Author’s reply to comment of Anonymous Referee #2

We thank the referee for the constructive review of our paper.

The main point that the referee makes is that not enough convincing evidence is given in the paper that the SCI is a useful indicator of scattering aerosols. He advises to show more comparisons with AERONET measurements and more simulations of UVAI of clouds.
We have followed the advice of the referee: we included more extensive cloud simulations and we added two AERONET stations in Southeast Asia to the comparison. We thus provide additional evidence that the SCI signal indicates aerosols (and not only clouds).

In the following, we give our replies to the reviewer’s specific comments.

Comment 1: More simulation results on the effects of clouds on the SCI are needed, and section 7 should be moved to section 3.
The paper was restructured in such a way that the sensitivity studies in the beginning encompass the cloud studies. We have recalculated the dependence of UVAI on cloud fraction and cloud optical thickness for different asymmetry parameters (0.70-0.90), for different solar and viewing geometries (SZA 20°, 40°, and 60°, viewing zenith angles 0°, 10°, 20°, and 30°; relative azimuth angles 0°, 90°, and 180°), and for more cloud altitudes (0-2 km, 2-4 km, 4-6 km, 8-10 km, and 12-14 km). For clarity, we will not show all results in the paper, but we will recap them here:
- SZA 20° and 40° give very similar results (SZA 40° has slightly smaller UVAI than SZA 20°), UVAI is significantly decreased for SZA 60°, but the trend with cloud fraction is the same as for the other SZA values;
- Changing the viewing geometry causes a small change in UVAI (on the order of 0.1-0.2 units), but does not change the overall shape of the dependence of UVAI on cloud fraction. At SZA 60°, the viewing angle dependence becomes much stronger;
- The asymmetry parameter was set to values between 0.7 and 0.9, which are values roughly representative of aerosols, water clouds, and ice clouds with different ice crystal shapes. Like the viewing geometry, the asymmetry parameter has only a minor effect on the UVAI, and does not change the overall dependence of UVAI on cloud fraction;
- Cloud altitude does not affect the shape of the UVAI dependence on CF, although the magnitude of UVAI decreases with increasing cloud altitude. Interestingly, high, thick clouds cause positive UVAI values under certain conditions, making them appear as absorbing aerosols.
In short, none of the varied parameters (SZA, viewing geometry, cloud altitude, cloud asymmetry parameter) influence the shape of the dependence of UVAI on cloud fraction, but all these parameters affect the magnitude of UVAI.
The revised Figure 2 is reproduced at the end of this supplement.
For more details on the effects of clouds, please have a look at our reply to referee #1.

Comments 2, 3, and 4: The SCI is simply the negative part of the residue. A new term “SCI” is not needed, and only confuses the literature. Similarly, the term “UV Aerosol Indices” is not needed, since it is identical to the term residue, which is also used in the paper.
Currently, three terms are used in the literature: residue or r_λ (e.g. in Torres et al., JGR 1998), UV Aerosol Index or UVAI (e.g. in Torres et al., JGR 2007), and Absorbing Aerosol Index or AAI (e.g. in the PhD thesis by de Graaf). The terms “residue” and “UV Aerosol Index” can be used interchangeably, but the Absorbing Aerosol Index is defined as the positive values of the UVAI.

We have adopted the three terms, and added a new, fourth term to indicate the negative part of the UVAI, called the SCattering Index (SCI).
We agree that it is confusing to use too many terms for a single quantity, but we believe that it is important to stress the fact that the negative UVAI contain information on scattering particles (clouds and aerosols), and that it is therefore a counterpart to the well-known AAI. Instead of dropping the term SCI, we propose dropping the term residue, and referring to AAI and SCI together as UV Aerosol Indices (UVAI).

Comment 5: There should be a comparison to one or more AERONET stations in the regions where the SCI indicates scattering aerosols. Especially in Asia there should be several comparisons.
The comparison with AERONET stations in the Southeast U.S.A. was shown because in this region, the largest aerosol contribution by far comes from scattering aerosols (especially in summer), the aerosols are quite homogeneously distributed over a large area, and they show a strong, clear seasonality. In other regions with high SCI values, AERONET stations were either not present (e.g., the Chinese inland and tropical Africa), or not representative due to their proximity to point sources (e.g., Chinese East coast). The two stations included in the revised manuscript, Mukdahan and Bac Giang, are located in Thailand and Vietnam, respectively, and fulfil the requirements mentioned above.
We have included the figures 5-7 as they will appear in the revised manuscript at the end of this comment.
The AERONET stations in the U.S.A. show high AOT values from scattering aerosols (high SSA) in summer, and the African stations each have high concentrations of absorbing aerosols during their respective biomass burning seasons. In contrast, the Southeast-Asian sites each show two distinctly different seasons, each with high aerosol loadings. This makes the interpretation of the results more complicated than for the other four AERONET stations. In the dry season (particularly from February-April), a lot of biomass burning occurs in Southeast Asia, and AOT caused by locally produced or transported (dark) smoke particles is high. This is seen at both AERONET locations, Mukdahan and Bac Giang, by their large AOT values and corresponding high (positive) UVAI. The low SSA values, indicative of dark smoke particles, are most clearly seen in the data recorded at Mukdahan, where statistics are better (larger dots, see Fig. 7).
In the latter part of the year, approximately from September to November, high amounts of scattering particles are detected at both Southeast-Asian stations. These are probably due to the formation of secondary organic aerosols from volatile organic compounds emitted by plants. The seasonal cycle with high AAI in spring and high SCI in autumn is particularly apparent in Bac Giang, which is also where higher AOT values were measured by AERONET.
The seasonal cycle is not very apparent in the SSA data, which may have several causes. One reason is that SSA at 440 nm may not be representative of SSA in the UV range (which is the case for certain aerosol types; see Sect. 7 in the manuscript, and references mentioned there). Another important reason is that the number of SSA measurements (relative to the number of AOT measurements) is quite small, so that different aerosols may be sampled for the SSA measurements than for the UVAI measurements. It is, however, known from fire counts and other (satellite) data that a lot of biomass burning occurs in spring in Southeast Asia. The high AOT values measured by AERONET in autumn cannot be attributed to either biomass burning or mineral dust particles, and therefore are, most probably, caused by scattering aerosols.

An extra statement was added to the manuscript that reads:

“These examples demonstrate the importance of studying not only the positive UVAI values, but also the negative UVAI values: if only the AAI is studied, no aerosols are detected at the two AERONET stations in the Southeast U.S.A., and in Southeast Asia only the biomass burning aerosols would be observed.”
We also added data from 2004 and from 2006 from all stations to the comparison to improve statistics.

Comment 6: Please indicate which SSA values belong to which aerosol types in section 3.
For Figure 1, a systematic study of the dependence of UVAI on aerosol parameters was performed. For this study, we did not attempt to model certain aerosol types. Generally, a small SSA (smaller than about 0.9) is associated with particles from biomass burning or other incomplete burning processes, but mineral dust usually also has small SSA values in the UV range. Sulphate aerosols and secondary organic aerosols, on the other hand, barely absorb visible light and have high SSA (>0.95). In-between values of SSA may occur for mixtures (either internal or external) of different types of aerosols.
We have included a short statement to this effect in the revised manuscript.

Comment 7: The last paragraph of the abstract should be removed, because this combination is not shown in this paper.
The paragraph was intended to indicate the future applications of the UVAI. It is changed in the revised manuscript.

Comment 8: Why not use the altitude of the scattering particles, as follows e.g. from the SCIAMACHY O_2 A-band, to separate aerosols from clouds?
This is a very good suggestion for future research. We have recently tried to distinguish between clouds and aerosols based on the altitude of the scattering layer, but the results were so far not conclusive (work presented at the Cloud and Aerosol Workshop in Berlin, March 2009). Problems may be caused by: a) the competing effects of clouds/aerosols in on the one hand shielding the trace gases below the clouds and on the other hand increasing the sensitivity for trace gases above the cloud; and b) the dependence of both effects on the accurate value of the surface albedo. It is well known that O_2 band techniques become rather insensitive for small effective cloud fractions or low optical depths. We are still working on improving these techniques and will include a statement in the conclusions of the revised version of the manuscript.

Comment 9: In the abstract and in later sections the term “ “scattering” aerosols” is used, with scattering in quotation marks. Please remove the quotation marks, and clearly define in the beginning what you mean with scattering aerosols and with absorbing aerosols.
The term “scattering” was placed in quotation marks, because nearly all physically relevant aerosol particles scatter more visible light than they absorb. Because, as the referee correctly pointed out, the quotation marks may lead to confusion, we have omitted them in the revised paper and have explained more clearly what our definition of scattering and absorbing aerosols is.

The smaller and textual comments were corrected in the manuscript, except for:
P. 13576, l.11: “The SCI have not been shown ... before.” The residue is shown in operational data products. Please remove sentence.
We believe the referee is mistaken here: in the (TEMIS) AAI products for GOME(2) and SCIAMACHY only positive UVAI values are shown in the downloadable figures, as is the case for OMI and TOMS data. The full UVAI dataset (including negative UVAI) can be

downloaded from TEMIS, but to the authors' best knowledge these negative UVAI data have not been published or discussed in any form yet.

Figures

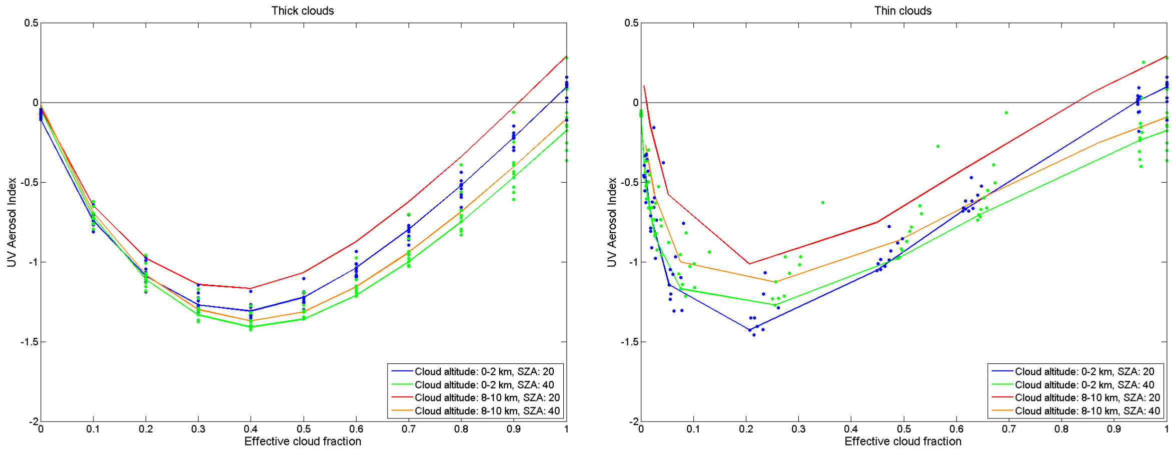

Figure 2. Results from RTM calculations using SCIATRAN 3.0. Cloud parameters: single scattering albedo 1.0, asymmetry parameter 0.85. Left: thick clouds with a total cloud optical thickness equal to 50 (cloud albedo equal to 0.8) with varying geometrical cloud fraction. Right: clouds with varying cloud optical thickness (between 0 and 50) and geometrical cloud fraction equal to 1. Surface albedo was constant, and set to 0.05. Calculations were performed for different SCIAMACHY viewing geometries (viewing angle 0-30°, relative azimuth angle 0-180°) and solar zenith angles of 20° (blue and red) and 40° (green and orange). The clouds were located at 0-2 km altitude (blue and green), or at 8-10 km (red and orange). The solid lines connect the points with nadir viewing geometry.

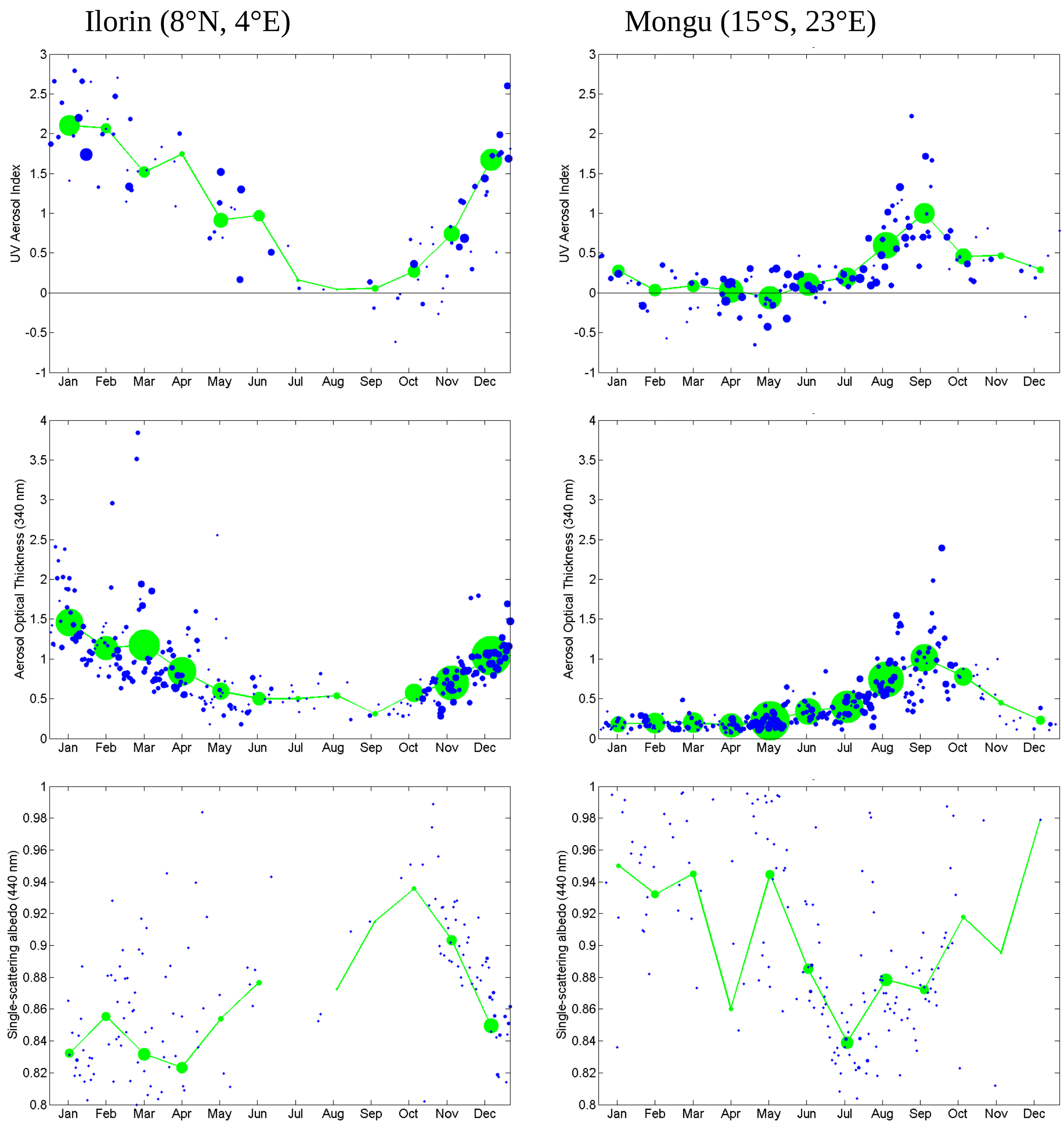

Figure 5. Time series of aerosol parameters at two AERONET stations in Africa: left, Ilorin (8°N, 4°E); right, Mongu (15°S, 23°E). Blue dots, daily averages; connected green dots, monthly averages. The size of the data points indicates the number of measurements included in the average value (minimum value: 1, maximum: 486 (monthly averaged AOT in May at Mongu).

Upper plots: daily and monthly averaged UVAI. Pixels included in the averaging were in a 2° x 2° box with the AERONET station in the centre. Pixels with SZA > 60° or with HICRU CF > 5% were discarded.

Middle plots: AERONET AOT at 340 nm (level 2.0, only data for the orange line in the Walker Branch figure are level 1.5). Measurements included in the average have SZA < 60°, and were measured between 9:30 and 11:30 A.M. (local time).

Lower plots: single-scattering albedo at 440 nm (level 1.5). The same criteria as for AOT measurements apply. Details are given in the text.

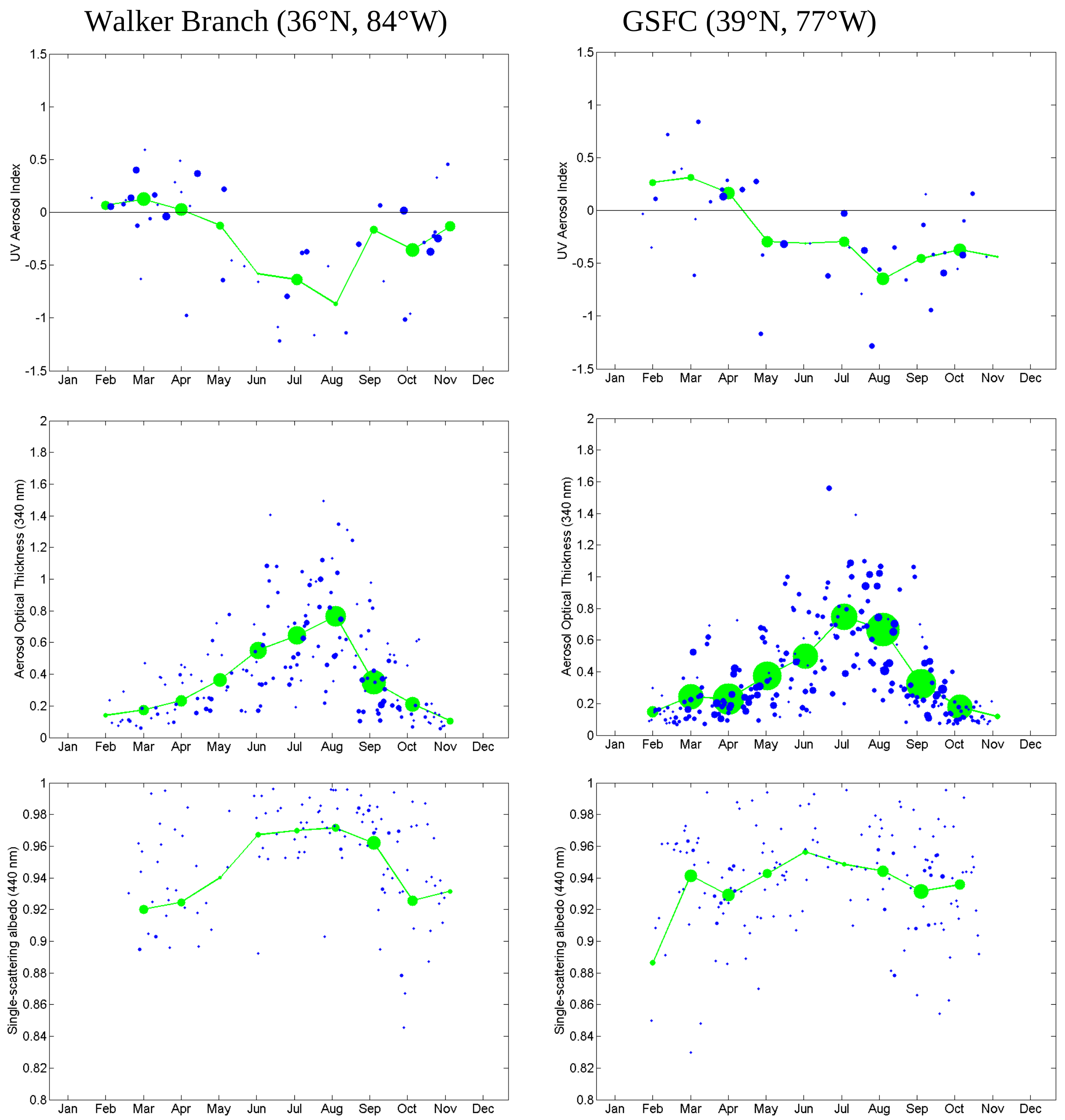

Figure 6. Time series of aerosol parameters at two AERONET stations in Southeast U.S.A.: left, Walker Branch (36°N, 84°W); right, GSFC (39°N, 77°W). Blue dots, daily averages; connected green dots, monthly averages. The size of the data points indicates the number of measurements included in the average value (minimum value: 1, maximum: 355 (monthly averaged AOT in August at GSFC).
Upper plots: daily and monthly averaged UVAI. Pixels included in the averaging were in a 2° x 2° box with the AERONET station in the centre. Pixels with SZA > 60° or with HICRU CF > 5% were discarded.
Middle plots: AERONET AOT at 340 nm (level 2.0, only data for the orange line in the Walker Branch figure are level 1.5). Measurements included in the average have SZA < 60°, and were measured between 9:30 and 11:30 A.M. (local time).
Lower plots: single-scattering albedo at 440 nm (level 1.5). The same criteria as for AOT measurements apply. Details are given in the text.

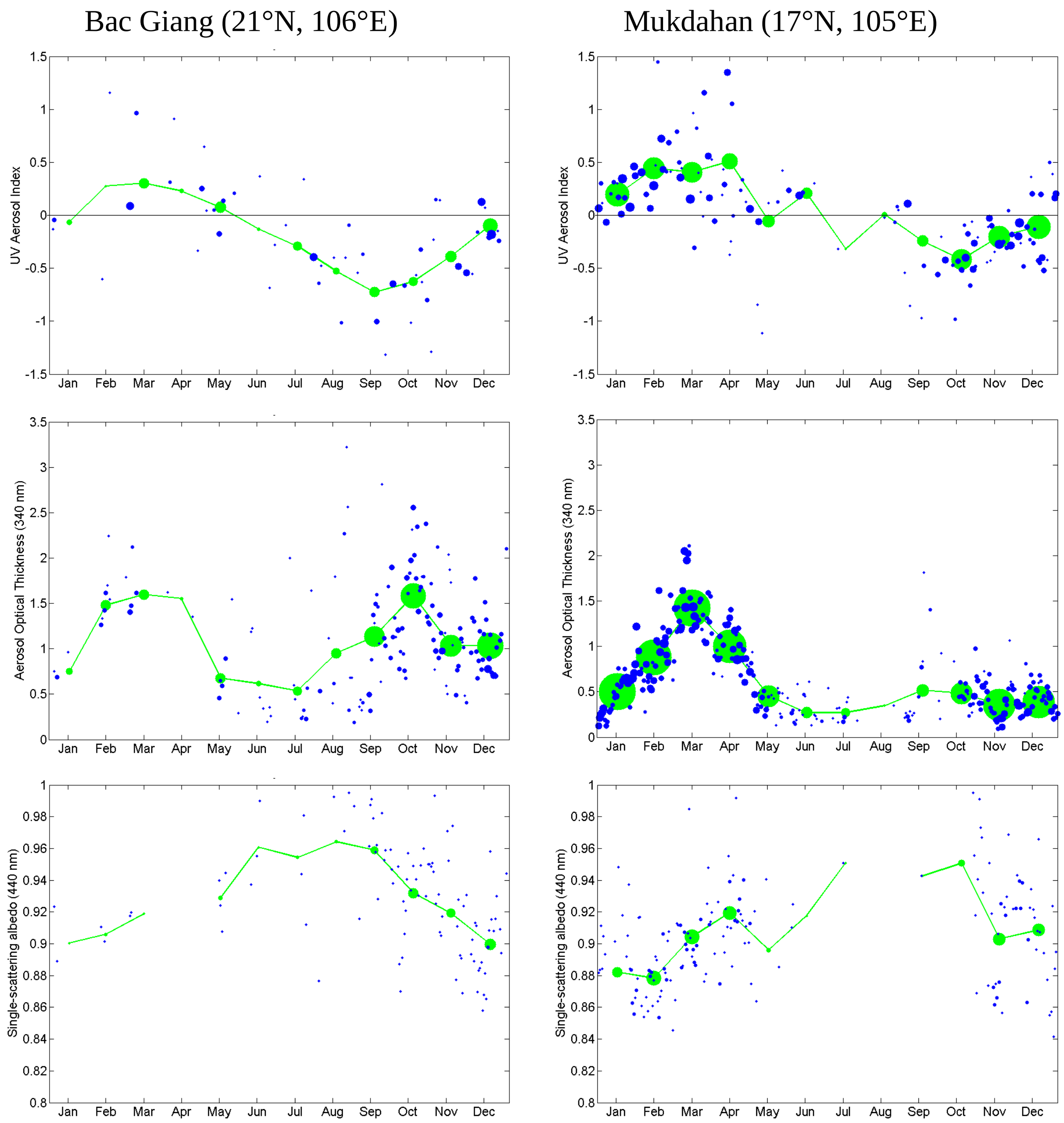

Figure 7. Time series of aerosol parameters at two AERONET stations in Southeast Asia: left, Bac Giang (21°N, 106°E); right, Mukdahan (17°N, 105°E). Blue dots, daily averages; connected green dots, monthly averages. The size of the data points indicates the number of measurements included in the average value (minimum value: 1, maximum: 440 (monthly averaged AOT in January at Mukdahan)).
Upper plots: daily and monthly averaged UVAI. Pixels included in the averaging were in a 2° x 2° box with the AERONET station in the centre. Pixels with SZA > 60° or with HICRU CF > 5% were discarded.
Middle plots: AERONET AOT at 340 nm (level 2.0). Measurements included in the average have SZA < 60°, and were measured between 9:30 and 11:30 A.M. (local time).
Lower plots: single-scattering albedo at 440 nm (level 1.5). The same criteria as for AOT measurements apply. Details are given in the text.