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# Deriving an effective aerosol single scattering albedo from spectral surface UV irradiance measurements

A.F. Bais, A. Kazantzidis*, S. Kazadzis, D.S. Balis, C.S. Zerefos, C. Meleti

*Laboratory of Atmospheric Physics, Aristotle University of Thessaloniki, Campus Box 149, 54124 Thessaloniki, Greece*

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**Abstract**

The modification of clear-sky global and diffuse irradiances and the direct-to-diffuse irradiance ratio by the aerosol single scattering albedo, $\omega$, as a function of aerosol optical depth, $\tau_a$, and solar zenith angle (SZA) is investigated using radiative-transfer model calculations. The model-derived relations are combined with UV irradiances at the surface and the aerosol optical depth measured with a Brewer MkIII spectroradiometer, with the aim to develop and test an indirect method of estimating an effective single scattering albedo. The uncertainties introduced from different sources are quantitatively discussed in order to determine the overall uncertainty of the method. The overall accuracy in determining $\omega$ indirectly depends strongly on the radiation quantity used and the amount of aerosols, increasing the uncertainty of the method in situations with $\tau_a$ at 340 nm below 0.2. From the three radiation quantities, the direct-to-diffuse irradiance ratio provides the highest accuracy in the estimation of the effective $\omega$. As an example, the effective $\omega$ is determined for 2 days with different aerosol loadings and composition. Finally, 5 years of measurements of global irradiance and $\tau_a$ under cloud-free conditions are analysed in order to estimate the range of variation of the effective $\omega$ at Thessaloniki, Greece. For $\tau_a$ at 340 nm larger than 0.8, $\omega$ ranges between 0.85 and 0.99, while for lower-aerosol optical depths the derived effective $\omega$ covers the entire range from 0.64 to 0.99.
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*Keywords:* Aerosol optical properties; Single scattering albedo; UV irradiance

## 1. Introduction

In recent years, great effort has been put in improving the simulation of the transfer of ultraviolet radiation through the atmosphere with detailed computer models. It has been recognized that the models are capable of estimating the ultraviolet irradiance at the Earth's surface under cloud-free conditions with accuracy of better than 10% (Wang and Lenoble, 1994; Zeng et al., 1994; Forster et al., 1995; Mayer et al., 1997; Weihs and Webb, 1997; Kylling et al., 1998; Kazantzidis et al., 2001) which is comparable to the uncertainties of most of the existing UV measurements. Apart from the uncertainties related to the precision of the radiative-transfer algorithms, the accuracy in the input parameters (e.g. ozone, aerosols, surface albedo) used in model calculations has been discussed extensively (e.g. Weihs and Webb, 1997; van Weele et al., 2000) and constitutes one of the major sources of uncertainty in simulations with radiative-transfer models.

Although the accuracy in measuring the total ozone and the aerosol optical depth, $\tau_a$, is nowadays sufficiently good, measurements of the optical properties of the aerosols (like the single scattering albedo, $\omega$, and the

*Corresponding author. Tel.: +30 2310 998049; fax: +30 2310 248602.
*E-mail address:* akaza@auth.gr (A. Kazantzidis).

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asymmetry factor and phase function) are either nonexistent or inadequate to represent their vertical variability, since they are mostly conducted near the surface. The importance of these parameters in modifying the transmission of ultraviolet radiation through the atmosphere has been already recognized and several studies (e.g. Kazantzidis et al., 2001; Slusser et al., 2001) were devoted to the investigation of their effects with respect to solar zenith angle (SZA), $\tau_a$, wavelength, and surface albedo. Slusser et al. (2001) compared measured UV irradiances at two sites of the USDA UVB Monitoring and Research Network with model calculations and attributed their differences to differences in $\omega$. The accuracy of TOMS-derived monthly erythemal exposure estimates changes from $\pm 6\%$ to $\pm 12\%$ under UV-absorbing aerosol plumes, consisting mainly of dust and smoke (Herman et al., 1999). Thus, the estimation of aerosol attenuation characteristics could lead to reduction of errors in satellite-derived surface UV irradiances (Kroktov et al., 1998).

Indirect methods for the determination of the aerosol properties from solar irradiance measurements have been proposed and tested in several studies, using either global irradiance spectra (Kylling et al., 1998; Kazantzidis et al., 2001), or the direct-to-diffuse irradiance ratio at selected wavelengths (King, 1979; Slusser et al., 2001) or radiances measured by satellites (Herman et al., 1999; King et al., 2003). Using data from the PAUR campaign, Kylling et al. (1998) concluded that $\omega$ affects largely the comparison between modelled and measured global UV irradiance spectra, and determined daily values of ω which led to better agreement of model calculated spectra with measurements. With the same technique, Kazantzidis et al. (2001) examined the relationship between model-derived $\omega$ and local weather conditions (wind speed and relative humidity) during the SUSPEN campaign. King (1979) applied a statistical procedure (King and Herman, 1979) in order to determine ground albedo and the effective imaginary term of the complex refractive index of atmospheric particulates from narrow-band measurements of downward hemispheric diffuse and global flux and radiative-transfer computations, as suggested by Herman et al. (1975). A similar technique was used (Yu et al., 2000) to estimate the single scattering albedo in south-eastern United States. Eck et al. (1998) combined broadband and spectral irradiance measurements, and measured aerosol optical properties (optical depth, asymmetry parameter and size distribution) in Amazonia with model calculations to retrieve $\omega$ for biomass burning aerosols. Petters et al. (2003) using measurements of global and diffuse irradiance and model calculations provided estimates of $\omega$ in the UV, suggesting that due to uncertainties in measurements and model input parameters this process works best for more turbid atmosphere ($\tau_a > 0.3$).

Aerosol properties can also be determined by inverting sky radiance measurements in the almucantur and direct sun measurements from specially designed photometers, such as those of the AERONET network (Holben et al., 1998; Dubovik and King, 2000). The error in the retrieval of $\omega$ at 440 nm from AERONET data is between 0.03 and 0.07, depending on the type and optical depth of aerosols (Dubovik et al., 2000).

Balis et al. (2003, 2004) investigated the combined use of the extinction to backscatter ratio measured by a Raman lidar system in the UV and $\omega$ derived from Brewer global irradiance measurements at Thessaloniki, Greece, for improving the characterization of aerosols from local pollution, Sahara dust and biomass burning.

Because they are based on measurements conducted at the ground, these indirect methods produce estimates of an effective single scattering albedo, being incapable to reproduce the actual variability of $\omega$ inside the aerosol column.

In this study, we investigate quantitatively the sensitivity of different solar UV radiation components to changes in $\omega$ under clear-skies, by combining spectral measurements of global and direct irradiance with model calculations. The final goal is to propose an indirect method for estimating the effective $\omega$, using the derived relations from the model and measured irradiances at the surface, and to investigate the limitations of its applicability, taking into account the uncertainties in the measurements and the model calculations. As an application of this methodology, global irradiance and $\tau_a$ at 340 nm measured at Thessaloniki during the period 1998–2002 with a Brewer MkIII spectroradiometer are combined with model calculations to derive the typical for the area range of variation of the effective $\omega$ under cloud-free conditions.

## 2. Spectral measurements

The spectral measurements used in this study were recorded during 1998–2002 with a double monochromator Brewer MkIII spectroradiometer, which operates regularly at Thessaloniki, Greece (40.5°N, 22.97°E). Usually, the principal component measured by these spectroradiometers is the global irradiance incident on a horizontal surface and additionally at some stations the direct irradiance (Huber et al., 1995; Bais, 1997). The latter can be used to derive the optical depth of the aerosols (Huber et al., 1995; Marenco et al., 1997; Kylling et al., 1998) in the operational spectral region of the instrument, which typically extends between 290 and 365 nm. The diffuse component of solar radiation can be in principle derived as the difference between the global and the direct irradiance. However, in scanning instruments, like the Brewer, the two spectra of global and direct irradiance are measured one after the other with a

time lag of several minutes. Hence, to derive the diffuse irradiance it is necessary to apply interpolation techniques for transforming one of the two spectra (usually the direct) to correspond to the time the other was acquired. Such techniques usually introduce additional uncertainties. Since 1995, the scanning procedure of the Brewer operating at Thessaloniki was modified to obtain samples of the direct component at eight fixed wavelengths (every 10 nm starting from 290 nm) during global irradiance scans, which are used for estimating the cosine correction of the global spectra (Bais et al., 1998). These measurements of the direct irradiance are practically synchronous with the global irradiance measured at the same wavelength (their time difference is the order of 5 s), allowing the computation of the diffuse component at these eight wavelengths.

The absolute calibration of the global irradiance spectra is achieved through the use of 1000-W standards of spectral irradiance, traceable to the National Institute of Standards and Technology standards, following internationally acceptable procedures. The uncertainty in global irradiance measurements is estimated to within $\pm 5\%$.

The calibration of the direct irradiance is derived by comparing the direct irradiance, as measured by the instrument, to the difference between global and diffuse irradiance, which are measured quasi-simultaneously (Bais, 1997) during selected days with clear skies and relatively low aerosol. The diffuse component is obtained from global irradiance measurements by shading the diffuser with a disk. The absolute calibration derived by this method is likely to be affected by the amount of aerosols, although the calibration factors determined by a number of independent measurements at Thessaloniki were consistent within the expected uncertainties of the measurements. Such measurements are performed at Thessaloniki on the average once every 1 or 2 months and the data used in this study are calibrated with this method. The direct calibration uncertainty is estimated to about $\pm 5\%$, and it is the result of combining the uncertainty of the global irradiance calibration and the uncertainty of the shadow-disk measurement procedure (i.e. the transfer of global irradiance calibration to the direct port of the Brewer).

The spectral aerosol optical depth $\tau_a(\lambda)$ is determined by comparing the measured cloud-free spectral direct irradiance at the surface to the direct irradiance which is calculated from the transfer of the high-resolution extraterrestrial spectrum ATLAS 3 (VanHoosier, 1996) through the atmosphere using the Beer's law and accounting only for molecular absorption (mainly $O_3$ and $SO_2$) and Rayleigh scattering. The calculations are performed at the resolution of the ATLAS 3 spectrum and the derived high-resolution irradiance at the ground is convoluted with the instrument's slit function to derive finally the optical depth. In the following any reference to $\tau_a$ refers to a wavelength of 340 nm, unless otherwise specified.

## 3. Radiative transfer modelling

In this study, the UVSPEC model (Mayer et al., 1997; Kylling et al., 1998) from the LibRadTran package version 0.99 (http://www.libradtran.org) was used to conduct a sensitivity study of the relation between different radiation quantities and $\omega$ as a function of aerosol optical depth and SZA. In the model calculations we assumed that the vertical distribution of $\omega$ is invariant, since the effective $\omega$ derived by the method presented here is representative of the total aerosol column only. UVSPEC solves the radiative-transfer equation using the pseudo-spherical discrete ordinates algorithm (Dahlback and Stamnes, 1991; Stamnes et al., 1988) running with 16 streams. Irradiance spectra were calculated in 0.15 nm steps and resolution and then they were convoluted with the slit function of the Brewer to match the measurements at 340 nm.

The atmospheric composition and structure used in the model is based on measured parameters (total ozone, $\tau_a$ at 340 nm) and on profiles taken from the literature. The AFGL midlatitude winter profiles were used for ozone, temperature and air pressure (Anderson et al., 1986). Ozone and air-pressure profiles were rescaled to match the measured ozone column and the mean surface pressure of the station, respectively. For the aerosol vertical distribution we used the Elterman (1968) profile which was scaled to match the measured optical depth at 340 nm.Taking into account the expected impact of the surrounding area on the composition of the aerosol loading, we used Mie calculations to generate a phase function that corresponds to urban aerosol type, as described by d' Almeida et al., 1991. This phase function results to an asymmetry parameter of 0.7 at 340 nm, which was assumed to be constant with altitude for all days considered. The presence of maritime aerosols, which should be expected at Thessaloniki, would not change significantly the proposed asymmetry parameter value. d' Almeida et al. (1991) report values between 0.66 and 0.77 for maritime clean and polluted aerosols and only for maritime mineral aerosols these values are larger, up to 0.84.

The area around the site where the measurements are performed is covered mainly by buildings with concrete roofs, extending for several kilometres to all directions. A few kilometres north-eastwards the area is mountainous, partially covered by pine trees, while the sea extends from a distance of 1 km to the WSW direction. This type of landscape suggests a relatively small surface albedo (Blumthaler and Ambach, 1988), thus a value of 0.03 was used, constant in the entire UV spectral region.

Rayleigh scattering cross-sections were calculated according to the analytic function proposed by Nicolet (1984). Finally, the high-resolution ATLAS 3 spectrum was chosen as the extraterrestrial solar flux, shifted to air wavelengths, and used in the model calculations at the highest available analysis (0.05 nm). Corrections to the extraterrestrial spectrum due to the elliptic orbit of the Earth around the Sun were calculated following the formulation by Spencer (1971).

## 4. Effect of single scattering albedo on UV irradiance

The single scattering albedo influences directly the diffuse radiation, since it describes the proportion of the scattered photons in a beam to the total attenuation (scattering plus absorption). In contrast, its effect on the direct irradiance could be considered negligible, especially at moderate-to-small SZAs and low-aerosol optical depth, when the fraction of diffuse radiation in the beam is very small. Calculations with the UVSPEC model revealed that for the Brewer's field of view (~2.5°) the diffuse component in the direct irradiance at 340 nm is about 0.5% at 60° SZA and $\tau_a = 0.8$. Thus, from the radiation quantities that are potentially measured at a station, it would be expected that variations of $\omega$ may influence the global and the diffuse irradiance, as well as the ratio of the direct-to-diffuse irradiance. These effects can be quantitatively described by radiative-transfer model calculations.

Based on model calculations, Fig. 1 illustrates the effect of $\omega$ variations on global irradiance, diffuse irradiance and direct to global irradiance ratio at 340 nm, under clear skies, for a range of $\tau_a$ from 0 to 1.2 in increments of 0.2 and for two SZAs, 30° and 60°. Differences in all radiation quantities were calculated

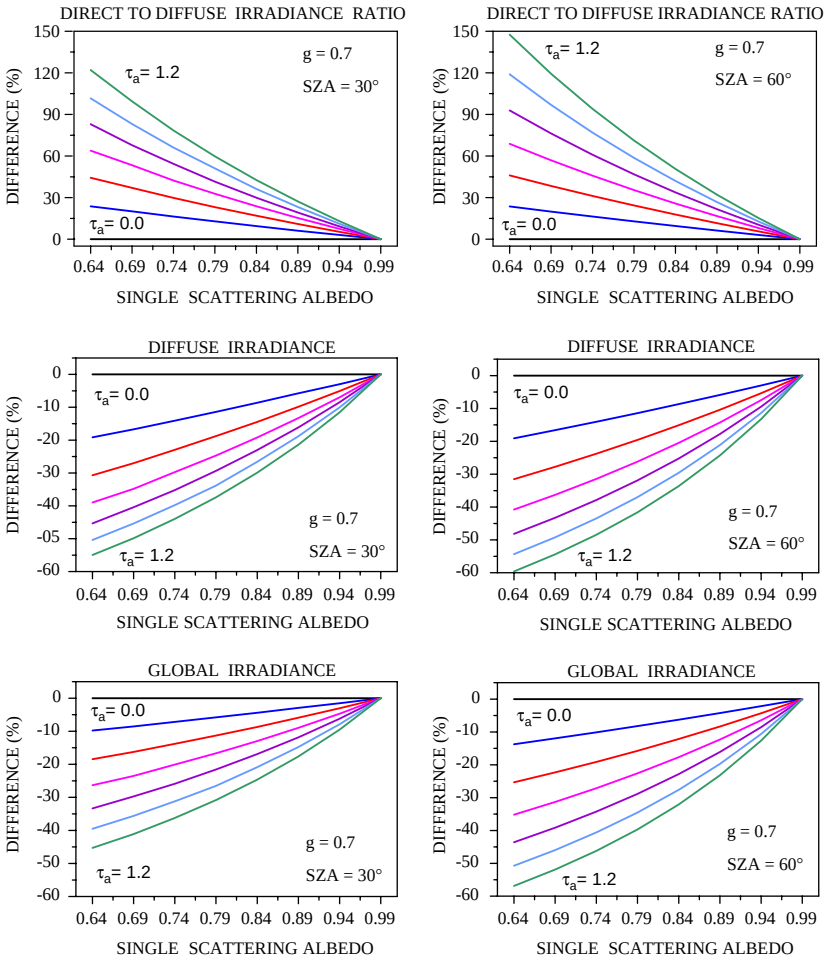


Fig. 1. Changes in direct-to-diffuse irradiance (top), diffuse irradiance (middle) and global irradiance (bottom) at 340 nm due to changes in $\omega$ from 0.99, for a range of aerosol optical depth from 0 to 1.2 in increments of 0.2, for each $\tau_a$. Differences in radiation quantities ($F$) are expressed in percent ($100[F_\omega - F_{\omega=0.99}]/F_\omega$). The left column refers to 30° and the right to 60° SZA.

from their values corresponding to $\omega = 0.99$, for each $\tau_a$ and SZA. In the model calculations we used an asymmetry parameter of 0.7, for the reasons discussed in the previous section. The wavelength of 340 nm was chosen because of the weak ozone absorption, making this sensitivity study practically independent on ozone. A similar analysis that was done for 300 nm (not shown here) reveals similar patterns for all three quantities.

In principle, these model calculations can be used for retrieving $\omega$ when one of the radiation quantities at a specific SZA and wavelength, and the aerosol optical depth are known. The accuracy of the retrieved $\omega$ depends on the sensitivity of the radiation quantities to changes in $\omega$, and is limited by the measurements' accuracy and the detection limit of the instruments at low-intensity conditions (large SZAs, small wavelengths, high-aerosol optical depth).

Fig. 1 suggests that the diffuse irradiance and the direct-to-diffuse irradiance ratio are the most sensitive to changes in $\omega$, and this occurs for a significant range of $\tau_a$. Apparently, in rather clean atmospheres (e.g. $\tau_a < 0.2$) the variation of all three quantities with $\omega$ is very small. On the other hand, in cases with high-aerosol concentrations (e.g. $\tau_a > 0.4$) also the global irradiance becomes sensitive to changes in $\omega$.

With regard to the effect of the SZA, it seems that again the diffuse irradiance and the direct-to-diffuse irradiance ratio maintain their sensitivity for the longest range of aerosol optical depth variation. As the contribution of the direct component weakens at larger SZAs, the behaviour of global irradiance becomes comparable to diffuse. Considering the analogous application to actual measurements, the uncertainty in determining $\omega$ at 340 nm depends on SZA, either because all three radiation quantities are less sensitive to changes in $\omega$ at small SZAs, or because spectral measurements become more uncertain at large SZAs.

## 5. Methodology to derive effective $\omega$ from Brewer measurements

The model-derived relationships of $\omega$ with irradiances, which were discussed in the previous section, were used to estimate the effective $\omega$ during cloud-free days with different aerosol conditions at Thessaloniki. Measurements of global and diffuse irradiance and the direct-to-diffuse irradiance ratios were compared with model calculations, which were based on the actually measured total ozone column and $\tau_a$. From these comparisons, we determined the values of $\omega$ for which the model and the measurements agreed to better than 1%. Depending on the sensitivity of each radiation quantity to changes in $\omega$, more than one values of $\omega$ could satisfy the above condition, because we considered meaningless to estimate the effective $\omega$ with precision higher of 0.01. The number of $\omega$ values for each case gives an idea of the achievable accuracy and the differences in estimating $\omega$.

Uncertainties in both the radiation measurements and the model input parameters contribute to the overall uncertainty of the methodology. Realistic uncertainties are the order of $\pm 2$–3% for the direct-to-diffuse irradiance ratio, $\pm 3$–5% for the global irradiance and $\pm 4$–6% for the diffuse irradiance. Only at a few stations or in cases where the measurements are fully controlled (e.g. during experimental campaigns) the lower limits of these uncertainties can be achieved (Bernhard and Seckmeyer, 1999; Bais et al., 2001). The uncertainty in direct-to-diffuse irradiance ratio is expected to be lower compared to the uncertainty in the other radiation quantities, since errors related to the calibration procedures are diminished. With regard to the determination of the optical depth from direct irradiance measurements, the uncertainty is estimated to less than 0.05 units (Kylling et al., 1998). Uncertainties in the model-derived radiation quantities at 340 nm are introduced by the assumed values for the asymmetry parameter, surface albedo, aerosol vertical profile and the extraterrestrial solar spectrum. According to d' Almeida et al. (1991), for the proposed aerosol model and for relative humidity up to 99% the asymmetry parameter at 350 nm varies between 0.66 and 0.77. Model sensitivity analysis revealed that the uncertainty in global irradiance calculations due to the asymmetry parameter value of 0.7 that was used in this study would be $\pm 1$%, resulting to an error of 0.02 in the calculated $\omega$ (Balis et al., 2003). Errors of similar magnitude are expected from the assumptions made for the aerosol vertical profile and surface albedo at Thessaloniki. Finally, the uncertainty in the extraterrestrial solar flux is within $\pm 3$% (Bais, 1997). The overall uncertainty in model-derived radiation quantities is estimated to $\pm 4$% for direct-to-diffuse irradiance ratio, $\pm 5.3$% for diffuse irradiance and $\pm 4.6$% for global irradiance.

The expected accuracy levels in estimating $\omega$ with the proposed method are summarized in Table 1 for two values of $\tau_a$ and two SZAs, and are comparable with those reported in other studies (Dubovik et al., 2000; Petters et al., 2003). Because of its lower uncertainty, the direct-to-diffuse irradiance ratio can be used for the estimation of the effective $\omega$ only when $\tau_a$ is higher than 0.2. On the other hand, diffuse and global irradiance measurements are useable only when $\tau_a > 0.4$. Even though the uncertainty in measuring the diffuse irradiance is higher compared to global irradiance, more accurate results can be achieved from the use of the diffuse component, because it is more sensitive to $\omega$. At large SZAs and high-aerosol optical depth (when the diffuse part is dominant) global irradiance can be also used to estimate the effective $\omega$. Using the methodology described above, significant changes in the aerosol

Table 1
Uncertainties in estimating the effective $\omega$ from measurements of global irradiance, diffuse irradiance, and the ratio of direct-to-diffuse irradiance for two aerosol optical depths and two solar zenith angles

| $\tau_a$ | Direct/Diffuse ratio | | Diffuse irradiance | | Global irradiance | |
|---|---|---|---|---|---|---|
| | 0.2 | 0.4 | 0.2 | 0.4 | 0.2 | 0.4 |
| 30° SZA | ±0.06 | ±0.04 | ±0.07 | ±0.05 | ±0.15 | ±0.08 |
| 60° SZA | ±0.06 | ±0.04 | ±0.07 | ±0.05 | ±0.13 | ±0.05 |

properties over a UV measuring site may be detected and typical values of $\omega$ may be derived, giving hints for the actual aerosol class, as far as $\tau_a$ is higher than 0.2.

Two examples of applying the proposed methodology are shown in Fig. 2 for 2 days in 2001 with different aerosol loading. The first day, 28 March 2001, was dryer with lower aerosol (relative humidity at local noon was ~28% compared to ~50% in the second day, 4 April 2001). Both days were cloud-free, according to observations made for every half an hour at the nearby meteorological station at the airport of Thessaloniki (~10 km far). As it would be expected from the results of Fig. 1, the low-aerosol content during 28 March 2001 reduces significantly the accuracy of the retrieved $\omega$. The effective $\omega$ during both days is estimated independently by comparing each of the three radiation quantities with the corresponding model calculations. If the different types of radiation measurements (in this case global and direct irradiances) were absolutely consistent, one would expect completely identical values of $\omega$ resulting from each type of measurement. In practice though, due to (a) the uncertainties introduced by the independent calibration and measurement procedures of the global and direct irradiance, (b) possible contamination of the measurements by the presence of thin clouds and (c) imperfect synchronization of global and direct measurements, some inconsistencies may occur between the two quantities, resulting into different estimates of the effective $\omega$. In both days, $\omega$ derived from global and diffuse irradiance measurements is overestimated with respect to that derived from the direct-to-diffuse irradiance ratio, probably because in the later the effect of absolute calibration errors are much smaller. In most cases, especially at small SZAs and high-aerosol optical depth, the range of $\omega$ is within the uncertainties proposed in Table 1.

Despite the uncertainties, it appears from the two examples of Fig. 2 that during the day with high $\tau_a$ (4 April) and for SZAs smaller than 75° the effective $\omega$ varies between 0.8 and 0.9, whereas in the second-half of 28 March (when $\tau_a \sim 0.2$) it ranges between 0.65 and 0.8, suggesting the presence of more absorbing aerosols. Since no direct method for measuring $\omega$ at Thessaloniki was available, the resulted effective $\omega$ could be assessed only by supplementary information about the composition of aerosols in the troposphere during these days.

To investigate the origin of air masses transported over Thessaloniki, analytical back trajectories at different pressure levels were used, provided by the German Weather Service (DWD). Model calculations were derived for 13.00 UTC of each day on a three-dimensional grid of 1.5° resolution (Kottmeier and Fay, 1988). The deviation between the calculated and the actual track of an air parcel is about 10% of the trajectory length (Stohl, 1998). In Fig. 3 (upper panel) the trajectories reveal that on 4 April air originated from upper levels and from north-eastern directions; 4 days before being over Russia. This suggests that the air masses transported over Thessaloniki were rather free of urban—and strongly absorbing—aerosols, which is in accordance with the estimated higher values of the effective $\omega$. On the other hand, the trajectories for 28 March (lower panel) reveal that air masses during that day were coming from western directions, passing over polluted or urbanized areas, such as the lignite mines and power plants in north-western Greece and over south Italy. Aerosols emitted from such areas contain soot and other absorbing substances, resulting to lower values of effective $\omega$. Moreover, the smaller $\omega$ values on 28 March could be also associated with mineral components in aerosol transported over North Africa at the 850 hPa level. The higher optical depth of the aerosols on 4 April 2001 is likely due to the prevailing local meteorological situation, with lighter winds and higher humidity levels in comparison to 28 March 2001. On 28 March, the smaller $\omega$ values seem to be originally in contrast to the mostly maritime origin of aerosols in this case. Thus, larger $\omega$ values than on 4 April should be expected, when aerosols have rather continental origin. However, the finally smaller $\omega$ values on 28 March than on 4 April result probably from mixing processes that the aerosols undergo when being transported over continental areas. This highlights the importance of such mixing processes, apart from the source of aerosol particles, in determining their composition and optical properties.

Even though global irradiance is less sensitive to changes in single scattering albedo, compared to the

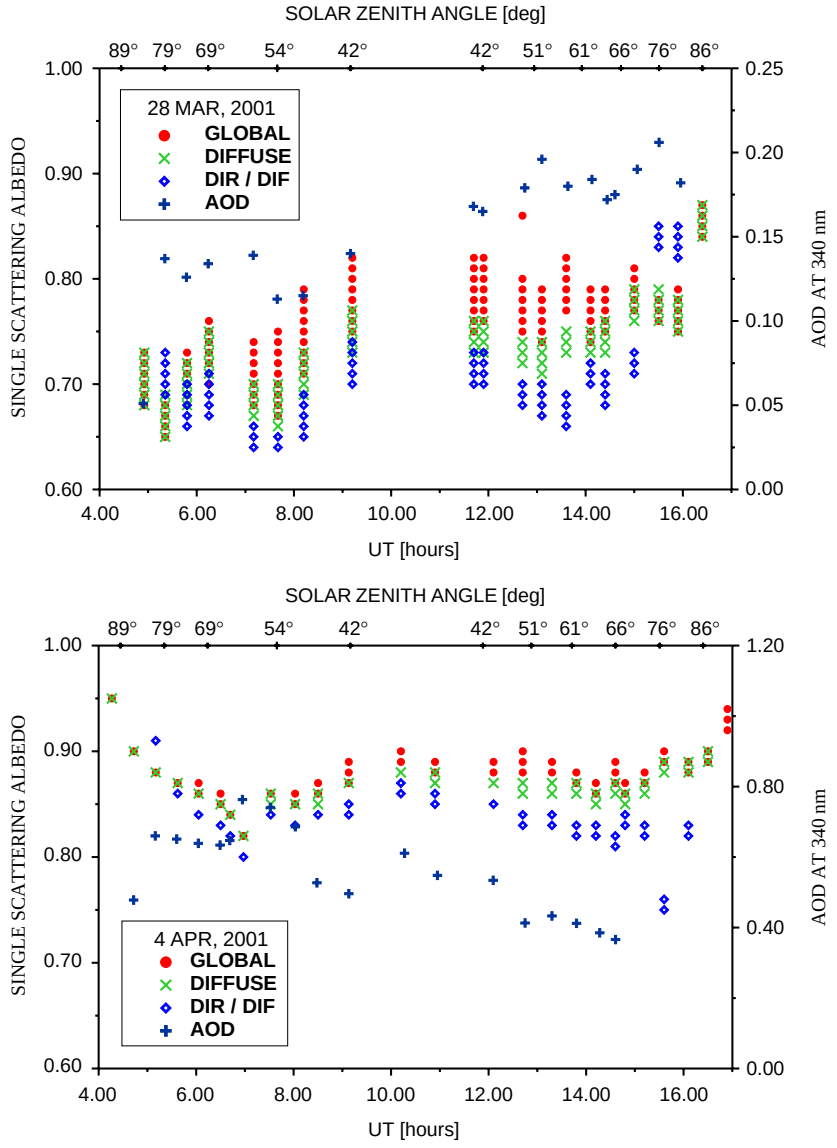


Fig. 2. Examples of deriving the effective $\omega$ by comparing measurements of global irradiance, diffuse irradiance and direct-to-diffuse irradiance ratio with model calculations, during 2 days with high (4 April 2001) and low (28 March 2001) aerosol content. The variation of the aerosol optical depth (AOD) during the day is also shown (crosses—right axis).

other two radiation quantities discussed above, this is the most commonly measured parameter at UV-monitoring sites. Therefore, a fairly long record of global irradiance measurements combined with aerosol optical depth measurements could be used to provide, at least qualitatively, information for the variability of the most likely effective $\omega$ at the specific location. Fig. 4 presents the variation of global irradiance at 340 nm in Thessaloniki under cloud-free conditions during the period 1998–2002 as a function of $\tau_a$ for two SZAs ($50^\circ \pm 1^\circ$ and $63^\circ \pm 1^\circ$). The later SZA was chosen for increasing the amount of measurements during the period of study and consequently the statistical significance of the results (measurements at $63^\circ$ degrees are available at Thessaloniki all year around). Model calculations were used to derive look-up tables of global irradiance at 340 nm as a function of $\tau_a$ and $\omega$. Since the influence of ozone on UV irradiance at 340 nm is negligible, the variability of global irradiance at a given $\tau_a$ is practically due to the variability of the aerosol optical properties, and to the measurements' uncertainties. Finally, because the calculated irradiances

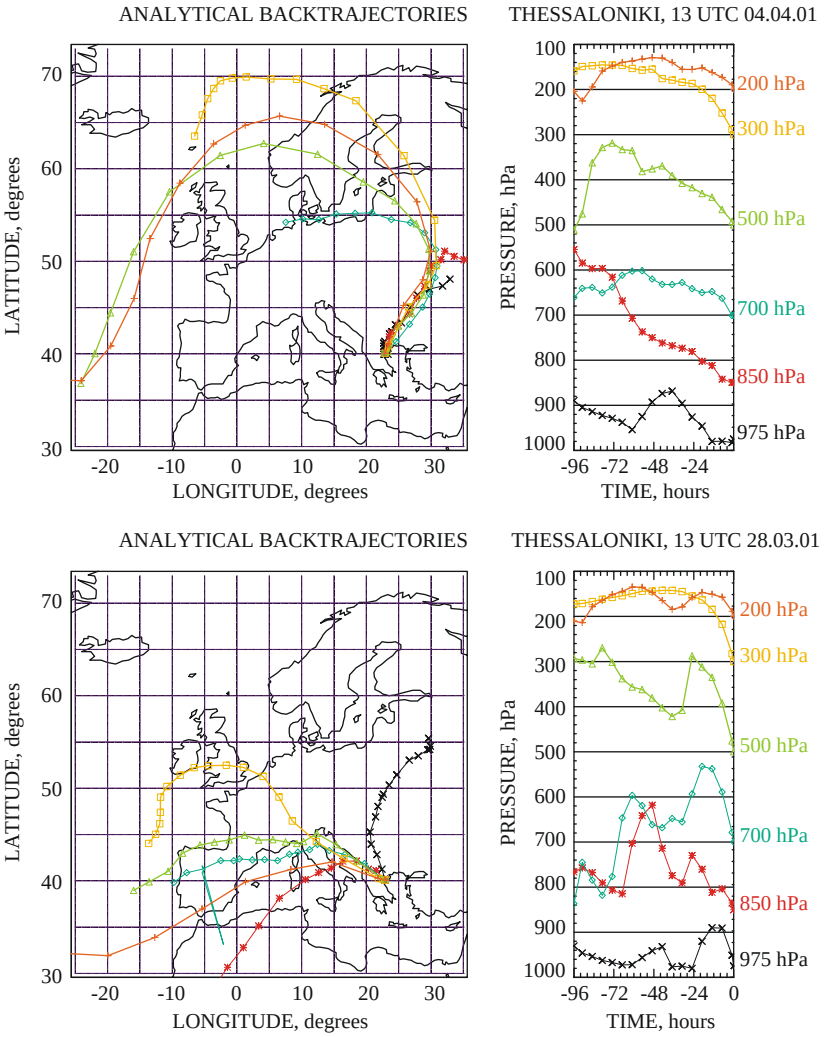


Fig. 3. Analytical back trajectories of air masses at different pressure levels for 4 April 2001 (upper panel) and 28 March 2001 (lower panel).

correspond to the mean sun–earth distance, the measurements were adjusted accordingly, following the formula of Spencer (1971). The comparison of these measurements with model calculations, performed for two extreme values of $\omega$ (0.64 and 0.99), reveals that the majority of them lay within the expected range, with a slight tendency to agree better with model estimates that correspond to higher values of $\omega$. This tendency is more apparent at large optical depths ($\tau_a > 0.8$), suggesting the presence of less absorbing aerosols in the ultraviolet. Because the range of the derived $\omega$ values is large the result of this comparison is rather qualitative than quantitative. Additional analysis with the use of more ancillary data is needed to investigate the behaviour of the derived effective $\omega$ at Thessaloniki in different seasons or weather patterns.

## 6. Conclusions

An indirect method for determining the effective single scattering albedo in the ultraviolet by comparing irradiance measurements derived from a Brewer spectroradiometer with model calculations was proposed. Three variants of this method, based on global irradiance, diffuse irradiance, and the ratio of direct-to-diffuse irradiance were investigated and inter-compared. It appears that the latter parameter has the highest sensitivity to $\omega$ variations and thus the lowest uncertainty. Consequently, the direct-to-diffuse irradiance ratio is expected to give the highest accuracy in the estimation of the effective $\omega$. A prerequisite for applying these techniques is the availability of the aerosol optical depth at the wavelength(s) of interest.

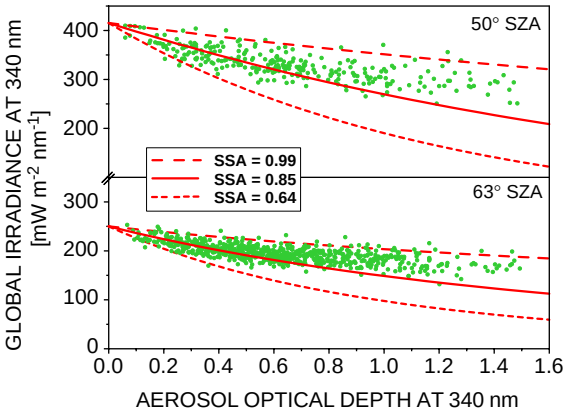


Fig. 4. Measurements of global irradiance at 340 nm (solid circles) during cloud-free days for years 1998–2002 at Thessaloniki as a function of $\tau_a$ at 340 nm. The upper panel shows measurements taken at $50^\circ \pm 1^\circ$ SZA and the lower panel those at $63^\circ \pm 1^\circ$ SZA. The two dashed and one solid curves correspond to model calculated global irradiance for three different values of $\omega$ (0.64, 0.85 and 0.99).

It appears that all three approaches give less uncertain results in situations with high-aerosol loads. The individual uncertainties of the measurements, the modelling and the aerosol optical depth determination contribute to the overall uncertainty in estimating the single scattering albedo. A realistic estimate of the achievable accuracy when using the direct-to-diffuse irradiance ratio at 340 nm is about $\pm 0.04$ units of $\omega$ for $\tau_a = 0.4$ and about $\pm 0.06$ units for $\tau_a = 0.2$, imposing some limitations to the applicability of the method when $\tau_a < 0.2$. The lower accuracy in estimating $\omega$ from diffuse and global irradiance measurements limits their applicability only to cases with $\tau_a > 0.4$.

The application of the proposed methodology to two days with aerosols of different origin and amount provides estimates of the effective $\omega$ that are in accordance with the expected—from back trajectory analysis—composition of the aerosols during the days in question. Comparisons of global irradiance measurements at 340 nm with model calculations for different aerosol optical depths for years 1998–2002 reveal that the effective $\omega$ at Thessaloniki covers the entire range of $\omega$ variation for $\tau_a < 0.8$. For higher values of $\tau_a$ the effective $\omega$ ranges between 0.85 and 0.99.

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