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The direct effect of aerosols on solar radiation over the broader Mediterranean basin

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Abstract

For the first time, the direct radiative effect (DRE) of aerosols on solar radiation is computed over the entire Mediterranean basin, one of the most climatically sensitive world regions, by using a deterministic spectral radiation transfer model (RTM). The DRE effects on the outgoing shortwave radiation at the top of atmosphere (TOA), DRE_{TOA}, on the absorption of solar radiation in the atmospheric column, DRE_{atm}, and on the downward and absorbed surface solar radiation (SSR), DRE_{surf} and $DRE_{netsurf}$, respectively, are computed separately. The model uses input data for the period 2000–2007 for various surface and atmospheric parameters, taken from satellite (International Satellite Cloud Climatology Project, ISCCP-D2), Global Reanalysis projects (National Centers for Environmental Prediction – National Center for Atmospheric Research, NCEP/NCAR), and other global databases. The spectral aerosol optical properties (aerosol optical depth, AOD, asymmetry parameter, g_{aer} and single scattering albedo, ω_{aer}), are taken from the MODerate resolution Imaging Spectroradiometer (MODIS) of NASA (National Aeronautics and Space Administration) and they are Supplemented by the Global Aerosol Data Set (GADS). The model SSR fluxes have been successfully validated against measurements from 80 surface stations of the Global Energy Balance Archive (GEBA) covering the period 2000–2007.

A planetary cooling is found above the Mediterranean on an annual basis (regional mean $DRE_{TOA} = -2.4\,Wm^{-2}$). Though planetary cooling is found over most of the region, up to $-7\,Wm^{-2}$, large positive DRE_{TOA} values (up to $+25\,Wm^{-2}$) are found over North Africa, indicating a strong planetary warming, as well as over the Alps ($+0.5\,Wm^{-2}$). Aerosols are found to increase the absorption of solar radiation in the atmospheric column over the region ($DRE_{atm} = +11.1\,Wm^{-2}$) and to decrease SSR ($DRE_{surf} = -16.5\,Wm^{-2}$ and $DRE_{netsurf}$ $-13.5\,Wm^{-2}$) inducing thus significant atmospheric warming and surface radiative cooling. The calculated seasonal and monthly DREs are even larger, reaching $-25.4\,Wm^{-2}$ (for DRE_{surf}). Sensitivity tests show that

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regional DREs are most sensitive to ω_{aer} and secondarily to AOD, showing a quasi-linear dependence to these aerosol parameters.

1 Introduction

Atmospheric aerosols influence the Earth's climate by modifying its energy balance through the direct, indirect and semi-direct effects. However, the uncertainty of aerosol effects on the Earth's radiation budget greatly exceeds that of any other climate forcing agent (Kaufman et al., 2002; Vardavas and Taylor, 2007; Forster, 2007). This is due to the fact that the aerosol physical, chemical and optical properties are highly variable in space and time, because of the short atmospheric lifetime of aerosols and of the inhomogeneous emissions compared to those of the greenhouse gases (Quinn and Bates, 2005; Forster, 2007). Improved assessments of aerosol radiative effects are essential for reducing the uncertainty of future climate changes (IPCC, 2007) and have to be performed not only on global but also on regional scales. Such assessments are also important in the context of changes of solar radiation at the Earth's surface, commonly known as solar (or global) dimming and brightening, which are of primary importance for the Earth's climate and have received much attention lately (see Wild, 2009).

The role of aerosols is even more crucial over climatically sensitive regions like the Mediterranean basin, which is a crossroad where aerosols from different sources are mixed. These aerosols include fine anthropogenic aerosols from Europe, desert dust from North Africa and the Middle East, and maritime aerosols from the Mediterranean Sea and the Atlantic Ocean (e.g. Mihalopoulos et al., 1997; Lelieveld et al., 2002; Sciare et al., 2003; Pace et al., 2006; Lyamani et al., 2006, Gerasopoulos et al., 2006). The role of aerosols is enhanced because of the large amount of solar radiation reaching the Mediterranean basin, especially during the summer cloud-free conditions. The regional annual mean value of aerosol optical depth at 550 nm (AOD_{550}), a good measure of atmospheric aerosol loading, has been recently determined at 0.22 (Papadimas et al., 2008) a value 1.3 times larger than the corresponding global average of 0.17 (Remer et al., 2008). Nevertheless, a typical feature in the region is the strong spatial and temporal variability of the aerosol, modulated by the local, synoptic and large-scale circulation (di Sarra et al., 2001; Kallos et al., 2007).

The aerosol radiative effects over the Mediterranean region have been studied by many researchers in the last decade (e.g. Formenti et al., 2002; Markowicz et al., 2002; Meloni et al., 2003; Haywood et al., 2003; Balis et al., 2004; Roger et al., 2006; di Sarra et al., 2008; Saha et al., 2008, Bergamo et al., 2008, Santos et al., 2008; Di Biagio et al., 2009, 2010; Kazadzis et al., 2009; Santese et al., 2010). Nevertheless, most of these studies are temporally and spatially limited, since they are based on station measurements at specific sites or experimental campaigns that took place in parts of the Mediterranean region (e.g. MINOS experiment, Sahara Dust Experiment, SHADE). Moreover, most of the earlier published DRE computations refer either to anthropogenic or to natural aerosols, separately. Understanding of the aerosol effects on the atmospheric dynamics and climate of the region requires investigation of the spatial variation of the aerosol effects on the regional radiation budget, accounting for both natural and anthropogenic aerosols that co-exist and interact in the region (e.g. Hatzianastassiou et al., 2009). To this aim, it is crucial to perform computations of aerosol radiative effects for the entire Mediterranean basin.

The present modelling study attempts to compute, for the first time to our knowledge, the direct effect of aerosols (natural plus anthropogenic) on the radiative energy budget of the broader Mediterranean Basin (29.5° N–46.5° N and 10.5° W–38.5° E) for the 7 yr period from March 2000 to February 2007 using contemporary satellite and reanalyses data. This is achieved by using the available observational data as input to a detailed spectral radiative transfer model (RTM). The RTM uses as input spectral aerosol optical properties (AOD, asymmetry parameter, g_{aer}, and single scattering albedo, ω_{aer}) taken mainly from the MODerate resolution Imaging Spectroradiometer (MODIS) of NASA (National Aeronautics and Space Administration) and supplemented by the Global Aerosol Data Set (GADS). The rest of the surface and atmospheric parameters that are

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required as input such as clouds, water vapour, surface albedo and ozone, are taken from comprehensive global climatological databases (mainly the International Satellite Cloud Climatology Project, ISCCP) and global reanalyses (NCEP/NCAR). The model computes the aerosol DRE on the various components of the regional SW radiation budget, namely at TOA (i.e. on the reflected solar radiation), within the atmosphere (on the absorbed solar radiation), and at the surface (on the incoming and absorbed solar radiation) at a spatial resolution of 2.5° × 2.5° latitude-longitude and on a monthly mean basis. The computations are performed under real all-sky conditions, but also under clear-skies. Apart from the geographical patterns and seasonal variability of DREs, the aerosol-induced inter-annual changes of solar radiation in the Mediterranean basin over the study period are also estimated by applying linear regression fits to the 7 yr time series of DRE components.

The methodology and the description of RTM and its input data are given in Sect. 2. The model results and the validation of SW fluxes and aerosol DREs are discussed in Sect. 3, whereas the results of the model sensitivity tests are presented in Sect. 4. Conclusions are summarized in Sect. 5.

2 Model and input data

2.1 The spectral radiative transfer model

The spectral radiative transfer model (Vardavas and Carver, 1984; Hatzianastassiou et al., 2004a, 2007a, b; Vardavas and Taylor, 2007) computes solar radiative fluxes at 118 wavelengths in the range 0.2–1.0 µm and 10 spectral intervals in the range 1.0–10 µm. The computations are performed for each 2.5° latitude × 2.5° longitude cell of the region under both clear- and cloudy-sky conditions, considering ozone absorption, Rayleigh scattering, and absorption by water vapour, carbon dioxide and methane. Scattering and absorption by clouds (low, middle and high) and aerosols, and reflection from the Earth's surface are also taken into account. A complete description and detailed model considerations can be found in previous publications (Hatzianastassiou and Vardavas, 1999, 2001; Hatzianastassiou et al., 2004a, b, c, 2005, 2007a). The aerosol DRE is computed as the difference between model computed fluxes with (F_i) and without ($F_{i,\text{no-aerosol}}$) the presence of aerosols:

$$\text{DRE}_{\text{atm}} = F_{\text{atm}} - F_{\text{atm,no-aerosol}} \tag{1}$$

$$\text{DRE}_{\text{surf}} = F_{\text{surf}} - F_{\text{surf,no-aerosol}} \tag{2}$$

$$\text{DRE}_{\text{TOA}} = F_{\text{TOA,no-aerosol}} - F_{\text{TOA}} \tag{3}$$

The DRE components quantify the effect of aerosols on solar radiation at TOA, DRE_{TOA}, on the absorbed radiation within the atmosphere, DRE_{atm}, and on the downward, DRE_{surf}, and net downward or absorbed radiation $\text{DRE}_{\text{netsurf}}$ at the Earth's surface.

Apart from DRE, the aerosol radiative efficiency ($E_{\text{AOD},i}$) which is used to isolate the AOD dependence of DRE, is also investigated in this study. $E_{\text{AOD},i}$ (Eq. 4) is the aerosol perturbation of solar radiation, at TOA, in the atmosphere and at the surface ($E_{\text{AOD,TOA}}$, $E_{\text{AOD,atm}}$, $E_{\text{AOD,netsurf}}$, respectively) per unit of optical depth (AOD_{550}^{-1}) and is expressed in Wm^{-2}

$$E_{\text{AOD},i} = \text{DRE}_i / \text{AOD}_{550} \tag{4}$$

(AOD is usually given at the 0.5 or 0.55 µm visible wavelengths). Provided that $E_{\text{AOD},i}$ is known, for a given AOD, the product of AOD and $E_{\text{AOD},i}$ provides an estimate of aerosol climatic effects (Anderson et al., 2005; Christopher and Jones, 2008). $E_{\text{AOD},i}$ is also useful since it removes the significant geographical dependence of AOD, and thus can be used to evaluate the performance of models.

As another useful measure of the impact of aerosols on solar radiation we define the so-called aerosol radiation (budget) efficiency (ARBE). This quantifies the ability of aerosols to modify the solar radiation fluxes, and can be defined as

$$\text{ARBE}_i = \text{DRE}_i / F_{i,\text{no-aerosol}} \tag{5}$$

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which, based on Eqs. (1), (2), and (3), is equivalent to

$$\mathrm{ARBE}_i = F_i / F_{i,\mathrm{no-aerosol}} - 1 \tag{6}$$

2.2 Aerosol optical properties

Spectral aerosol optical properties (AOD, ω_{aer}, and g_{aer}) are required for model computations at all wavelengths and spectral bands from 0.25 to 10 µm. In this study, we use mean monthly AOD and g_{aer} MODIS Terra (Collection 5 and 5.1) Level-3 data (http://modaps.nascom.nasa.gov/), for the available wavelengths, i.e. 0.47, 0.55, 0.66, 0.87, 1.24, 1.64, and 2.13 micrometers, supplemented by GADS data for the remaining wavelengths. The MODIS data are available over land and ocean and provide complete spatial and temporal coverage over the broader Mediterranean basin. The pre-launch uncertainty of the MODIS AOD is ±0.05 ±0.2 × AOD over land (Chu et al., 2002), and ±0.03 ±0.05 × AOD over ocean (Remer et al., 2002). Over the Mediterranean basin, according to an extensive validation of MODIS Collection 005 against AERONET (Papadimas et al., 2009), the MODIS AOD data are slightly (by 1 %) underestimated. The ω_{aer} data are taken from GADS because MODIS ω_{aer} data (Deep Blue) are not available for the study period, they are limited over land and, in addition, they do not entirely cover the study region, especially its northern part (southern European coastal areas). Furthermore, MODIS Deep Blue ω_{aer} data have not been adequately evaluated. The originally taken from GADS ω_{aer} data, are re-computed for actual relative humidity values for the aerosol layer as explained by Hatzianastassiou et al. (2004a) and extensively discussed by Hatzianastassiou et al. (2007a). We have assessed our re-computed, and finally used, GADS ω_{aer} data through comparisons against ω_{aer} data from MODIS deep-blue (Hsu et al., 2004). It was found that at 550 nm they are underestimated with respect to MODIS ω_{aer} data by 0–10 %, with an overall, i.e. regional mean, underestimation of 8 %. We have performed a similar inter-comparison for the globe, which revealed an overall underestimation by GADS ω_{aer} equal to 3 %.

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2.3 Surface and atmospheric data

The monthly mean water vapour and temperature data were taken from the NCEP/NCAR Global Reanalysis Project (Kalnay et al., 1996; Kistler et al., 2001). Monthly mean cloud properties (cloud amount, cloud-top pressure, cloud-top temperature, liquid water path, cloud albedo, and optical depth for total, as well as low-, mid-, and high-level clouds) on $2.5^{\circ} \times 2.5^{\circ}$ grid cell resolution were taken from the latest D2 stage data of ISCCP (Rossow et al., 1996; Rossow and Schiffer, 1999). The treatment of surface reflection is explained in detail by Vardavas and Koutoulaki (1995) and Hatzianastassiou et al. (2004a, 2005, 2007a). The mean monthly 2.5-degree latitude-longitude grid cell data of the total O_3 column abundance (in Dobson Units) were taken from TOVS, archived in the ISCCP-D2 package. For CO_2 a fixed total atmospheric amount is taken, equal to $0.54\,\mathrm{g\,cm^{-2}}$, corresponding to 345 parts per million by volume (ppmv). The treatment of solar absorption by atmospheric air molecules is explained by Hatzianastassiou et al. (2004a, 2007a), whereas the Rayleigh scattering, due to air molecules, is considered in the model in the same way as in Vardavas and Carver (1984) and Hatzianastassiou et al. (2004a).

3 Results

3.1 Model validation

The aerosol direct effect on surface solar radiation, i.e. $\mathrm{DRE_{surf}}$, is very important because SSR has a key role for many processes of the Earth-atmosphere system, for example surface heating or evaporation. Before the presentation of model computed DRE results, it is important to assess their reliability. Unfortunately, direct validation of the model computation of $\mathrm{DRE_{surf}}$ is prohibited by the absence of measurements of the surface solar radiation fluxes in absence of aerosol particles, $F_{\mathrm{surf,no-aerosol}}$ (Eq. 2). Therefore, the quality of model computations of $\mathrm{DRE_{surf}}$ can be assessed only by the

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validation of the computations of the other component, i.e. of surface solar radiation in presence of aerosols, F_{surf} (Eq. 2).

The quality of model DRE_{surf} in this study was assessed by validating the monthly mean $2.5^{\circ} \times 2.5^{\circ}$ downward surface solar radiation (SSR) fluxes over the broader Mediterranean basin, through comparisons with corresponding high-quality surface measurements, over the study period (2000–2007). The surface SSR measurements were taken from the Global Energy Balance Archive (GEBA, Gilgen and Ohmura, 1999). GEBA database, redesigned and updated in 1994 and 1995, contains quality controlled monthly mean energy fluxes at the Earth's surface from sites all over the world. The GEBA data are widely used for validation of model and satellite remote sensing retrieval algorithms because of the large number of stations and the long observational period (Wild et al., 1995; Kiehl and Trenberth, 1997; Hatzianastassiou et al., 2005).

GEBA and modelled SSR fluxes have been compared for those pixels including GEBA stations that are located within the study region and operated within the period 2000–2007. A total of 80 GEBA stations satisfied this criterion and were used in this study for the model evaluation. These stations ensure a satisfactory spatial coverage of our study region being homogeneously distributed. Figure S1a in the Supplement presents the overall comparison between model and GEBA monthly SSR fluxes. There is a large number of matched data pairs ($N = 4843$), enabling thus good statistics. The scatterplot comparison reveals a good agreement between model and GEBA SSR, with a bias equal to $-8.1\,\mathrm{Wm^{-2}}$ (or $-5.3\,\%$), indicating a general model underestimation of SSR, with a relatively small scatter ($\mathrm{SD} = 19.08\,\mathrm{Wm^{-2}}$). The correlation coefficient between model and GEBA fluxes is satisfactory and equal to 0.95.

Apart from the overall comparison, the ability of model SSR to reproduce the interannual variability and tendencies of SSR based on surface measurements, has been also tested on the basis of complete time-series. For this purpose, in order to ensure reliable estimations of SSR tendencies, those GEBA stations providing at least 80 % of the total number of monthly data over our study period (2000–2007) have been

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selected. Only four (Eq. 4) GEBA stations met this criterion. In Table S1 of Supplement the model computed changes of SSR, Δ (SSR), over the period 2000–2007 are compared with the corresponding changes from GEBA measurements, for each station. The computed correlation coefficients and the relative differences between the two sets of SSR fluxes are also given. Figure S1b in Supplement shows the inter-annual variability and tendencies of the model and GEBA SSR fluxes, based on applied linear regression fits to the two time-series. The comparisons reveal that our model is able to adequately reproduce the inter-annual variation and the tendencies of SSR.

3.2 Regional mean patterns

3.2.1 DRE_{TOA}

The seven-year (2000–2007) average geographical distribution of aerosol direct radiative effect on the outgoing shortwave radiation at TOA, for all-sky conditions, over the broader Mediterranean basin is given in Fig. 1i-a. In general, negative DRE_{TOA} values prevail indicating that aerosols increase, through reflection, the outgoing solar radiation at TOA (see Eq. 3), i.e. they cool the Earth-atmosphere system above much of the study region. In contrast, more rare positive DRE_{TOA} values indicate that local planetary warming also occurs. The model 7 yr regional mean aerosol DRE_{TOA} (see also Table 1) is equal to $-2.4 \pm 0.3\,\mathrm{W\,m^{-2}}$ (standard deviation corresponds to inter-annual variability), indicating that on an annual basis, aerosols induce a significant "planetary" cooling over the broader Mediterranean basin, equivalent to an increase of the mean regional planetary albedo by 2.3 %. Our analysis shows, however, that there is spatial variability of DRE_{TOA} within the study region, for example between the eastern, central and western Mediterranean basins (DRE_{TOA} values equal to −2.5, −3.4, and $-1.2\,\mathrm{W\,m^{-2}}$, respectively, see Tables S2, S3, S4 of Supplement). Local DRE_{TOA} values range mostly between −7 and $0\,\mathrm{W\,m^{-2}}$ (planetary cooling) but reach positive values (planetary warming) up to 28 $\mathrm{W\,m^{-2}}$. The planetary warming arises from the significant absorption of solar radiation either by natural mineral dust aerosols over the

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bright reflecting surfaces of Sahara desert, or by polluted anthropogenic aerosols over the ice- or snow-covered mountain slopes of the Alps. This effect is enhanced through multiple reflections between the absorbing aerosol layers and the highly reflecting surfaces underneath (e.g. Charlson et al., 1992; Haywood and Shine; 1997; Myhre et al., 1998; Hatzianastassiou et al., 2004a, b; 2007a).

The spot of aerosol planetary warming over the Alps is interesting and similar, though smaller in magnitude (due to smaller AOD and surface albedo values), to the corresponding warming effect of Arctic haze that has been documented in the literature (Rinke et al., 2004; Treffeisen et al, 2005; Law and Stohl, 2007). The very large positive DRE_{TOA} values over the Sahara desert (up to 28 Wm^{-2}) are due to high aerosol loading (AOD) and to low ω_{aer} values there (down to 0.88 at 550 nm). Note that computed DRE_{TOA} over the Sahara desert are very sensitive to the adopted ω_{aer} values since a decrease by up to 10.6 Wm^{-2} is calculated when ω_{aer} at 550 nm is increased by 10 %. A similar to Sahara desert aerosol effect (planetary warming, DRE_{TOA} >0) is also observed over the Middle East. On the other hand, the largest negative values, up to −7 Wm^{-2}, are computed over the Mediterranean Sea, and are due to high aerosol loads (up to 0.4, see Papadimas et al, 2008) and to low sea surface albedo (0.08 to 0.1).

The largest (negative or positive) DRE_{TOA} (absolute) values are found over the southern part of the study region, i.e. Mediterranean Sea, northern Africa and Middle East, and arise from the combination of significant aerosol loads (AOD), cloud-free conditions, and high solar radiation and land surface albedo. On the opposite, the highly populated and industrialized urban areas of central and west Europe are found to have relatively smaller DRE_{TOA} values (up to −1.5 Wm^{-2}) due to the absence of synergistic effect of the above mentioned factors. To investigate the role of cloudiness, DRE_{TOA} has been computed under clear-sky conditions (see Fig. 1i-b). The results show that the aerosol effect is larger than under all-sky conditions, with a 7 yr regional mean DRE_{TOA} equal to -4.5 ± 0.4 Wm^{-2} (Table 1), i.e. by 87.5 % larger than all-sky, and with local DRE_{TOA} values ranging between −11 and 38 Wm^{-2}.

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On a seasonal basis (Fig. S2, Supplement) the largest negative DRE_{TOA} values over the sea are found in the eastern Mediterranean basin during winter and spring, over the western Mediterranean during summer and over central Mediterranean in autumn and spring. This is in accordance with the MODIS-based aerosol load seasonal patterns (Papadimas et al., 2008) supporting thus the importance of AOD in determining the magnitude of DRE_{TOA}. The seasonal regional mean aerosol DRE_{TOA} values, and the corresponding standard deviations, are equal to -0.9 ± 0.1, -2.4 ± 0.7, -4.3 ± 0.5, -1.9 ± 0.5, for winter (December to February), spring (March to May), summer (June to August), and autumn (September to November), respectively (see also Table 1), showing a clear maximum in summer. The corresponding seasonal regional mean AOD_{550} values are 0.13 ± 0.02, 0.22 ± 0.03, 0.24 ± 0.03, $0.16 \pm 0.03\,\mathrm{Wm^{-2}}$, also with a summer maximum, but not so distinct as that of DRE_{TOA} and comparable to the spring AOD_{550} maximum. Note that the summer/winter ratio of DRE_{TOA} is equal to 4.8, which is larger than the corresponding summer/winter ratio of AOD_{550} (1.8). The summer/spring ratios of DRE_{TOA} and AOD_{550} are equal to 1.8 and 1.1, respectively, i.e. again higher for DRE_{TOA} than AOD_{550}. These results highlight the important role of available solar radiation for the magnitude of DRE_{TOA}, apart from that of aerosol optical properties. The summertime primary maxima of DRE_{TOA} reflect the high solar radiation and the impact of pollution aerosols that are abundant in the absence of rain during summer. Furthermore, the secondary maxima in spring demonstrate the influence of desert dust transport, from North Africa and Middle East, to the Mediterranean during spring (Barnaba and Gobbi, 2004; Papadimas et al., 2008).

3.2.2 DRE_{atm}

Aerosols are found to increase the atmospheric absorption (Fig. 1ii) of solar radiation over the Mediterranean basin mostly by up to $12\,\mathrm{Wm^{-2}}$, but the effect can be much larger, up to a several tens of $\mathrm{Wm^{-2}}$, over specific areas with absorbing dust aerosols (e.g. Markowicz et al., 2002), like the Anatolia peninsula, Middle-East and northern Africa. Annually, the regional average DRE_{atm} is equal to $11.1 \pm 0.7\,\mathrm{Wm^{-2}}$ (Table 1).

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According to our computations, this value is about triple that of the global mean DRE_{atm} value of 3.9 Wm^{-2}, verifying thus that Mediterranean has one of the most significant absorbing aerosol loadings in the world (e.g. Lelieveld et al., 2002). DRE_{atm} values as high as 12 Wm^{-2} are also computed for central Europe, associated with absorbing pollution aerosols, while over the Mediterranean Sea the values vary between 5 and 10 Wm^{-2}. The DRE_{atm} values are higher over the Mediterranean Sea than the Atlantic Ocean by factors of 2–3, because the Mediterranean Sea experiences higher aerosol background levels than the open Atlantic Ocean, due to long range and regional transport from upwind continental areas (e.g. Querol et al., 2009; Kanakidou et al., 2011). The excessive amounts of aerosol solar absorption (>30–40 Wm^{-2}) over the Middle-East and North Africa are partly attributed to the low ω_{aer} values there (down to 0.88 at 550 nm). Increasing ω_{aer} by 10 %, results in a decrease of the computed DRE_{atm} values over these areas by 22–29 Wm^{-2}.

On a seasonal basis, the DRE_{atm} values are even stronger. In summer, the regional mean value is equal to 17.5 Wm^{-2} (Table 1) whereas local values exceed 100 Wm^{-2} (over Africa and the Middle East) mainly during summer, but also in spring (Figs. S3iii and S3ii, respectively, Supplement). On the other hand, in winter DRE_{atm} decreases down to 4.5 Wm^{-2}, showing again that DRE_{atm} is strongly dependent on the available solar radiation, and certainly more than on AOD. Indeed, the computed ratio DRE_{atm}-summer/DRE_{atm}-winter (equal to 3.9) is roughly double the corresponding ratio of seasonal AODs (AOD_{550}-summer/ AOD_{550}-winter equal to about 2). It is interesting to note the local maxima of DRE_{atm} (about 15–20 Wm^{-2}, Fig. S3i, S3ii) computed for central and northern Europe only in winter and spring that can be attributed to absorbing aerosols from local anthropogenic activities. These aerosols are known to be transported towards the Arctic and constitute the Arctic haze.

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3.3 DRE_{surf} and $DRE_{netsurf}$

Aerosols, through scattering and absorption, decrease drastically the downwelling (DRE_{surf}) and absorbed ($DRE_{netsurf}$) solar radiation at the Mediterranean surface, by 16.5 and 13.5 Wm^{-2}, respectively (Table 1), on an annual mean basis, producing thus an important surface radiative cooling. This aerosol surface cooling is significant (about 10 %) as compared to the downward and absorbed solar radiation at the region's surface (172.4 ± 1.8 and 147.0 ± 1.7 Wm^{-2}, respectively). The patterns of DRE_{surf} and $DRE_{netsurf}$ are very similar, and thus only the latter is shown in Fig. 1iii. The largest (absolute) values are found over the deserts of Middle East and North Africa, where the surface solar absorption is decreased by 25–55 Wm^{-2} (19–60 %). Smaller, though still important $DRE_{netsurf}$ values (12–15 Wm^{-2}) are found over the Mediterranean Sea, whereas values <10 Wm^{-2} are found over the European continent. On a seasonal basis, the mean regional $DRE_{netsurf}$ values are equal to −5.4 ± 2.1, −17.1 ± 3.2, −21.8 ± 0.9, and −9.6 ± 4.6 Wm^{-2} for winter, spring, summer, and autumn, respectively (Table 1). Nevertheless, the values are much larger at the 2.5° × 2.5° grid cell level, rising up to 90 Wm^{-2} in summer (Fig. S4iii, Supplement). In general, the seasonal patterns of $DRE_{netsurf}$ are similar to those of DRE_{atm} already discussed, for example, they also exhibit the local maximum in central-northern Europe during winter (Fig. S4i, Supplement).

Given that DRE_{TOA} and $DRE_{netsurf}$ depend on aerosol scattering and extinction (scattering+absorption) respectively, their ratio, $DRE_{TOA}/DRE_{netsurf}$ is a good indicator of aerosol absorption. When it takes values close to 1.0, aerosols are almost pure scatterers, while strongly absorbing aerosols are characterized by $DRE_{TOA}/DRE_{netsurf}$ values approaching zero. Figure 2i displays the 7 yr average spatial distribution of $DRE_{TOA}/DRE_{netsurf}$ ratio. Over the seas, the ratio values are higher than 0.5, associated with the presence of scattering sea-salt aerosols, while over land the ratio takes values smaller than 0.2 indicating more absorbing continental aerosols. The computed mean regional ratio $DRE_{TOA}/DRE_{netsurf}$ is equal to 0.18, indicating the presence

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of strongly absorbing aerosols over the Mediterranean basin. Figure 2ii shows the seasonal variation of regional mean $DRE_{TOA}/DRE_{netsurf}$ values. Relatively smaller values (0.09–0.16) are found in spring and early autumn (September), associated with the presence of absorbing aerosols, mainly dust, spread out over extended areas of the Mediterranean basin under frequently occurring southerly cyclonic circulation. Larger $DRE_{TOA}/DRE_{netsurf}$ values are seen in summer and winter (0.19–0.27) owing to the contribution of scattering, mainly anthropogenic, aerosols emitted in Europe and subsequently transported southwards through favourable anticyclonic circulation (e.g. Azores anticyclone in summer). The large winter $DRE_{TOA}/DRE_{netsurf}$ values (peak in November) might be associated with the efficient wash-out of aerosols by rain, which is maximum during this season in the Mediterranean, controlled thus by the influence of regional/local emissions, composed mainly by sea-salt and sulfate aerosols.

The significant SSR reduction induced by aerosols translates to a surface cooling that, according to our computations, can be as large as $-0.029\,\mathrm{K\,day^{-1}}$ over the Mediterranean Sea, and up to $-2.04\,\mathrm{K\,day^{-1}}$ over North Africa. Such a large surface cooling, together with the significant atmospheric warming of up to $0.22\,\mathrm{K\,day^{-1}}$ over the Mediterranean Sea and $2.2\,\mathrm{K\,day^{-1}}$ over North Africa, can influence the regional atmospheric dynamics by decreasing the atmospheric temperature gradient, thus leading to more stable atmospheric conditions. This can have important implications for the region's climate (e.g. possible effects on clouds and precipitation) in view of the changing aerosol loadings and composition observed over the Mediterranean basin on decadal timescales (e.g. Perez et al., 2008; Papadimas et al., 2008; Koukouli et al., 2009).

3.4 Aerosol radiative efficiency

Aerosol DREs strongly depend on AOD, apart from other parameters like the available solar flux itself (e.g. Hatzianastassiou et al., 2004b). The dependence of DRE on AOD is here investigated by means of aerosol radiative efficiency, i.e. the aerosol perturbation of solar radiation, at TOA, in the atmosphere and at the surface ($E_{AOD,TOA}$,

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$E_{AOD,atm}$, $E_{AOD,netsurf}$, respectively) per unit of optical depth (AOD_{550}^{-1}), computed by Equation 4. The slope of the applied linear regression fit to E_{AOD} values as function of AOD_{550}, can be also defined as the radiative efficiency (E_{AOD}; Eq. 4). This slope is actually a more robust measure of E_{AOD} than the simple ratio (Christopher and Jones, 2008). Therefore, we followed this methodology (linear regression fitting) to estimate the aerosol radiative efficiency in the Mediterranean. The scatterplots, linear fits, and the obtained relationships between AOD_{550} and DRE_i can be seen in Fig. 3. The derived relationships enable a rough estimation of aerosol radiative effects over the Mediterranean basin if we know the AOD_{550} value. The larger E_{AOD} values at the surface and in the atmosphere indicate the strong dependence of surface and atmospheric aerosol radiative effects on AOD. Thus, knowing AOD_{550} or AOD_{550} changes, $DRE_{netsurf}$ and DRE_{atm} changes can be reasonably well estimated. On the contrary, the aerosol effects at TOA depend much less on AOD. E_{AOD} under clear-sky conditions does not significantly change from that under all-sky conditions (see Fig. 3i,ii,iii-b). In all scatterplots, there appears a group of points (red colour) which is separated from the main body of blue points. Although their number is relatively small, 4 % of the total number of points, they seriously affect the applied linear regression fits to the overall population of points. Therefore, linear regression fits have been applied separately to the two groups of points. It is seen that for $E_{AOD,atm}$ and $E_{AOD,netsurf}$ the two liner fit lines are almost parallel, but the intercept is larger (absolute terms) in the case of red points, indicating larger DREs for them. Moreover, for $E_{AOD,TOA}$, the slope changes sign and becomes positive, opposite to the negative slope for blue points. Positive and negative slopes for $E_{AOD,TOA}$ indicate planetary warming and cooling due to aerosols. The performed analysis revealed that red points (planetary warming) correspond to geographical cells that are systematically located in northern Africa, and correspond to absorbing dust aerosols. In all cases however, the obtained linear fit equations indicate that increasing AODs produces amplified magnitudes of DRE_{TOA}, DRE_{atm} and $DRE_{netsurf}$.

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To further examine the impact of aerosols on solar radiation, the aerosol radiation (budget) efficiency (ARBE, Eqs. 5 and 6) has been computed. The $ARBE_i$ values (Fig. 4) indicate above the broader Mediterranean basin, a 1–22 % increase of the outgoing solar radiation (OSR) at TOA due to aerosols under all-sky conditions, with a stronger influence over the Mediterranean Sea and over north Africa (Fig. 4i-a). Under all-sky conditions, the aerosol influence is largely dominated by the presence of clouds that have much larger optical depth than aerosols, or by the strong land-surface (desert) reflection. The influence of aerosols is stronger in case of small cloudiness or for massive aerosol loads, like those over deserts. Under clear-sky conditions (Fig. 4i-b) the aerosol influence is amplified, especially over dark ocean surfaces. For example, see the large increase of $ARBE_{TOA}$ over the Atlantic Ocean and Black Sea, while similarly small values are found over the European continent. The $ARBE_{TOA}$ values under clear-sky conditions can be as large as 33 %. The modification of solar atmospheric absorption by aerosols above the study region (Fig. 4ii-a, b) is larger than that of reflected radiation to space, with values ranging between 5 and 105 %. Values of 10–20 % are computed above extended areas of the study region, whereas they reach 30–100 % above vast desert areas with high aerosol loads (AOD_{550} up to 0.4). Similar geographical patterns, but with even higher values (10–150 %) are computed under clear-sky conditions. The significant effect of aerosols on solar atmospheric absorption over the industrial regions of central Europe can be seen, with values up to 25 % in all-sky and 50 % in clear-sky conditions, respectively. Our results highlight the key role of aerosols for solar heating of the region's atmosphere, since they can even double it in case of significant absorbing aerosol amounts. Nevertheless, it is worth noting the absence of large $ARBE_{atm}$ values over the Mediterranean Sea or the Atlantic Ocean, even under clear-skies, opposite to $ARBE_{TOA}$. This indicates the very weak absorption ability of sea-salt or sulphate maritime aerosols.

Aerosols reduce the downward solar radiation at the Mediterranean's surface by 3–38 % under all-sky conditions (Fig. 4iii-a), with the largest percent decrease observed primarily over North Africa, Middle-East and the Anatolian peninsula, and secondarily

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over central and northern Europe. Under clear-sky conditions, the geographical patterns of $ARBE_{surf}$ remain about the same with those in case of all-sky, but the values are higher by about 25 %.

Figure 4 reveals the significant role of aerosols in modifying the solar radiation budget of the Mediterranean basin, inducing important changes in the solar radiative fluxes at TOA, in the atmosphere, and at the region's surface. Yet, our results suggest that this role becomes even more important in the case of solar atmospheric absorption.

3.5 Seasonal and inter-annual variation of aerosol DREs

Figure 5 shows the seasonal variation of aerosol DREs for the Mediterranean basin under all-sky and clear-sky conditions separately. For all-sky conditions, negative and positive values of $DRE_{netsurf}$ and DRE_{atm}, respectively, indicate that in the Mediterranean, aerosols reduce the absorbed solar radiation at the surface, $DRE_{netsurf}$, by 3.9–22.6 Wm^{-2}, and enhance the absorbed solar radiation within the atmosphere, DRE_{atm}, by 3.2–18.4 Wm^{-2}. Hence, $DRE_{netsurf}$ and DRE_{atm} exhibit significant seasonal variations, contrary to DRE_{TOA} that shows weak seasonality varying between −0.7 and −4.2 Wm^{-2}. The maximum aerosol radiative effects are observed in summer and spring, and the minimum in winter, following thus the seasonal variation of the corresponding solar radiation fluxes, but also of the aerosol loading (AOD_{550}, seasonal cycle shown in Fig. 5).

The role of cloudiness for aerosol DREs can be assessed by comparing the results of Fig. 5 for all- and clear-skies. As expected, the clear-sky DREs absolute values are consistently larger than the all-sky ones. $DRE_{netsurf}$ is reaching about 27 Wm^{-2} and presents clear maxima in summer an spring, reflecting primarily the seasonality of the available solar radiation fluxes and secondarily that of AOD. The role of AOD is strengthened in the absence of clouds, as shown by the significant spring peaks that appear in clear-sky conditions for all DRE components.

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The time-series of regional mean aerosol DREs are displayed in Fig. 6. All-sky DRE_{TOA} (Fig. 6a) exhibits an annual cycle with clear maximum absolute values in summer (see also Fig. 5) when it ranges between −4.0 and −5.5 Wm^{-2}, and minimum values in winter (DRE_{TOA} between 0 and −0.9 Wm^{-2}). There is, however, a significant year-to-year variability, in terms of both maximum and minimum values. For example, the summer values of 2002 and 2003 are larger (in absolute terms) by about 1 Wm^{-2} than those of other years, while the winter values of 2003 and 2005 are very close to zero. Under clear-sky conditions, the magnitude of DRE_{TOA} is consistently larger, with a secondary peak in spring (mainly in April) reflecting the AOD seasonality as earlier discussed. The inter-annual variation of DRE_{atm} and $DRE_{netsurf}$ is smoother than DRE_{TOA}, with maximum and minimum absolute values observed in summer and winter, respectively, regardless of sky conditions. DRE_{atm} values vary within the range 2–24 Wm^{-2}, with slightly (by about 2–3 Wm^{-2}) larger magnitude under clear-skies, whereas the $DRE_{netsurf}$ regional mean values range from −2.7 to −31 Wm^{-2} over the period 2000–2007 again being larger (by up to about 6 Wm^{-2}) under clear-sky than under all-sky conditions.

Given that aerosol properties are changing with time (e.g. Mischenko et al. 2007; Perez et al., 2008; Papadimas et al., 2008) it is expected that aerosol radiative effects, DREs, are also subject to variations with time. Such variations produce modifications of solar radiation that contribute to the overall solar radiation changes (solar dimming and brightening when referring to the Earth's surface) that, apart from aerosols, are also produced by other parameters of the Earth-atmosphere system, namely clouds, water vapour or other trace gases. Here, we attempted to estimate the aerosol-induced inter-annual changes of solar radiation in the Mediterranean basin over the study period, by applying linear regression fits to the 7 yr time series of DRE components. Our results indicate that $DRE_{netsurf}$ has decreased in magnitude by 2.48 Wm^{-2} (or by 18.4 %) from 2000 to 2007, resulting thus in an equivalent increase of surface solar radiation (solar brightening). According to our results, decreases of $DRE_{netsurf}$ are calculated for all seasons, as shown in Table S5 of Supplement. Over the same period, the surface

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solar radiation over the broader Mediterranean basin has decreased by 25.6 Wm^{-2} (solar dimming). Therefore, it appears that the aerosol brightening has been overwhelmed by the dimming produced by the other parameters. Indeed, ISCCP data analysis shows an increase in cloudiness over the Mediterranean from 2000 to 2007 equal to 7 %, and a corresponding increase in total precipitable water by 5.6 % (2 mm). The decrease in $DRE_{netsurf}$ from 2000 to 2007 is almost identical under clear- and all-sky conditions, which means that it is not related to clouds, but to the regional aerosol loading. Indeed, over the same period AOD_{550} has decreased by 0.05 or by 27 %, in line with the increased aerosol removal by enhanced precipitation (see Papadimas et al., 2008).

4 Sensitivity of aerosol DREs

Despite the considerable number of past studies (e.g. Koepke and Hess, 1988; Charlson et al., 1992; Penner et al., 1994; Hansen et al., 1997; Haywood and Shine, 1997; Schult et al., 1997; Penner et al., 1998; Haywood and Ramaswamy, 1998; Myhre et al., 1998; Ramanathan et al., 2001; Yu et al., 2006; Hatzianastassiou et al., 2004a, b; 2007a) the magnitude of aerosol radiative effects and forcing, is still uncertain (IPCC, 2007). This uncertainty is largely attributed to the aerosol properties used as input data in the radiation transfer and climate models. Among these aerosol properties, the most important are AOD, g_{aer} and ω_{aer}. Therefore, a series of sensitivity tests were performed with our model to assess the uncertainty of the obtained DREs, due to possible inaccuracies of the utilized input AOD, g_{aer} and ω_{aer} data.

The results of the sensitivity tests are given in terms of absolute and percentage changes of DREs (Eqs. 7 and 8, respectively), denoted as $\Delta(DRE_i)$ and $\Delta(DRE_i)\,\%$ respectively, and defined as

$$\Delta(DRE_i) = DRE_i - DRE_{ref,i} \tag{7}$$

$$\Delta(DRE_i)\% = \left[(DRE_i - DRE_{i,ref})/DRE_{i,ref}\right] \times 100 \tag{8}$$

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where $DRE_{ref,i}$ are the aerosol DREs for the reference simulation and DRE_i are the corresponding DREs for each sensitivity simulation. The reference DREs are those for the year 2003. Their annual regional mean values, namely $DRE_{ref,TOA}$, $DRE_{ref,atm}$, and $DRE_{ref,surf}$, are equal to −2.6, 11.2 and −16.9 Wm^{-2}, respectively (Table 2).

To examine the sensitivity of the reference DREs to the uncertainty of the three aerosol optical properties, we have increased and decreased each one of them by 10 %, and computed the resulting changes in DRE_i, $\Delta(DRE_i)$, that are given in Table 2. Increasing/decreasing AOD by 10 % resulted in an increase/decrease of all DRE components. More specifically, DRE_{TOA} has increased by 8.4 %, DRE_{atm} by 9.3 % and DRE_{surf} by 9 %. So, it appears that aerosol DRE at TOA, surface and in the atmosphere, show similar sensitivity to variations of AOD. Therefore, the slight (1 %) underestimation of MODIS C005 AOD used in our modelling study (with respect to AERONET, see Sect. 2.2) as evaluated by Papadimas et al. (2009), induces a smaller than 1 % underestimation of the computed DREs discussed in Sect. 3. Based on these sensitivity tests the impact of possible current or future variability of natural or anthropogenic AOD in the Mediterranean can be estimated. For example, increased AOD values due to increased anthropogenic emissions would result in enhanced "planetary" cooling over the region, arising from enhanced surface cooling and atmospheric warming. The opposite effects, i.e. planetary and surface warming along with atmospheric cooling, are expected for decreased AOD values that, for example, can result from actions aiming to reduce aerosol emissions, like the Air Framework Directive or the Clean Air for Europe Programme.

Furthermore, a similar to AOD percent increase/decrease of ω_{aer}, i.e. by ± 10 %, results in significantly stronger variations of aerosol DREs. As expected, the biggest effect is on aerosol atmospheric warming (DRE_{atm}), which is modified by up to about ± 70 %, whereas the effects on DREs at TOA and surface are smaller (up to about ± 40 %) though still very important. Therefore, possible future changes in ω_{aer}, as for example smaller ω_{aer} values associated with increasing anthropogenic or natural biomass burning activities or increasing emissions of black carbon, may lead to

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enhanced atmospheric heating, combined with a strengthened surface cooling. These effects may be even stronger if they are accompanied by increased AOD levels. In this case, sensitivity tests performed with the RTM have shown that the effects of modifying ω_{aer} and AOD on DRE should be additive.

On the other hand, increasing/decreasing the aerosol asymmetry parameter by 10 %, mostly affected the DRE_{TOA} (by about 30 %). DRE_{surf} is modified by about 10 % while DRE_{atm} is practically unaffected, as expected, given that changing g_{aer} does not affect the absorption ability of aerosol particles, it only modifies the angular distribution of scattered solar radiation.

Variations of aerosol optical properties by up to 10 %, as above examined, are probably realistic only for AOD. For example, AOD is found to have decreased over the Mediterranean basin by about 20 % from 2000 to 2007 (see e.g. Papadimas et al., 2008). On the contrary, similar changes of ω_{aer} and g_{aer}, associated with anthropogenic or natural variability, are quite difficult to take place on decadal timescales based on the observed range of values of these parameters accounting for their spatial and temporal variability; they rather represent the upper bound of possible natural or anthropogenic variability. Therefore, the sensitivity of model DREs to smaller changes of aerosol optical properties as well has been further examined. For this, AOD, ω_{aer} and g_{aer} have been modified by smaller amounts and the $\Delta(DRE_i)$s have been re-computed. Note that only one parameter has been changed for each simulation. Nevertheless, as indicated previously, the effects of changing the aerosol optical properties are additive. In Fig. 7, the computed $\Delta(DRE_i)$ values are plotted as function of changing aerosol properties (AOD, ω_{aer} and g_{aer}) by ± 1, ± 3, ± 5, ± 7 and ± 10 % for the 30 performed simulations. The results are given, again, in terms of regional means for the year 2003. Figure 7 offers the possibility to estimate the modification/dependence of aerosol DREs due to realistic changes of AOD, ω_{aer} and g_{aer}. It appears that aerosol DREs over the broader Mediterranean basin, depend almost linearly on changing aerosol optical properties (correlation coefficients of linear regression fits ranging from 0.989 to 1). The derived relationships (Fig. 7) enable an easy,

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first, estimate of the expected aerosol effects on the regional solar radiation budget, arising from potential changes of aerosol optical properties in various future climatic scenarios.

5 Conclusions

In this study, a detailed spectral radiative transfer model along with spectral aerosol optical properties from MODIS and other satellite and reanalysis datasets has been used to compute the direct radiative effect of natural plus anthropogenic aerosols on the solar radiation budget of the climatically sensitive Mediterranean basin. To our knowledge this is the first study that focuses on the entire Mediterranean and computes separately all aerosol effects, DREs, namely on the outgoing SW radiation at TOA, DRE_{TOA}, on solar atmospheric absorption, DRE_{atm}, and on the downward, DRE_{surf}, and absorbed SW radiation, $DRE_{netsurf}$, at surface. The model computations are performed under both all-sky and clear-sky conditions, using realistic data for all surface and atmospheric parameters, for the 7 yr period 2000–2007.

Aerosols are found to modify significantly the regional solar radiation budget. During the studied 7 yr period aerosols increase the SW reflection back to space above the broader Mediterranean basin by 2.4 Wm^{-2} and 4.5 Wm^{-2} under all- and clear-sky conditions, thus producing a planetary cooling over the region. The Mediterranean atmosphere is heated by aerosols due to enhanced solar absorption by 11.1 Wm^{-2} (14.3 Wm^{-2} in clear-skies) whereas the surface is cooled by aerosols through reduced reception at surface (16.5 Wm^{-2} all-sky, 22.9 Wm^{-2} clear-sky) and subsequent absorption (13.5 Wm^{-2} all-sky, 18.8 Wm^{-2} clear-sky). Even larger DREs are computed on a seasonal basis, reaching 26.2 and 31.7 Wm^{2} in case of all-sky and clear-sky DRE_{surf}, respectively. The maximum absolute DRE values are computed for summer (July) and the minimum ones for winter (December). However, under clear-sky conditions secondary DRE maxima appear in spring (April) in line with a corresponding spring AOD maximum. According to our results, the seasonal variation of aerosol direct radiative

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effects is driven by the available solar radiation fluxes under both clear- and all-sky conditions. The role of aerosol loading, i.e. AOD, is important mostly in cloudless skies.

High aerosol absolute DRE values, locally exceeding 100 Wm^{-2}, are computed and show a north-to-south gradient, with the highest values over the deserts of northern Africa and Middle-East. Also, relatively high values are calculated over central and northern Europe, associated with anthropogenic emissions of polluted aerosols. The larger DREs are computed over the eastern Mediterranean during winter and spring, and over the western Mediterranean during summer, whereas in autumn about equal DREs are computed in the eastern, western and central Mediterranean. The $2.5^{\circ} \times 2.5^{\circ}$ pixel-level DRE_{TOA} values range between −7 and 25 Wm^{-2}. Positive values, indicating planetary warming, are computed for over North Africa and Middle-East, where absorbing dust is trapping SW radiation. The same phenomenon, though much weaker, occurs over the Alps due to the presence of polluted aerosol over snow, throughout the year except summer. The geographical patterns of DREs in the atmosphere and at surface are similar, with the largest values over deserts extending from Sahara up to Middle-East and the Anatolian peninsula.

Aerosols over the Mediterranean basin have a strong absorbing character. This is reflected on the estimated regional values of the ratio of DREs at TOA (DRE_{TOA}) and at surface ($DRE_{netsurf}$) that vary between 0.09 and 0.27 throughout the year. Higher ratios $DRE_{TOA}/DRE_{netsurf}$ are found over the Mediterranean Sea (0.35–0.5) and the Atlantic Ocean (0.45–0.75) than over European and African continents (0.05–0.45). This indicates the presence of significantly scattering aerosols (sea-salt) over oceanic areas against more absorbing aerosols, natural or anthropogenic, above continental regions. Yet, the smaller ratios $DRE_{TOA}/DRE_{netsurf}$ above the Mediterranean Sea than the Atlantic Ocean point to the increased background of absorbing natural or/and anthropogenic aerosols (dust or/and black carbon) over the Mediterranean Sea. The computed DRE_{atm} highlights the existence of a significant aerosol effect on thermal dynamics over the Mediterranean basin. The strong surface SW cooling for the region, estimated equal to −13.5 Wm^{-2} on annual basis, combined with an equally strong

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warming of the atmospheric column (11 Wm^{-2}), can decrease the vertical atmospheric temperature lapse rate and thus create more stable atmospheric conditions, affecting the atmospheric circulation, cloud and precipitation formation. Such consequences can exacerbate the already documented (IPCC, 2007) threatening desertification processes occurring in the Mediterranean. The computed aerosol DREs are validated by thorough comparisons against reference measurements of SW fluxes from surface stations (Global Energy Balance Archive, GEBA).

Subject to equal percent modifications in aerosol properties, aerosol DRE is found to be mostly sensitive to aerosol single scattering albedo. Nevertheless, for realistic modifications of aerosol optical properties, i.e. within the range of observed natural or anthropogenic variability, AOD seems to be the main responsible parameter for modifications of aerosol radiative effects. Changes of DREs (ΔDREs) are found to depend quasi-linearly on the changes of aerosol optical properties (AOD, ω_{aer}, g_{aer}). The obtained relationships enable a first estimate of the expected benefits from actions aiming to reduce aerosol emissions (e.g. Air Framework Directive or Clean Air for Europe Programme). Future reductions of anthropogenic aerosol loadings in the Mediterranean basin of the order of 10 % that could result from such actions may reduce aerosol radiative effects up to about 10 %, producing a solar brightening mainly in summer, and hence surface heating, and a cooling of the atmospheric column owing to decreased aerosol solar absorption.

The model aerosol DREs presented in this study are columnar and do not account for the vertical distribution of aerosol optical properties. Such information over the Mediterranean is available from ground-based lidar measurements (EARLINET, Bösenberg et al., 2003) which however do not ensure the required complete spatial coverage in the present study. Vertically distributed aerosol information covering the entire Mediterranean basin has been made available recently from CALIOP instrument onboard the CALIPSO satellite (Winker et al., 2006). Nevertheless, this information is not yet extensively validated, it provides only AOD, while it is more limited spatially and temporally than MODIS, because of the relatively small swath of CALIOP (64 km × 64 km), inducing thus probably inadequate coverage for our study. The sensitivity of Mediterranean aerosol DREs to the vertical distribution of aerosol properties in the region will be examined in a future work, when aerosol vertical information will be available over the entire Mediterranean basin, for all properties, and at the required spatial and temporal resolution and coverage.

Supplementary material related to this article is available online at: http://www.atmos-chem-phys-discuss.net/11/30009/2011/ acpd-11-30009-2011-supplement.pdf.

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Table 1. Regional mean values of aerosol direct radiative effect (DRE, in Wm^{-2}) over the broader Mediterranean basin under all-sky conditions. DRE on outgoing solar radiation at TOA (DRE_{TOA}), on solar radiation absorbed in the atmosphere (DRE_{atm}), on downward surface solar radiation (DRE_{surf}), and on net downward (or absorbed) radiation at surface ($DRE_{surfnet}$). Seasonal mean values are given for winter (November-December-January, DJF), spring (March-April-May, MAM), summer (June-July-August, JJA), autumn (September-October-November, SON) and for the entire period (2000–2007). The values in parentheses indicate the corresponding quantities under clear-sky conditions.

	TOA (DRE_{TOA})	Atmosphere (DRE_{atm})	Surface DRE_{surf}	Surface $DRE_{netsurf}$
Year	−2.4 ± 0.3 (−4.5 ± 0.4)	11.1 ± 0.7 (14.3 ± 0.7)	−16.5 ± 1.0 (− 22.9 ± 1.2)	−13.5 ± 0.9 (−18.8 ± 1.0)
DJF	−0.9 ± 0.1 (−2.6 ± 0.1)	4.5 ± 2.2 (7.4 ± 3.3)	−7.0 ± 2.9 (−13.0 ± 4.6)	−5.4 ± 2.1 (−10.0 ± 3.4)
MAM	−2.4 ± 0.7 (−5.2 ± 0.9)	14.7 ± 2.7 (19.2 ± 2.9)	−20.9 ± 3.9 (− 29.7± 4.0)	−17.1 ± 3.2 (−24.4 ± 3.6)
JJA	−4.3 ± 0.5 (−6.3 ± 0.4)	17.5 ± 0.7 (20.1 ± 1.1)	−26.2 ± 1.0 (−31.7 ± 1.3)	−21.8 ± 0.9 (−26.5 ± 1.1)
SON	−1.9 ± 0.5 (−3.8 ± 0.5)	7.7 ± 4.0 (10.4 ± 4.0)	−11.8 ± 5.7 (−17.3 ± 5.6)	−9.6 ± 4.6 (−14.2 ± 4.4)

Table 2. Modifications $\Delta(\mathrm{DRE}_i)$ of reference model annual aerosol $\mathrm{DRE}_{\mathrm{ref},i}$ (in Wm^{-2} and %) for the broader Mediterranean region, arising from variations of AOD, single scattering albedo (ω_{aer}), and asymmetry parameter (g_{aer}), equal to ± 10 %. The reference aerosol $\mathrm{DRE}_{\mathrm{ref},i}$ (in Wm^{-2}) for the year 2003 are also given below.

	$\Delta(\mathrm{DRE})_{\mathrm{TOA}}$				$\Delta(\mathrm{DRE})_{\mathrm{atm}}$				$\Delta(\mathrm{DRE})_{\mathrm{surf}}$			
ΔV(%)	+10		−10		+10		−10		+10		−10	
	Wm^{-2}	[%]	Wm^{-2}	[%]	Wm^{-2}	[%]	Wm^{-2}	[%]	Wm^{-2}	[%]	Wm^{-2}	[%]
AOD	−0.22	8.4	0.22	−8.4	1.04	9.3	−1.05	−9.4	−1.52	9.0	1.54	−9.1
ω_{aer}	−0.99	37.9	1.12	−42.9	−8.22	−73.4	7.60	67.9	5.78	−34.2	−6.46	38.2
g_{aer}	0.79	−30.3	−0.76	29.3	−0.07	−0.6	0.07	0.6	1.64	−9.7	−1.57	9.3

$\mathrm{DRE}_{\mathrm{ref,TOA}}$	$\mathrm{DRE}_{\mathrm{ref,atm}}$	$\mathrm{DRE}_{\mathrm{ref,surf}}$
−2.6	11.2	−16.9

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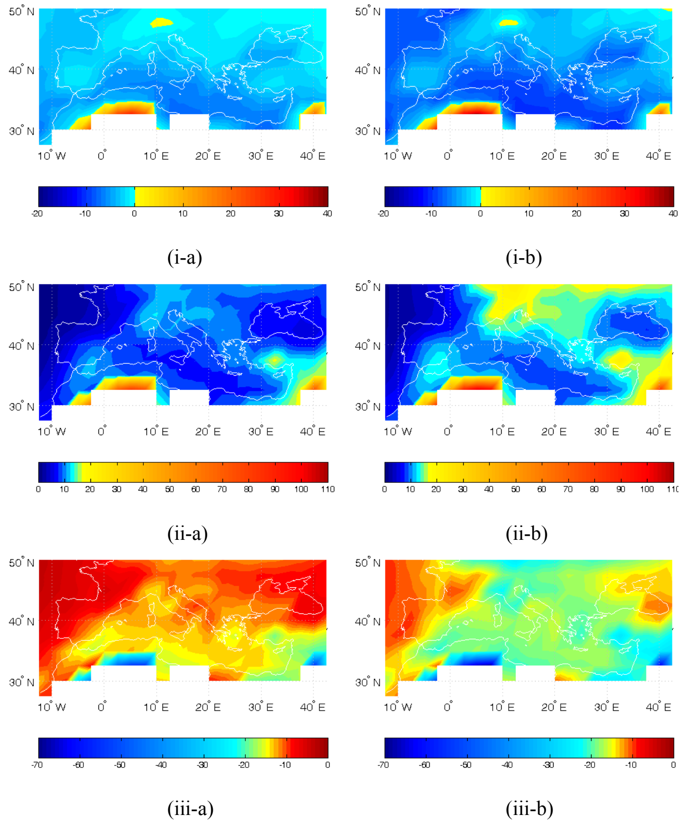

Fig. 1. Seven-year (2000–2007) average geographical distribution of aerosol direct radiative effect: **(i)** on the outgoing shortwave radiation at the top of the atmosphere (DRE_{TOA}, in Wm^{-2}), **(ii)** on the atmospheric absorption of shortwave radiation (DRE_{atm}, in Wm^{-2}), and **(iii)** on the absorbed solar radiation at the Earth's surface ($DRE_{netsurf}$, in Wm^{-2}) over the broader Mediterranean basin. The results are given under: **(a)** all-sky conditions and **(b)** clear-sky conditions.

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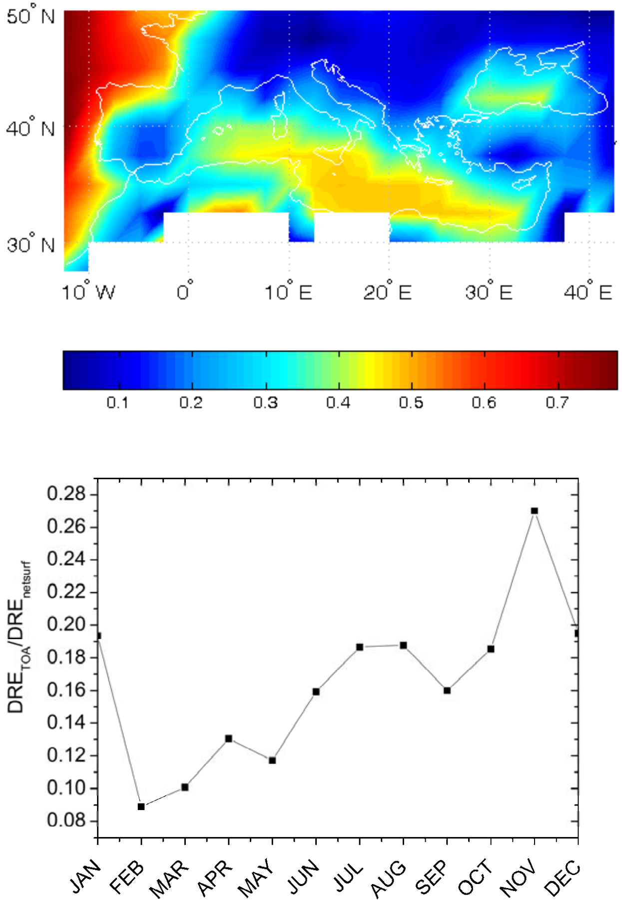

(i)

(ii)

Fig. 2. **(i)** Seven-year average (2000–2007) spatial distribution of the ratio $DRE_{TOA}/DRE_{netsurf}$ over the broader Mediterranean region, and **(ii)** seasonal variation of the 7 yr mean regional $DRE_{TOA}/DRE_{netsurf}$ values.

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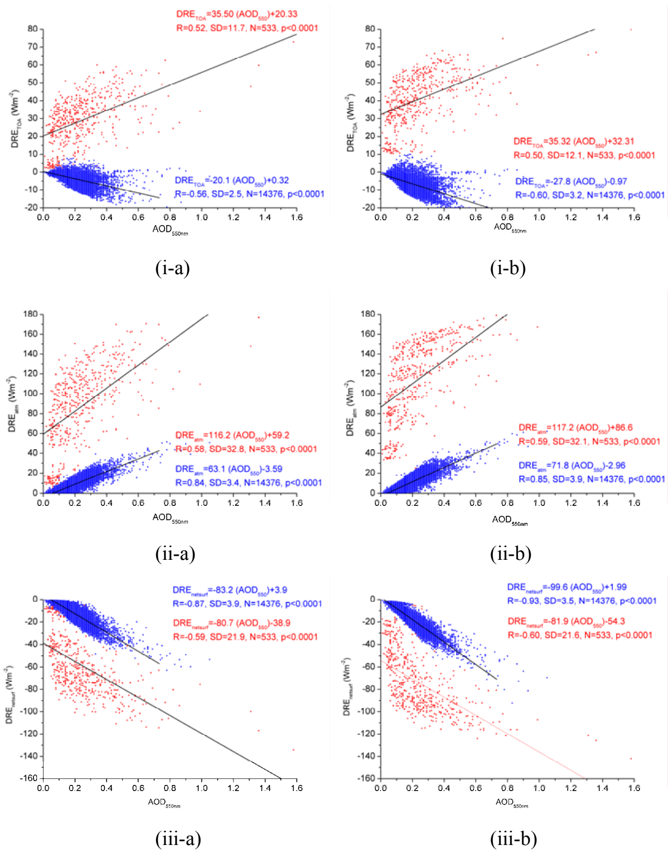

Fig. 3. Scatterplot comparison between DRE and AOD$_{550}$ values: **(i)** at TOA, **(ii)** in the atmosphere and **(iii)** at surface of the broader Mediterranean basin under: **(a)** all-sky and **(b)** clear-sky conditions. Applied linear regression fit lines are also shown, along with the computed slope, intercept and correlation coefficient values, and have been applied separately to the main blue (96 % of total number points) and secondary red (4 % of total number points) groups of points.

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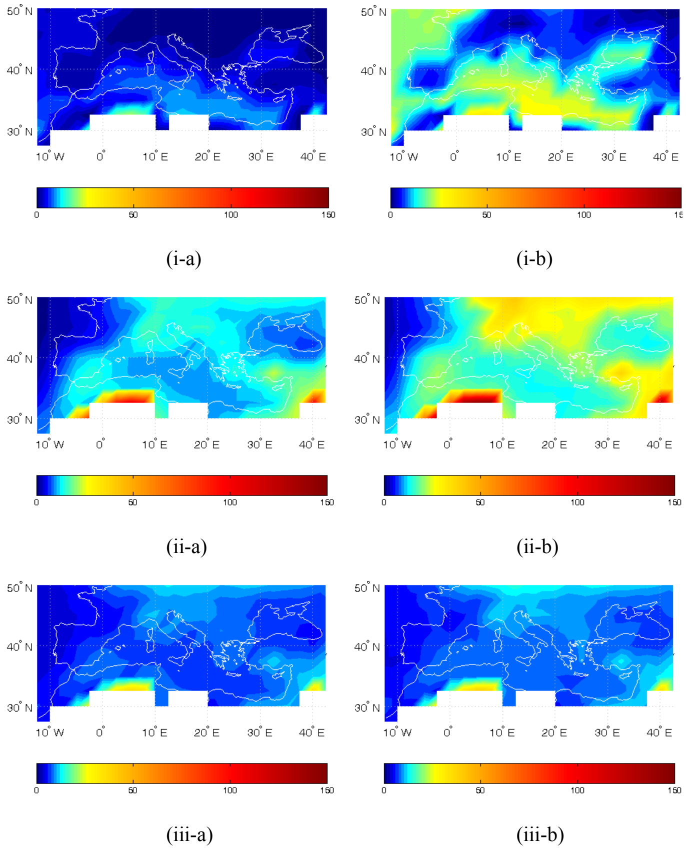

Fig. 4. Aerosol radiation budget efficiency, namely the percent change of solar radiative fluxes (in %) due to the presence of aerosols. Results are given at TOA **(i)**, in the atmosphere **(ii)**, and at surface **(iii)**. Results are given separately under: **(a)** all-sky, and **(b)** clear-sky conditions.

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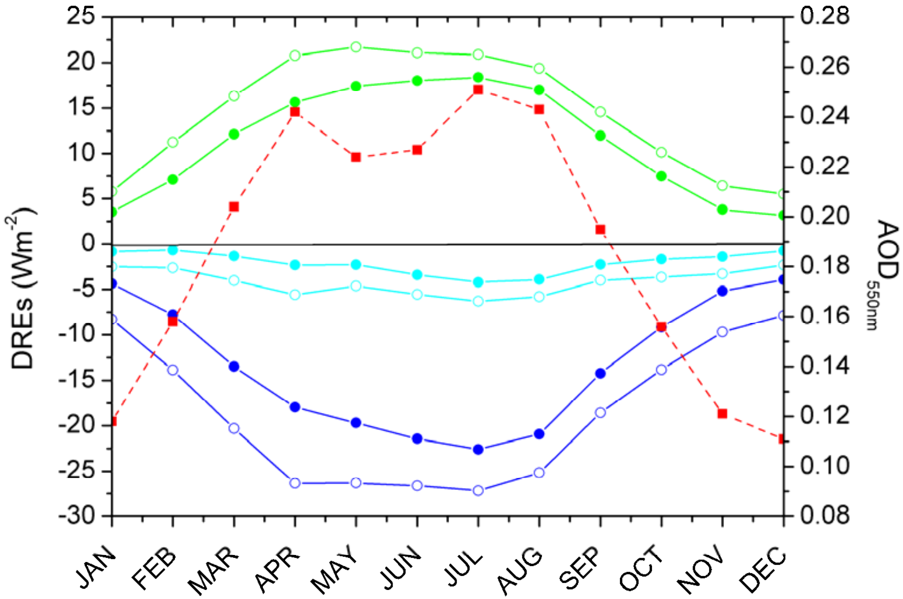

Fig. 5. Intra-annual variation of seven-year (2000–2007) regional mean values of aerosol DREs (at TOA: DRE_{TOA} cyan lines, in the atmosphere: DRE_{atm} green lines, at surface: $DRE_{netsurf}$ blue lines) under all-sky (full circles) and clear-sky (open circles) conditions. The intra-annual variation of visible AOD_{550} (dashed red line, rectangles) is also given (right y-axis).

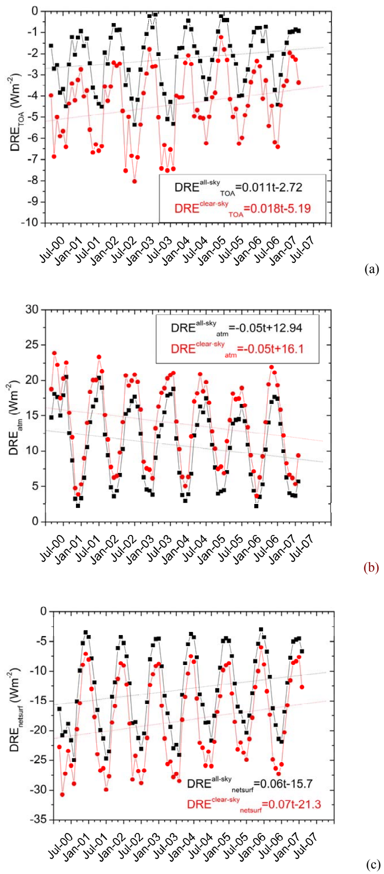

Fig. 6. Time series (2000–2007) of mean regional monthly values of: **(a)** DRE $_{TOA}$, **(b)** DRE$_{atm}$ and **(c)** DRE$_{netsurf}$ (in Wm^{-2}) for the broader Mediterranean basin, under all-sky (black line) and clear-sky (red line) conditions. Linear regression fit lines and the associated equations (t is the number of month starting from March 2000) are also shown.

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The direct effect of aerosols on solar radiation

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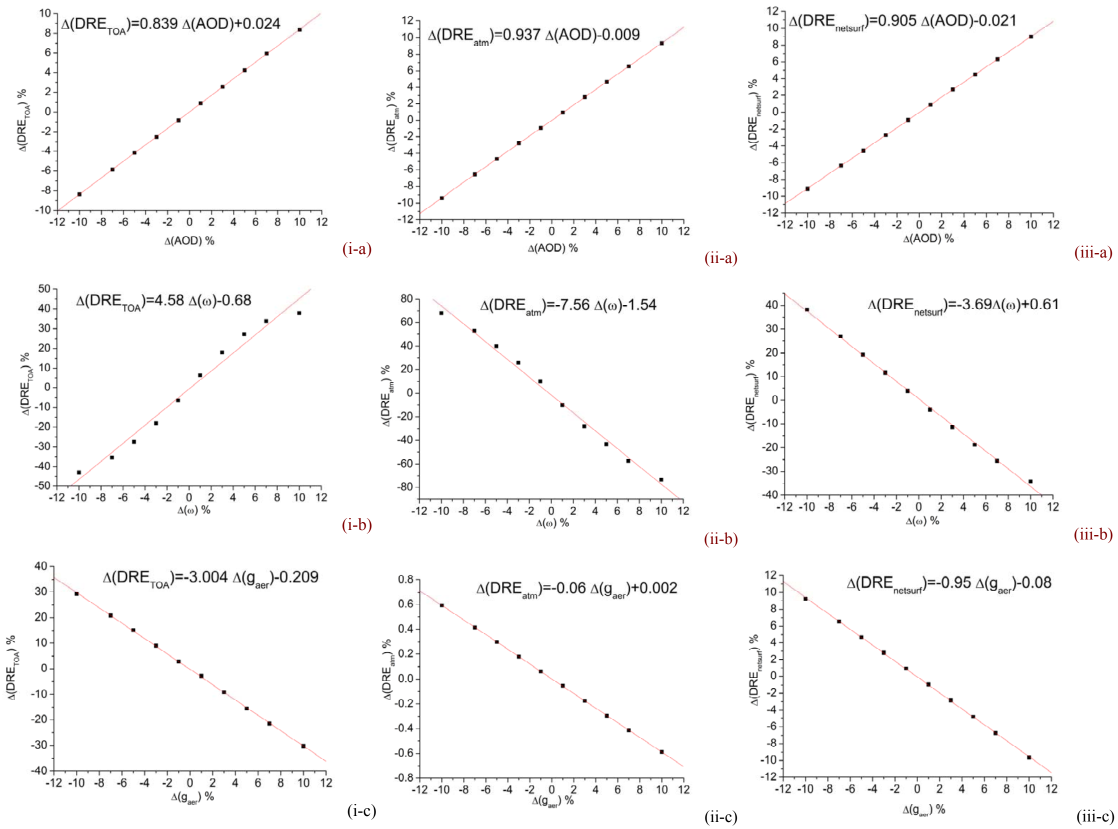

Fig. 7. Modifications of aerosol radiative effects: **(i)** at TOA, $\Delta(\mathrm{DRE_{TOA}})$, **(ii)** in the atmosphere, $\Delta(\mathrm{DRE_{atm}})$, and **(iii)** at surface, $\Delta(\mathrm{DRE_{surf}})$, as function of changing **(a)** AOD, **(b)** ω_{aer}, and **(c)** g_{aer}, by ± 1, ± 3, ± 5, ± 7 and ± 10 %. Results are given in terms of regional means for the year 2003.

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