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# Investigation of the accuracy for single scattering albedo retrieval from global UV irradiance measurements

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## Abstract

Retrieval of aerosol optical absorption properties in the UV spectral range such as single scattering albedo (SSA), using remote sensing techniques, is currently an open scientific issue. We investigate the limitations on calculating columnar SSA using a combination of global UV spectral measurements (that are common in various UV monitoring stations worldwide) in combination with radiative transfer modeling. To point out the difficulties in such retrieval we have used the travelling reference spectroradiometer (QASUME) results from 27 visits to UV monitoring stations around Europe, using the QASUME instrument as relative reference, analyzing absolute differences and also temporal and spectral deviations of UV irradiances, that are used as basic input for the SSA retrieval.

The results comparing the mean SSA derived by all instruments, measuring synchronous UV spectra, showed that 5 were within  $\pm 0.02$  difference from the SSA calculated from the QASUME instrument, while 17 were within  $\pm 0.04$ , for the Solar zenith angle of 60 degrees. As for the uncertainty that has been calculated using the  $2\sigma$  standard deviation of the spectral measurements, a mean 0.072 and 0.10 ( $2\sigma$ ) uncertainties have been calculated for  $60^\circ$  and  $30^\circ$  respectively. Based on such results we show that only very few instruments could be able to detect long term SSA changes. However, such measurements/results are useful in order to retrieve SSA at UV wavelengths, a product needed for various applications such as, inputs for modeling radiative forcing studies and satellite retrieval algorithms.

## 1 Introduction

Aerosols affect the Earth's radiative balance both directly and indirectly (e.g. Charlson et al., 1992). Even though the level of understanding of direct effects is higher than that of indirect effects (Rosenfeld and Lensky, 1998) there are still open issues, mainly

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related to the absorption component by complex mixtures of aerosols in highly diverse environments. A comprehensive review, was given by IPCC (2007) and Yu et al. (2006), describing an assessment of the aerosol direct effect, regarding both the current status and those issues that still urgently require further research. Both emphasize, that the significant uncertainties in global columnar single scattering albedo (SSA) may constitute the largest single source of uncertainty in the current modeling estimates of aerosol climate forcing. SSA is the ratio of scattering to total extinction (scattering plus absorption). Since both quantities depend strongly on chemical composition, particle size, state of mixture, relative humidity and wavelength, comprehensive measurements are crucial to understand the effects and thus reduce SSA uncertainties propagated into aerosol radiative forcing estimates. However, the SSA is still very difficult to measure by ground based or satellite remote sensing techniques and in-situ methods (Corr et al., 2009). A large portion of this difficulty emerges from current weakness to account for accurate aerosol absorption measurements (AAPCI, 2009; Bergstrom et al., 2007).

The most common method for the determination of aerosol optical properties like SSA in the visible spectral range using measurements of sun and sky radiances by sun photometers and radiometers have been introduced to the Aerosol Network (AERONET – <http://aeronet.gsfc.nasa.gov>). It is based on the retrieval of aerosol optical and microphysical properties using inversion techniques applied to observations and the angular distribution of sky radiances at visible wavelengths demonstrated by Dubovik et al. (2002). Kassianov et al. (2005) and Krotkov et al. (2005a) have proposed measurements of direct and diffuse irradiance combined with radiative transfer modeling (RTM) for the retrieval of SSA. Compared to the visible range, use of such methods for the determination of SSA in the ultraviolet (UV) are more difficult, due to enhanced molecular scattering and the effect of NO<sub>2</sub>, O<sub>3</sub> and SO<sub>2</sub> absorption (Krotkov et al., 2005b). In addition, irradiance and radiance measurements at UV wavelengths require more accurate instrument characterizations and accuracy of determining various, instrument related, parameters following guidelines for instrument specifications (Seckmeyer et al., 2001) and quality control procedures (Webb et al., 1998) such as:

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the wavelength and absolute calibration, the measurement signal to noise ratio, stray light effects and filters stability (Bais, 1997).

There are only few publications dealing with measurement techniques for SSA retrieval at UV wavelengths from which column average absorption can be inferred using two basic methodologies to determine SSA. They both use UV measurements and aerosol optical depth (AOD) information as inputs in an RTM code. The first is using global spectral irradiances (GSI), (Kylling et al., 1998; Bais et al., 2005; Ialongo et al., 2010) and the second (Petters et al., 2003; Krotkov et al., 2005b) is using measurements of global and diffuse irradiance ratios (GDIR) from UV multi-filter rotating shadow-band radiometers. However, all these retrievals have not been sufficiently validated yet. The main difference of these two approaches is that the GSI method requires accurate absolute calibrated global irradiance measurements and the GDIR method requires accurate relative measurements of direct and diffuse irradiance.

Taking into account the amount of UV spectroradiometers (especially Brewer type) that are used for monitoring purposes worldwide (e.g. Bais et al., 2001; Gröbner et al., 2005), we aim to investigate the possibility and the related accuracy in order to use such measurements for retrieving SSA at UV wavelengths. Such measurements with the use of the GSI method, can provide information on long term and short term SSA variability at various sites. In addition it is an essential tool for RTM input used as inputs either for radiative forcing (e.g. Hatzianastassiou et al., 2007; Kazadzis et al., 2009) or satellite retrieval algorithms (e.g. Arola et al., 2009). However, the GDIR method is impossible to be used for such purposes as only very few of such scanning instruments have the opportunity to measure both global and direct spectral irradiance. Even if practically Brewer spectroradiometers are able to perform such measurements (e.g. Kazadzis et al., 2005; Groebner and Meleti, 2004) only very few of them include them in their measuring schedule mainly due to difficulties to derive the absolute calibration of the direct spectral irradiance (Bais et al., 2005). In this work we are investigating the limitations and the level of SSA retrieval accuracy through global UV measurements (GSI method) for investigating the possibility to use such measurements

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from long term UV monitoring stations. Final aim is to assess the retrieval accuracy in order to investigate the possibility of using such measurements for deriving time series of aerosol absorption characteristics through the calculation of SSA.

## 2 Instrumentation and data

### 2.1 Spectral UV measurements

In order to achieve the above mentioned goals we have used the European travelling reference spectroradiometer (QASUME) results during the years 2002–2005. We have to point out that this instrument was used in this work as a relative global irradiance standard and not as an absolute irradiance standard, measuring with strict protocols and maximum control effort for the period of the EU project QASUME (<http://lap.physics.auth.gr/qasume/>). Its measurements were used in order to analyze absolute differences and deviations of synchronized global irradiance measurements measuring in parallel, at the same atmospheric conditions, with 27 UV monitoring spectroradiometers (Gröbner et al., 2006).

The QASUME instrument is a part of the European Ultraviolet Calibration Center that aims to improve the data quality in the European Global Atmospheric Watch UV network, to harmonize the results from different stations and monitoring programs, in order to ensure representative and consistent UV radiation data on a European scale. The transportable instrument approach was the main idea behind the EU project QASUME (2002–2004). The main idea behind this project was, that despite the progress made in the various inter-comparison exercises since 1991 (e.g. Bais et al., 2000; Webb et al., 2003), this inter-comparison process has several limitations and faults as a means of quality assurance (Bernhard and Seckmeyer, 1999). The QASUME reference spectroradiometer system has been previously described in detail in Gröbner et al. (2005). During the period of the QASUME project, 27 UV monitoring sites were visited with the transportable QASUME reference spectroradiometer. A summary of

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the sites and the site instruments can be found in Table 1 and Fig. 1 of Groebner et al. (2006). The protocol of the measurements at each site are described in detail in Gröbner et al. (2003a,b, 2004b). In addition all individual visits of the QASUME instrument are accompanied by a scientific audit of achievements, problems and a final assessment for each visited site. All the audit-documents can be found at: [http://www.pmodwrc.ch/euvc/euvc.php?topic=qasume\\_audit](http://www.pmodwrc.ch/euvc/euvc.php?topic=qasume_audit).

## 2.2 SSA retrieval

The attenuation of solar UV irradiance reaching the earth's surface by aerosols is mainly linked with aerosol optical properties such as AOD and SSA and in addition depends on solar zenith angle (SZA) at the time of the measurement. To demonstrate this effect in Fig. 1 we have calculated with the help of LibRadTran (Mayer and Kylling, 2005) radiative transfer code this attenuation for two solar zenith angles. Using such results and based on the methodology of Bais et al. (2005), we have constructed look up tables (LUT) of this UV attenuation as a function of AOD, SSA and SZA. With the help of these LUT's (as described in the GSI method) someone can calculate the SSA which matches a measured UV attenuation for certain (also measured) AOD at a given SZA (Bais et al., 2005). It is clear from Fig. 1 that for higher solar zenith angles the attenuation for the same AOD's and SSA's is higher. So the uncertainty of the GSI method decreases for higher SZA's and AOD's and for lower SSA's.

In this work we are aiming to investigate realistic (with the use of QASUME campaign results) limitations in the accuracy for the SSA retrieval using the GSI method linked with the spectroradiometer's global irradiance measurement's deviations. In all the 27 inter-comparison campaigns, the UV monitoring spectroradiometers have been visited by the QASUME instrument in order to analyze possible sources of UV measurement deviations at each visited site. More specifically we have used these results in order to investigate how absolute differences and standard deviation of the inter-comparison periods could affect the GSI method for SSA retrieval. In Fig. 2 we have summarized the mean differences and the  $2\sigma$  (standard deviation) of all visiting instruments shown

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as ratios of any instrument with the QASUME at two UV wavelengths

As it is shown in the above Fig. 2, 23 out of 27 instruments mean differences are within  $\pm 5\%$  compared with the QASUME instrument. In addition, deviations from this mean difference are shown. The reasons of such deviations of two spectroradiometers measuring side by side and performing synchronized spectral measurements are described in detail in (Bais et al., 1997, 2000; Gröebner et al., 2004, 2005). The instruments at each campaign were measuring about 20–30 spectral measurements per day for a period of 2–4 days depending on the site. The results used here could be used as a unique realistic example of the accuracy of such measurements in a very well organized campaign.

The analysis in order to investigate the uncertainty budget in order to calculate SSA using the GSI method was based on the:

1. Absolute mean difference of the two spectroradiometers
2. Standard deviation of the differences during the inter-comparison period

The methodology used in order to calculate the difference in the retrieved SSA from the two instruments was the following:

The LUT's that have been calculated with the LibRadTran RTM were used., with input parameters: the spectral measurements of each instrument, the solar zenith angle of every measurement and an AOD (for each location and for the specific month of the measurement campaign) based on the aerosol Climatology database of AeroComm (Kinne et al., 2009). The AOD that was used was constant during the whole campaign period at each site. This was decided, as the main interest of this work was to calculate the SSA retrieval uncertainty only from UV spectral measurement deviations and not to mix other sources of uncertainties such as ones related with RTM input parameters. The latest will also be discussed in the conclusion section. In addition, synchronized AOD measurements were not available for a number of the sites analyzed.

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The methodology for calculating SSA from global UV irradiance measurements illustrated in Fig. 3. There, as an example a  $-3\%$  difference and a  $3\%$  ( $2\sigma$ ) standard deviation between the QASUME and a specific spectroradiometer (SP) at 340 nm have been used (crosses), with an AOD of 0.44 and an SSA of 0.9, for two different solar zenith angles of  $30^\circ$  and  $60^\circ$  (red and blue line respectively). The figure shows that for the case of  $60^\circ$  the instrument SP would lead to an SSA calculation from 0.825 to 0.9 with a mean value of 0.865. Similar for  $30^\circ$  it would calculate an SSA from 0.795 to 0.9 with a mean of 0.845.

AOD and irradiance differences shown here were chosen as they were the mean values for the 27 sites analyzed. However, Fig. 3 is just used as an example to demonstrate visually the methodology used here. Dash lines represent other AOD values. It is shown that higher AOD values (blue dash line with an AOD of 0.7 at 340 nm) lead to better accuracy for the SSA retrieval, while very low AOD values (red dash line with an AOD of 0.1 at 340 nm) lead to a very high uncertainty. The same procedure has been repeated with all instruments for the UVB (310–325 nm) and UVA (325–360 nm) spectral range using the results demonstrated in Fig. 2.

### 3 Results and conclusions

As shown in Fig. 2 there were 4 instruments that showed mean deviations outside the  $\pm 5\%$  area compared with the QASUME instrument. Such deviations were due to calibration problems or instrument failures at the time of the inter-comparison so we have excluded them from the following statistical analysis. The results of the 23 remaining stations comparing the mean SSA derived by both instruments showed that 5 out of 23 were within  $\pm 0.02$  difference from the QASUME instrument, while 17 were within  $\pm 0.04$  for the SZA of 60 degrees. As for the uncertainty that has been calculated using the  $2\sigma$  standard deviation of the spectral measurements, a mean 0.072 and 0.10 ( $2\sigma$ ) uncertainties have been calculated for  $60^\circ$  and  $30^\circ$  respectively. The results from the 23 stations are shown in Fig. 4.



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The differences in the mean value but also the  $2\sigma$  deviations shown in Fig. 4 are affected from various factors. Main factor is the absolute difference and the standard deviation of the global spectral UV irradiance measurements between each instrument and the QASUME instrument. It has to be clarified that the QASUME instrument is not used here as the “absolute truth” concerning absolute irradiance UV measurements, but mainly as a stable, quality assured instrument, that can determine relative deviations among the instruments operating at the visited UV monitoring stations around Europe. Also, an instrument that can be used for homogenizing both UV spectral measurements around Europe but also by-products such as the SSA retrieval presented here. Analyzing the results at each site, the SSA deviations are affected by the different optical path inside the aerosol layer (SZA effect) and the absolute AOD value used for each site. This last factor shows that for cases of sites with low AOD’s the retrieval of SSA becomes highly uncertain. Similar results for one site has been presented in Bais et al. (2005).

Moreover, additional sources of errors in the SSA calculations at a particular site are the uncertainty of RTM input parameters such as:

- AOD measurement: Calibrated sun-photometers can provide AOD measurements with an uncertainty of 0.05. Using satellite aerosol AOD information or other model based AOD databases, the uncertainty increases due to spatial AOD features especially at urban areas and temporal ADO variability that can not be easily provided with this means.
- Assymetry parameter, aerosol profile and extraterrestrial spectrum uncertainties: According to Bais et al. (2005) this uncertainty can be within 3% for Thessaloniki, Greece, a place with a complex environment in terms of aerosol type and different aerosol transported and local sources.

A recent study (Ialongo et al., 2010) has been using the GSI method for the retrieval of SSA using UV global spectral measurements at solar (local) noon. In such studies due to the SZA variations during the year and also due to the AOD annual variations, the

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uncertainty for calculating the SSA is not constant (proportional to UV global irradiance measurement uncertainties), but depends on the SZA and AOD of each individual retrieval.

It has been mentioned that the calculation of the columnar (effective) SSA using the GSI or the GDIR method are at the moment the only existing methods for calculating this parameter in the UV region. We decided to analyze the possibilities to use the GSI method because of the number of UV monitoring station that have been performing global UV measurements from the start of the 90's. On the contrary only very few stations (starting also after the year 2000) perform simultaneous global and direct measurements. So, only results from the GSI method can be used in order to investigate possible long term SSA changes in a number of locations worldwide. Such changes can enlarge or diminish the effects of AOD changes in UV radiative forcing. Recent studies especially for Europe and North America showed a certain decrease of AOD values (Norris and Wild, 2009; Ohmura, 2009; Ruckstuhl and Norris, 2009). In parallel with that effect, a possible change to less absorbing aerosols would have a negative feedback/effect on the calculated UV increase due to the negative AOD change.

The results shown here demonstrate the possibility to use the retrieved SSA using the GSI method for aerosol related studies. For example for exploring aerosol absorption trends in the UV, only few instruments around Europe could be used to achieve this goal. This is demonstrated by the fact that the mean difference and the  $2\sigma$  variability of the SSA retrieval among two synchronized instruments is in the order of the magnitude of the possible trends that could be detected. For regional or global radiative forcing studies in the UV region, such measurements accompanied with their uncertainty, become very important as they are the only existing methodology at this point. In addition, satellite aerosol and radiation retrieval algorithms could use such time series in order to improve the aerosol absorption assumptions used as input optics in their retrieval schemes. As an example the Ozone Monitoring Instrument (on board of Aura satellite) surface UV retrieval recently introduced a post correction methodology that needs aerosol effective SSA worldwide (Arola et al., 2009).

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Finally, the data used here are the results of very well organized inter-comparison campaigns of very sophisticated spectral instruments. The direct comparison of two well characterized instruments measuring synchronous UV measurements under the same conditions and having the maximum effort from the local operators are making this comparison results the best available in terms of data quality. Such results were used here as a tool in order to realistically assess the possibilities of the SSA retrieval. Based on the above facts, any operator of other (broadband or filter) instruments that measure global UV irradiance has to put much more effort in order to reach the absolute accuracy/long term stability, similar to the spectroradiometers, in order to retrieve SSA with similar uncertainty budget.

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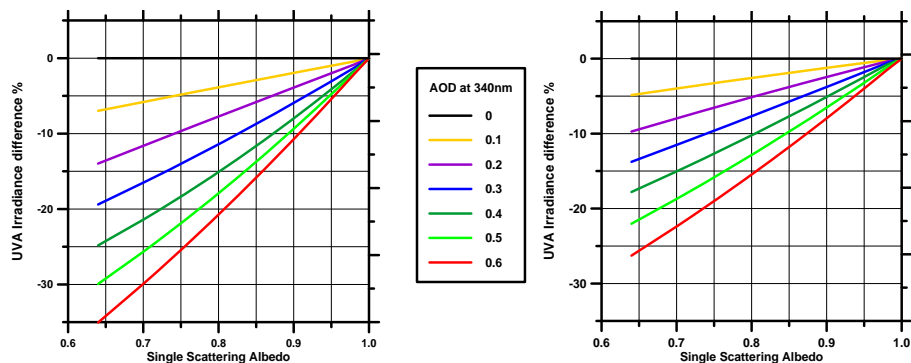
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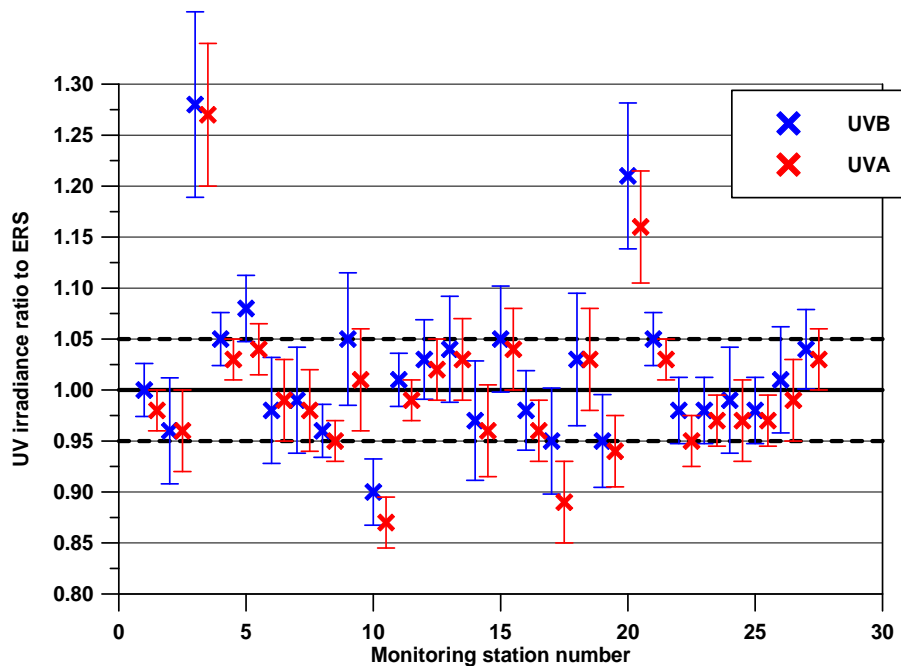
**Fig. 1.** Per cent attenuation of UV irradiance at 340 nm as a function of different AOD and SSA for SZA's of 30° (left) and 60° (right).

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**Fig. 2.** Mean differences and the  $2\sigma$  (standard deviation) of all 27 visiting instruments shown as ratios of any instrument with the QASUME at UVB and UVA wavelengths.

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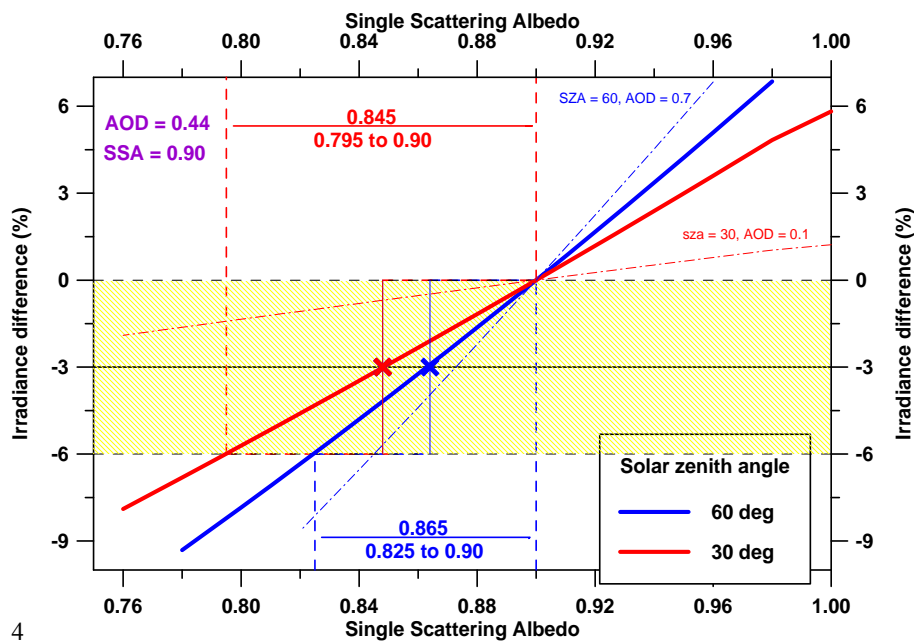
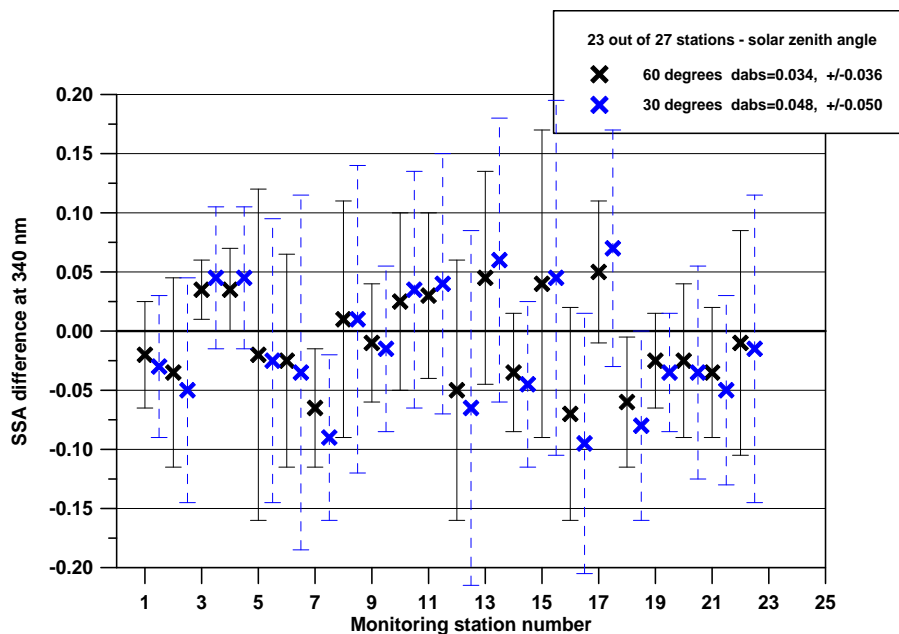


Fig. 3. Retrieval GSI methodology for calculating effective SSA.

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**Fig. 4.** Results of SSA differences between each SP and the QASUME instrument for two solar zenith angles.

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