presented below and in the manuscript were calculated using both the vector RTM SCIATRAN3.0 and the Backward-Monte Carlo RTM McArtim. The results of the two RTMs agreed very well in this study. On these grounds, and because our results are in excellent agreement with those of de Graaf et al., [2005], we have no reason to doubt our findings.

Following the referee's suggestion, we investigated the effects of clouds on UVAI in more detail.
Clouds have several effects on the observed radiances: they shield the atmosphere below and they increase the path length of the (multiply reflected) light. Interestingly, however, the main effect is probably the so called 'albedo effect': white (bright) surfaces increase the probability of multiple scattering (which depends non-linearly on the (cloud top) albedo, see Fig. S1). This becomes important for partially clouded scenes, which constitute the by far largest part of all SCIAMACHY observations.

For the modelling of thick clouds we use the independent pixel approximation: a satellite observation is assumed to consist of a white (cloudy) part and a black (clear) part. Hence, the radiance of the complete ground pixel depends linearly on cloud fraction at all wavelengths. Consequently, R_{335} also depends linearly on R_{376} for all values of TOA albedo (or cloud fraction), see Fig. S1, blue dashed line.
The radiances calculated for the LUT used in our UVAI algorithm are shown in black in Fig. S1. They were calculated for surface albedo values between 0 and 1. The fact that R335/R376 is not constant for all surface albedo values is due to the non-linear behavior of radiance (w.r.t. surface albedo) as a result of multiple surface and Rayleigh scattering in this wavelength range. This behavior is more clearly seen in Fig. S2, where the difference between the R335 for cloudy and clear sky is plotted against R376.
In Figs. S1 and S2 it can be seen why negative UVAI are found for partially clouded scenes: the contrast between 335.5 nm and 376.5 nm is smaller for partly clouded pixels than for clear pixels with higher surface albedo, or in short: $(R_{335}/R_{376})_{cloud} > (R_{335}/R_{376})_{clear}$. Because for the UVAI calculation $(R_{376})_{cloud} = (R_{376})_{clear}$, it follows that for partly clouded pixels $(R_{335})_{cloud} > (R_{335})_{clear}$, and with: UVAI = $-100*\log[(R_{335})_{cloud}/(R_{335})_{clear}]$ we find that UVAI < 0 for this case.
In conclusion, we find that the major effect of (partial) cloud cover is simply the effect of inhomogeneity within a satellite ground pixel.

The altitude of a cloud thus has little influence on the shape of the UVAI dependence on cloud fraction, although the value of UVAI is affected (see also Figure 2 in the revised manuscript and our reply to referee #2 for the effects of other parameters on UVAI of clouds). The magnitude of UVAI decreases for higher clouds, because the R_{335}/R_{376} ratio for large cloud fraction is smaller than for low clouds (due to decreased amount of Rayleigh scattering occurring in the case of high clouds). This also causes UVAI to become positive for high clouds with large cloud fractions (as mentioned in the revised manuscript).
For thin clouds the reasoning is less intuitive, but very similar to the thick cloud case.

d. How are clouds treated in residue calculation?
The clouds are assumed to be homogeneous clouds (within the clouded part of the satellite ground pixel) with a geometrical thickness of 2 km (see also section 4 in the revised manuscript). The cloud droplets are simulated using a Henyey-Greenstein asymmetry parameter (g) of 0.85, and a single-scattering albedo of 1.00. Thin clouds have geometrical cloud fraction of 1 and varying cloud optical thickness (assumed independent of wavelength). Effective cloud fraction is determined from the radiance calculated at 760 nm (O_2 A-band, similar to the FRESCO cloud retrieval scheme). Thick clouds have a total cloud optical thickness of 50; the radiances for cloud fractions smaller than 1 were calculated using the independent pixel approximation (e.g. for a cloud fraction of 0.2: R(CF=0.2) = 0.2*R(CF=1)+0.8*R(CF=0).
An explanation of the term “effective cloud fraction” was added in the revised manuscript.

e. Authors should compare UVAI with MODIS/MISR AOT, and with ocean colour maps.
This is a good suggestion. We compared maps of monthly averaged MODIS AOT with monthly averaged UVAI maps (with cloud fractions smaller than 5%) for the summer months of the year 2005. The major aerosol events on both maps are in good agreement. There are also some differences, such as the pattern of aerosol loading over China, and the generally low AOT over ocean. Two reasons for these discrepancies are: the dependence of UVAI on aerosol layer height and the fact that the presence of absorbing and scattering aerosols at the same time may cause a blindness of UVAI to aerosols (high SCI + high AAI ≈ 0). Over ocean, cloud remnants and ocean colour effects may also influence the UVAI results. A more extensive comparison would involve taking into account aerosol layer height and aerosol type (absorbing or scattering), and will be performed in the future.
The comparison between monthly averaged UVAI and ocean colour maps was also performed, and is discussed above.

f. Sun glint angle of 18 deg is too small to screen out sun-glint effects.
This is true, although using an angle of 18° does remove the largest portion of sun glint affected region. The angle is a (rather conservative) compromise between removing all effects of sun glint and discarding too many pixels (see also: de Graaf et al, ACP 5, 2005).

g. Clouds do not change the spectral dependence (line 194). The statement by the authors contradicts without any proof previously reported analysis.
As explained in detail above, small and thin clouds cause a significant UVAI signal (see section 4 and Figure 2 in the revised manuscript, and answer c above). No extensive study of cloud effects on UVAI has been published up to date, to the authors’ best knowledge.

h. Authors ignore effects of ocean colour (line 225)
The referee is right; a reference to ocean colour effects has been added (as noted in answer b).

i. The reported weak correlation (AOD vs SCI) is not surprising: signal separation (aerosol from non-aerosol) is less complicated over land than over ocean.
The reason that the comparison with AERONET data was performed with stations over land is mainly because AERONET stations are generally located on land. Nevertheless, the referee is right, and the fact that surface effects probably do not significantly influence our results near the AERONET stations is an extra advantage, making our results more convincing.

Figures

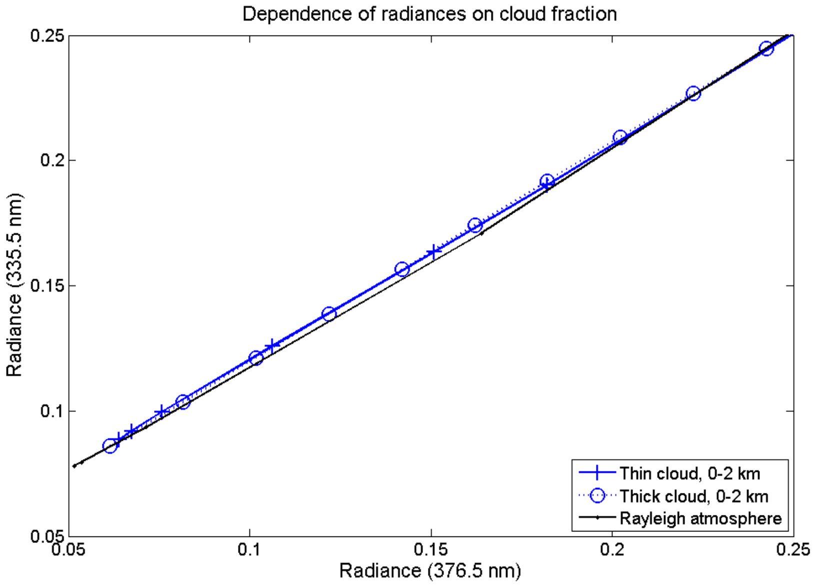

Figure S1. Radiances used for the calculation of UVAI plotted against each other for cloudy and clear-sky cases. The thin clouds (crosses, solid blue line) were calculated as described in the paper, with cloud optical thickness varying between 0.5 and 50, thick clouds (circles, dotted blue line) have effective cloud fractions between 0 and 1. The clouds are located between 0 and 2 km altitude. The clear-sky cases (dots, solid black line) have surface albedo values between 0 and 1. Other simulation parameters: SZA 20°, nadir viewing geometry.

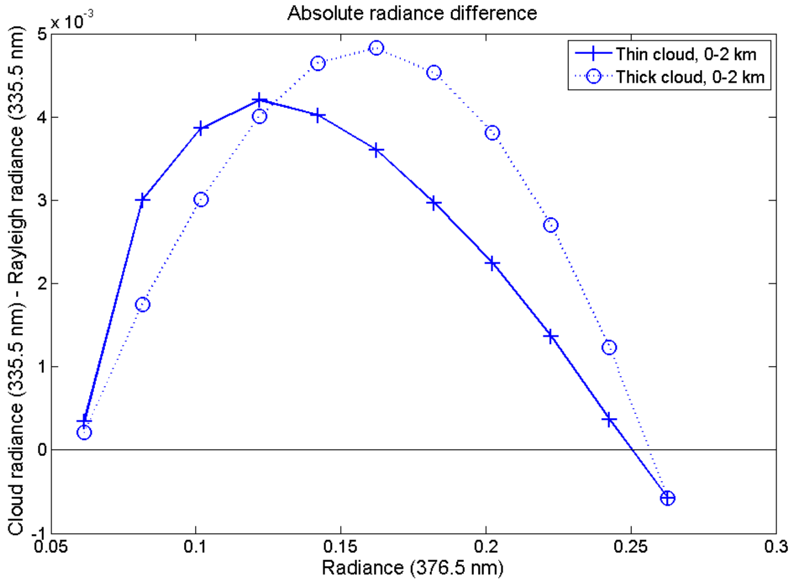

Figure S2. The data from Figure S1 plotted as the difference between cloud radiances and clear radiances at 335.5 nm.