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Aerosol forcing efficiency in the UVA region from spectral solar irradiance measurements at an urban environment

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Abstract. Spectral Ultraviolet (UV) measurements using a Brewer MKIII double spectroradiometer were used for the determination of the aerosol forcing efficiency (RFE) under cloud free conditions at Thessaloniki, Greece for the period 1998–2006. Using measured spectral UVA irradiance in combination with synchronous aerosol optical depth (AOD) measurements at 340 nm, we calculated the seasonal and the percent RFE changes with the help of radiative transfer model calculations used for cloud and aerosol free conditions reference. The calculated RFE for the 325–340 nm wavelength integral was found to be $-0.71\pm0.30\,\mathrm{W\,m^{-2}}/\tau s_{340\mathrm{nm}}$ and corresponds to a mean calculated RFE% value of $-15.2\%\pm3.8\%$ ($2\,\sigma$) per unit of $\tau s_{340\mathrm{nm}}$, for the whole period. This indicates a mean reduction of 15.2% of the 325–340 nm irradiance for a unit of aerosol optical depth slant column increase. Lower RFE% was found during summertime, which is a possible indication of lower absorbing aerosols. Mean AOD slant at 340 nm for the city of Thessaloniki were processed in combination with RFE% and a mean monthly UVA attenuation of ∼10% for the whole period was revealed. The nine years' analysis results showed a reduction in RFE%, which provides a possible indication of the changes in the optical properties over the city area. If such changes are only due to changes in the aerosol absorbing properties, the above finding suggests a 2% per decade increase in UVA due to changes in the aerosol absorption properties, in addition to the calculated increase by 4.2%, which is attributed only to AOD decrease at Thessaloniki area over the 1998–2006 period.

Keywords. Atmospheric composition and structure (Aerosols and particles) – Meteorology and atmospheric dynamics (Radiative processes)

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1 Introduction

Aerosols modify the Earth-atmosphere energy budget through various atmospheric processes such as the direct effect by scattering and absorption of the solar and earth's radiation (e.g. Charlson et al., 1992), the semi-direct effect by changing atmospheric thermodynamics and cloud formation (e.g. Koren et al., 2004), and the indirect effect by changing cloud microphysics (Rosenfeld and Lensky, 1998). The impact of aerosols to the Earth's radiation budget is a major component of global and regional climatic patterns, influencing processes in the troposphere (Yu et al., 2002), the hydrological cycle (Ramanathan et al., 2001), and surface temperatures over the globe (Charlson et al., 1992). The cooling by anthropogenic aerosols may be comparable in magnitude to greenhouse gas warming on a global scale but can be much larger on a regional scale (Intergovernmental Panel on Climate Change (IPCC), 2001). However, uncertainties remain substantial for the direct and for the indirect effect (IPCC, 2007).

In the past decade several papers suggested a reduction of solar irradiance, reaching the surface of the earth under cloudless conditions caused by anthropogenic aerosols (Liepert, 2002). Further studies have shown that over Europe and N. America the negative tendencies in solar radiation have reversed in the past decade as a result of improving air quality (Wild et al., 2005). AOD records from six remote locations in Europe confirmed the recent clearing of the air, particularly in the lower troposphere, showing a 60% decline of AOD and hence anthropogenic aerosol concentration since 1986 (Ruckstuhl et al., 2008; Philipona et al., 2009). They also confirmed increasing atmospheric transmission by surface solar irradiance measurements at a large number of stations in Northern Germany and Switzerland since 1981, showing significant global radiation increases. Most of these results were derived using the visible component of solar irradiance. Also in the UVA range, where the radiative transfer

is much more sensitive to aerosols, the change in solar radiation correlates well with increasing anthropogenic activities (Zerefos et al., 2000; Balis et al., 2003; Gerasopoulos et al., 2003).

The radiative effect of aerosols in the atmosphere observed is currently examined using radiative transfer calculations with AOD as the sole changing parameter for the characterization of any possible relationship between the radiation trend and the aerosol variability. Changes in the absorbing properties of aerosols in a global scale would add significant uncertainty in any future trend analysis of solar radiation. This is because the direct radiative effect of aerosols is also very sensitive to the single scattering albedo (SSA), which is a measure of the probability of a photon being scattered into some direction rather than absorbed when interacts with an aerosol particle. For example, a change in SSA from 0.9 to 0.8 can often change the sign of the direct effect, depending on the albedo of the underlying surface and the altitude of the aerosols. (IPCC, 2007) In this work, we investigate the possibility to quantify how UVA radiation trends can be linked with the aerosol optical depth variability. Additionally, we examine the possible effect on UVA radiation from possible changes of the SSA. We quantify the use of aerosol optical depth trends and their impact on solar irradiance levels taking into account also annual AOD patterns, using ground based irradiance and AOD measurements performed in Thessaloniki, Greece during a nine year period (1998–2006).

2 Measurement data

The UV monitoring station at Thessaloniki has one of the longest records of spectral UV irradiance measurements, from a single and a double Brewer spectroradiometers. In this study we are concerned for spectral measurements of solar ultraviolet (global) irradiance on a horizontal surface. The measurements were performed by a Brewer (MKIII #086) spectroradiometer, which operates on a regular basis at the Laboratory of Atmospheric Physics, University of Thessaloniki, Greece, since 1993. The instrument is positioned on the roof of the Physics Department building (40.63° N, 22.96° E, at 60 m a.s.l.), in the city center of Thessaloniki. The instrument has participated in a large number of experimental (Thiel et al., 2008) and intercomparison campaigns (e.g. Bais et al., 2001; Gröbner et al., 2006). 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 ±4%. More details on the instrument characteristics, the calibration history and the quality control of the measurements can be found in Garane et al. (2006). For the selection of the cloud free measurements we used the methodology described in Vasaras et al. (2001). The method has been used in a number of publications including cloudless Brewer measurement analysis and is based on the variability of the measurements from a collocated pyranometer.

Measurements of the aerosol optical depth (AOD) have been conducted in Thessaloniki, since 1997, a city with a population of approximately 1.2 million. The site is facing the Aegean Sea to the south and is situated along the expected pathway through which pollution from central and Eastern Europe influences aerosol loading over the Eastern Mediterranean (Amiridis et al., 2005). The retrieval of AOD is based on the absolute calibration of the solar spectral irradiance measured by the Brewer (Bais, 1997; Kazadzis 2005). The uncertainty analysis of the AOD retrieval using the MKIII Brewer showed an error on the AOD which is estimated to within 0.07 in the UV-B and 0.05 in the UV-A for measurements at solar zenith angles between 15 and 75 degrees (Kazadzis et al., 2005). This error is the result of the propagation of errors due to the direct irradiance measurement and calibration uncertainties, the determination of the extraterrestrial spectrum, and the measurement of ozone and SO_2. Local emissions and regional pollution transport contribute to the poor air quality of the city which is reflected mostly in particulate matter (PM) concentrations. The 24 h limit values for PM_{10} ($50\,\mu gr/m^3$) are exceeded for more than half of the days during a year in the city centre's environmental monitoring sites. The amount of aerosols in this location is quite high, with a annual mean AOT at 340 nm equal to 0.45 and monthly values ranging between 0.3 and 0.7, for winter and summer months, respectively (Kazadzis et al., 2007).

The cloud-free UV irradiance values were calculated with the UVSPEC model (Mayer and Kylling, 2005). The radiative transfer model (RTM) equation was solved with the discrete ordinates algorithm (Stamnes et al., 1988), using six streams and pseudospherical correction. The Air Force Geophysical Laboratory US standard atmosphere profiles (Anderson et al., 1986), were used for ozone, temperature and air pressure. According to Kazantzidis et al. (2005), possible uncertainties for the UVA spectral region and for solar zenith angles lower than 60 degrees are less than 1%. The high resolution ATLAS-3 solar spectrum (ftp://susim.nrl.navy.mil/pub/atlas3) was used. The type of landscape suggests a relatively small surface albedo, thus the constant value of 0.03 was used for the whole UV region. The Elterman's (1968) vertical profile, scaled to match the measured optical depth at 340 nm, was used to describe the aerosol vertical distribution. Mie calculations were performed 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. A comprehensive analysis about the introduced uncertainties on model calculations, due to the above-mentioned assumptions, could be found in Bais et al. (2005).

3 Methodology

The aerosol radiative forcing efficiency (RFE) is defined as the rate at which the irradiance at a certain wavelength range is "forced" (changing) per unit of aerosol optical depth (τ_{AOD}) (Bush and Valero, 2003). In this study, the irradiance change DI was calculated as:

$$DI_i = I_i - MI_i \quad (1)$$

where I_i and MI_i are the integral of 325–340 nm wavelength range calculated from the Brewer measurements for cloud free conditions and from the RTM for the same solar zenith angle and aerosol free conditions, respectively.

Most of the studies relating solar irradiance changes with aerosol changes, present results of solar irradiance in the visible wavelength range. In this work we chose to analyze UV-A irradiances, which are much more sensitive to aerosols, a major factor affecting the down-welling solar radiation, which correlates well with increasing anthropogenic activities. More specific we have chosen the 325–340 nm integral, as we would like to avoid analyzing UV-B wavelengths that are affected by ozone changes and could introduce additional uncertainties to the results. Also, measurements higher than 350 nm that are performed on a different slit with the Brewer spectroradiometer. Finally, we wanted to avoid analyzing single wavelength irradiance analyses.

AOD was calculated from direct sun spectral measurements averaged within ±15 min from each of the UV global irradiance measurement. For the determination of RFEs, the AOD in the slant path (AODs or τs) was calculated, when normalizing the retrieved AOD with the air mass factor for each measurement (Garcia et al., 2006). In addition, the percent RFE (RFE%) was calculated. RFE% is defined as the percent change of the irradiance at the given wavelength range per unit of AOD.

$$\text{RFE\%} = 100 \cdot \frac{DI}{MI} \quad (2)$$

The methodology described in Garcia et al. (2006) was used for data analysis. The monthly aerosol forcing efficiency under cloudless and snow-free surface conditions was calculated through the slope method, limiting the analysis to solar zenith angles (sza) less than 60 degrees (Jayaraman et al., 1998). Summarizing this approach, the regression fits were computed by dividing the range of the instantaneous AOD in bins of 0.1, previously normalized with the cosine of sza (AODs), in order to consider the effect of the optical path. Also, a minimum number of cases per AOD bin accepted for the calculations were set to be 2.5% of the total UVA spectral measurements. These fit conditions provide the same weight for every bin in the regression model avoiding the greater weight for the smallest AOD slant, whereas the points used have statistical significance.

RFE and RFE% values were calculated using monthly and seasonal measurement data sets, where seasonal is defined as three month period data (e.g. Fig. 1, summer RFE is calculated from June, July and August data for every year). The RFE% were calculated using the Garcia et al. (2006) bin analysis. Summarizing, the seasonal RFE% under cloudless conditions was calculated through the slope method limiting the analysis to solar zenith angles less than 60 degrees. This regression fit was computed by seasons dividing the range of the instantaneous aerosol optical depth in bins of 0.1, previously normalized with the cosine of sza (AOD in slant) to consider the effect of the optical path. Also, a minimum number of cases per bin was established, 2.5% of the total data set for station. The fact that cloud free measurements of UV irradiance during wintertime (December–February) are limited, thus the minimum number of cases per bin was not accomplished, in addition to the criterion of using solar zenith angles lower than 60 degrees, lead to the exclusion of these periods from the analysis. The coefficients used for monthly analysis were lower than the ones for the seasonal periods (70% of the latest were higher than 0.75). The number of measurements varied from 650–1050 for the summer, 320–450 for spring and 250–410 for autumn depending on the year. These numbers are directly related with the availability of cloud free days per season and also gaps in the measurement periods linked with the participation of the instrument in campaigns away from the monitoring site. An example of the RFE% calculation for summer of 1998 is shown in figure where RFE% versus AODs is presented. The RFE% calculated from all data are $-11.44\%/\tau s_{340nm}$ and $-11.40\%/\tau s_{340nm}$, respectively. The correlation coefficient of the bin analysis is equal with -0.99.

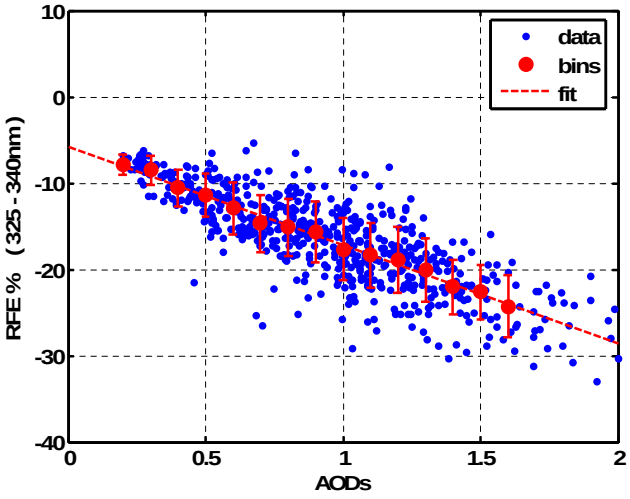

Fig. 1. Percent change (%) of the irradiance at the 325–340 nm wavelength integral versus the AOD slant. Data represent measurements of June–August 1998.

The uncertainties related with the RFE and RFE% are linked with the ones mentioned for spectral UV measurements and AOD determination reported in Sect. 2.

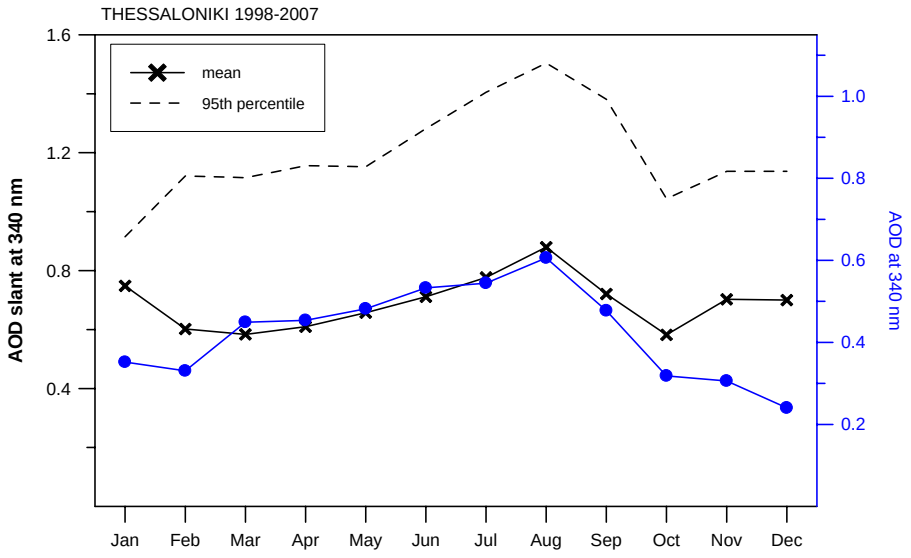

Fig. 2. Mean AOD (blue) and AODs (black line and symbols) values at 340 nm measured from 1998–2006 at Thessaloniki. The 95th percentile values of the mean AODs are also shown.

4 Results

Monthly average RFEs at different wavelengths, derived from multi-year analysis (1998–2006), were calculated at Thessaloniki. The calculated values of RFE for the 325–340 nm integrals were found to be $-0.71\pm0.30\,\mathrm{W\,m^{-2}}/\tau_{340\mathrm{nm}}$. Looking at seasonal differences, the RFE values for spring, summer and autumn, were -0.81, -0.65, $-0.76\,\mathrm{W\,m^{-2}}/\tau_{340\mathrm{nm}}$, respectively. These numbers suggest that aerosols with lower absorption properties are present for the summer months over the site. Similar results were found in Garcia et al. (2006) and Groebner and Meleti (2004) at Ispra, Italy. In addition, for Thessaloniki area, lower aerosol absorption during summertime was found in Arola et al. (2005) and Bais et al. (2005).

The seasonal variation of AOD over Thessaloniki shows an AOD maximum for summer months (0.62 in August), and minimum in wintertime (0.3 in December) (Kazadzis et al., 2007). As a first approach this could lead to higher UVA attenuation during summer. However, the solar zenith angle effects and the aerosol absorption properties have to be also considered.

Taking into account the differences of average solar zenith angle during different months of the year, we calculated the AODs annual variation for the 1998–2006 period, for Thessaloniki. The seasonal variation of AODs is shown in Fig. 2 together with the actual monthly mean AOD measurements. Mean values of AODs at 340 nm over Thessaloniki for the given period were found to be 0.75, while the 95th percentile statistics gave maximum values from 0.9 to 1.5. The annual variability of the AOD is smaller when calculating the AODs, because the wintertime slant path is larger than the one during summertime. However, maximum values of AODs were still observed in the summer months and especially in August (0.9). Precipitation is a parameter that also determines the seasonal variation of AOD values in the area and generally in the Mediterranean basin. Thus, the high summer AOD values are related to the low precipitation over the Mediterranean owing to the deflection of tracks of depressions by the northward shifted Azores anticyclone, whereas the lower winter AOD values are determined by high precipitation associated with deep depressions traveling across the basin (Papadimas et al., 2008). In that study, the satellite derived AOD measurements at the Mediterranean area showed up to 45% higher AOD values in the summer than in winter. Those values demonstrate the role of precipitation and atmospheric circulation, resulting in smaller AOD values during the rainy seasons of autumn and winter and in higher AOD values due to accumulation of (fine) aerosol particles in summer.

The AOD measurements at 340 nm, performed by the spectroradiometer during the period 1997–2006, revealed a negative trend of about -3.2% per year when analyzing AOD and -3.9% for AOD slant column values (Fig. 3). Similar negative trends were found analyzing the particulate matter (PM_{10}) measurements held in the city (Kazadzis et al., 2007). These results are most probably linked with a series of norms that had been taken during the 1990s concerning many aspects of the city activities (Petrakakis et al., 2005), like the consumption of crude oil with low sulphur content for industrial activity, the improvement of fuel quality, the renewal of the fleet with vehicles using anti-pollution technology and the annual check in the exhausts control etc. To date, several other studies (Koukouli et al., 2006; Kazadzis et al., 2007; Mishchenko et al., 2007; Papadimas et al., 2008) using also

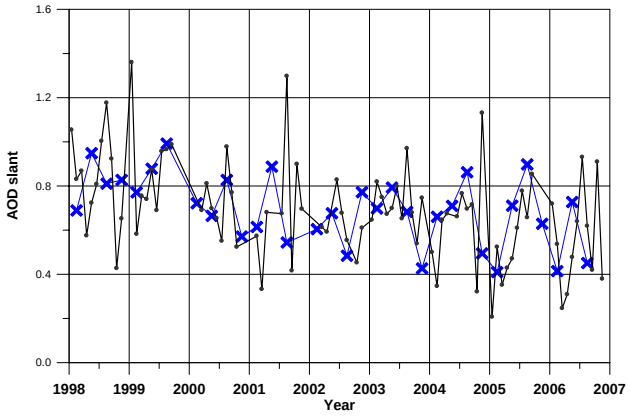

Fig. 3. Seasonal (blue symbols) and monthly (black line) AOD slant changes for Thessaloniki, Greece.

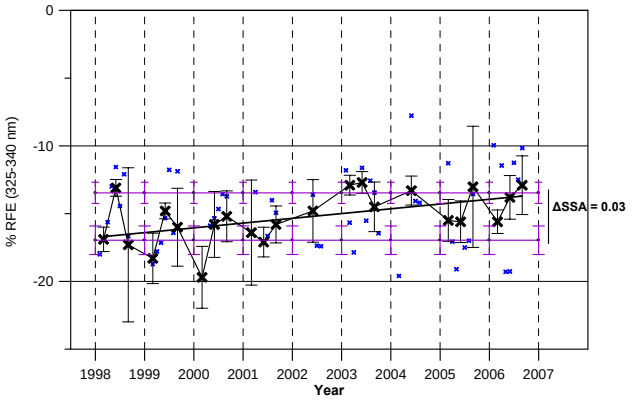

Fig. 4. Seasonal (black dots) and monthly (blue dots) RFE% changes calculated for Thessaloniki area over the period 1998–2007. Magenta points represent SSA calculations and their uncertainty estimation.

satellite datasets (e.g. Moderate Resolution Imaging Spectroradiometer, MODIS, Total Ozone Mapping Spectrometer, TOMS, or Advanced Very High Resolution Radiometer, AVHRR) reported a decreasing tendency in AOD values for the Mediterranean area during the last years.

As RFE% is independent of any AOD changes during the period, the calculated standard deviation, could provide an estimate of the aerosol forcing variability not due to AOD changes but to the possible change of aerosol absorption properties. The RFE% values for spring, summer and autumn were equal to −16.5%, −14.3% and −15.2% per unit of τs_{340nm}, respectively. Using RFE%, the real aerosol forcing can be calculated, when combining the above percentages and AODs results of Fig. 3. For spring, summer and autumn these calculations provide a 9.9%, 11.0% and 9.2% per unit of τs_{340nm}, UVA attenuation, respectively. In addition, the 95th percentiles (maximum AOD) included in the figure can provide the maximum aerosol impact on UVA irradiance reaching the surface (18.2%, 18.3%, 18.2% for the three seasons).

UVA spectral measurements performed by the Brewer spectroradiometer revealed a positive change over the 1998–2006 period (Meleti et al., 2009). In addition, the observed AOD decrease (Kazadzis et al., 2007; Meleti et al., 2009) can only partly explain such changes. Choosing the 325–340 nm wavelength region where ozone changes are negligible for UVA ones, we tried to investigate RFE% possible changes (which is independent of any AOD changes). Such RFE% changes can be linked with changes in the absorbing characteristics of aerosols. RFE% is a relatively stable property sensitive to the single-scatter aerosol properties (primarily single-scatter albedo) and environmental properties (primarily surface albedo and length-of-day) (Conant, 2000).

Analyzing the seasonal RFE% for the period 1998–2006, a positive change of RFE% of 3.8%/τs_{340nm} was calculated. As RFE% is a parameter that is independent of AOD changes such an increase can be a result of changes in the optical properties of aerosols or other factors like changes in the radiation in the top of the atmosphere. Concerning the changes in the optical properties, an asymmetry parameter (AP) change of 0.05 can lead to a 1.5% change per τs_{340nm} to the calculated RFE%. Concerning SSA, a decrease in the absorbing properties of aerosols (SSA increase) could also lead to such an RFE% change. In the following section we tried to calculate this SSA change in the case that it is the only changing RFE%-affecting parameter.

As observed change in RFE% can be considered as a possible hint of a change of the aerosol absorbing properties over the area and has to be taken into account in addition (compensating) to the observed AOD negative trends. The retrieved RFE%s were used in order to calculate possible SSA changes. Radiative transfer model calculations have been used in order to calculate the SSA value that can be used in the model input in order to match the calculated RFE% values. Magenda lines in Fig. 4 represent the calculated SSA for RFE%=$-13.2/\tau s_{340nm}$ and $-17/\tau s_{340nm}$ that represent the first and last points of the linear regression of RFE%s during the 1998–2006 period and the error bars the model SSA uncertainty introduced by the use of solar zenith angles varying from 20 to 60 degrees. The analysis provided here suggests that the observed RFE% change could be explained by a possible change of 0.03 in the mean SSA over the area during this specific 9 year period.

Using the AOD slant column results combined with RTM calculations to estimate the UVA (325–340 nm) increase due to aerosol optical properties changes, described in Figs. 3 and 4. We found a 4.2% UVA increase per decade when taking into account a mean RFE% (no change in the aerosol absorbing properties but only the AOD change) and a 6.2% UVA increase per decade when taking into account both the AOD decrease and the possible increase of SSA.

5 Summary and conclusions

We used measurements of global spectral irradiance and AOD in the UVA region to estimate the aerosol radiative forcing efficiency (RFE) at an urban location in the Eastern Mediterranean during a nine years time period (1998–2006). RFE was found equal to $(-15.2\pm3.8\%)/\tau s_{340nm}$ (2σ), representing the average reduction of the UVA global irradiance reaching the ground by the presence of an aerosol layer with AOD slant column of 1. In addition, we calculated different RFE% for different seasons and a positive change of this parameter was found. As RFE% is independent of any AOD slant column changes we linked this RFE% variability with possible changes in the aerosol SSA over the area. When taking into account only AOD changes, a 4.2% per decade increase of UVA irradiance was calculated. Additionally, a 2% can not be explained by this AOD decrease and in case that SSA changes are the only aerosol optical parameter responsible for such an RFE% change this increase can be attributed to decreases in the aerosol absorption properties. As AOD decrease in the area has been attributed to improvements in fuel of vehicles, industry crude oil content and exhausts control, a change of the aerosol absorbing properties have to be considered as possible. Such a conclusion is difficult to corroborate without further studies on aerosol absorbing properties observations, like long term aerosol chemical composition measurements. However, SSA measurements in the UV are difficult and in current literature there are only experimental techniques that are used for such a purpose (Corr et al., 2009, summarizes all such methods). Significant uncertainties in global aerosol absorption properties (SSA) may constitute the largest single source of uncertainty in the current estimates of aerosol climate forcing. For example, in the city of Thessaloniki, changes in aerosol absorption properties distribution over time, caused, for example, by industrialization, urbanization, deforestation, or regional environmental controls, complicate the ability to predict aerosol climatic effects.

The quantity and sometimes the quality of observational data on SSA in the UV part of the solar spectrum currently, do not match that available for aerosol optical depth. However aerosol absorption information has to be included in future studies related with aerosol impacts on UV radiation. Current and future simulation of future UV radiation levels in a global scale are based only in ozone recovery scenarios and only few of them include AOD trends. Changes in the absorbing aerosol properties globally could have a large effect on the uncertainty budget of any of the above simulations.

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