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Sensitivity analysis of aerosol direct radiative forcing in ultraviolet–visible wavelengths and consequences for the heat budget

By N. HATZIANASTASSIOU[1]*, B. KATSOULIS[2] and I. VARDAVAS[3], [1]*Laboratory of Meteorology, Department of Physics, University of Ioannina, 45110 Ioannina, Greece, and Foundation for Research and Technology-Hellas, Heraklion, Crete, Greece;* [2]*Laboratory of Meteorology, Department of Physics, University of Ioannina, Ioannina, Greece;* [3]*Department of Physics, University of Crete, and Foundation for Research and Technology-Hellas, Heraklion, Crete, Greece*

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ABSTRACT

A series of sensitivity studies were performed with a spectral radiative transfer model using aerosol data from the Global Aerosol Data Set (GADS, data available at http://www.meteo.physik.uni-muenchen.de/strahlung/aerosol/aerosol.htm) in order to investigate and quantify the relative role of key climatic parameters on clear-sky ultraviolet–visible direct aerosol radiative forcing at the top of the atmosphere (TOA), within the atmosphere and at the Earth's surface. The model results show that relative humidity and aerosol single-scattering albedo are the most important climatic parameters that determine aerosol forcing at the TOA and at the Earth's surface and atmosphere, respectively. Relative humidity exerts a non-linear positive radiative effect, i.e. increasing humidity amplifies the magnitude of the forcing in the atmosphere and at the surface. Our model sensitivity studies show that increasing relative humidity by 10%, in relative terms, increases the aerosol forcing by factors of 1.42 at the TOA, 1.02 in the atmosphere and 1.17 at the surface. An increase in aerosol single-scattering albedo by 10%, in relative terms, increased the aerosol forcing at the TOA by 1.29, while it decreased the forcing in the atmosphere and at the surface by factors of 0.2 and 0.69, respectively. Our results show that an increase in relative humidity enhances the planetary cooling effect of aerosols (increased reflection of solar radiation to space) over oceans and low-albedo land areas, whilst over polar regions and highly reflecting land surfaces the warming effect of aerosols changes to a cooling effect. Thus, global warming and an associated increase in relative humidity would lead to enhanced aerosol cooling worldwide. The sensitivity results also demonstrate that an increase in surface albedo due to, for example, a reduction in land vegetation cover, would lead to enhanced atmospheric warming by aerosols leading to a reduction in cloud formation and enhancement of the desertification process. On the contrary, a decrease in surface albedo over polar regions due to, for example, ice-melting associated with global warming, would reduce the planetary warming effect of aerosols over polar areas. Aerosol forcing is found to be quite sensitive to cloud cover, as well as to aerosol optical thickness and the asymmetry parameter, and to the wavelength dependence of the aerosol optical properties.

1. Introduction

There is strong evidence that the Earth's climate is being changed. Changes in climate are the result of both internal feedback processes in the climate system and external factors (natural and anthropogenic), whose influence is expressed by the concept of radiative forcing. Apart from other factors (like clouds, trace gases and solar irradiance) aerosols, both natural and anthropogenic, constitute a major factor leading to changes in radiative forcing. It has been recognized that aerosols affect the Earth's radiation budget both directly and indirectly, inducing an average global negative radiative forcing (a cooling effect) which may counteract global warming (positive forcing due to increases of the well-mixed greenhouse gases), estimated to be 2.43 W m^{-2} (IPCC, 2001) since the beginning of 19th century. Aerosols exhibit strong temporal and spatial variability, producing globally uneven patterns of radiative forcing (compared with greenhouse gases). Besides, there is a large uncertainty in the crucial aerosol radiative parameters (e.g. optical thickness and single-scattering albedo), which along with the small residence time of atmospheric aerosols (a few days to weeks) and their

*Corresponding author.
e-mail: nhatzian@cc.uoi.gr

variable concentrations, makes it difficult to characterize aerosols as climate-forcing agents and quantify their radiative forcing. Although significant progress has been achieved to better characterize the direct aerosol forcing of different types of aerosols (e.g. sulfate but also carbonaceous or organics), climate forcing due to aerosols remains one of the largest uncertainties in climate variability and climate change studies (Hansen et al., 1997; Tegen et al., 2000; Harvey, 2000). The level of scientific understanding relevant to direct forcing is still very low, thus producing large uncertainties. The range of estimates, guided by the spread in the published forcing values, is comparable to or even larger than their magnitude (IPCC, 2001). Much work and many modelling studies are required in order to improve our understanding of aerosol forcing mechanisms.

Hatzianastassiou et al. (2004) presented a model for estimating the global distribution of direct aerosol forcings in the ultraviolet–visible (UV–VIS) range of wavelengths (0.2–0.85 μm) at the top of the atmosphere (TOA), within the atmosphere and at the Earth's surface, arising under clear skies. The model accounts for the effect of relative humidity (RH) on the aerosol optical properties by computing the extinction aerosol optical thickness (AOT), single-scattering albedo (ω_{aer}) and asymmetry parameter (g_{aer}), which were taken originally from the Global Aerosol Data Set (GADS, Koepke et al., 1997), for computed mean monthly values of ambient RH within the aerosol layer. The values for scattering and absorption aerosol optical thickness are derived from the AOT and ω_{aer}. These computations were performed, as explained by Hatzianastassiou et al. (2004), by using water vapour and vertical temperature profiles from the National Centers for Environmental Prediction and National Center for Atmospheric Research (NCEP/NCAR) Global Reanalysis Project. The aerosol forcings were computed for actual clear-sky fractions, based on cloud cover data taken from the International Satellite Cloud Climatology Project (ISCCP) D2 series (Rossow and Schiffer, 1999). A complete description of the model and input data is given in detail in Hatzianastassiou et al. (2004).

Evaluation of the effects of various key climatic parameters on aerosol forcing is very important when attempting to draw conclusions, especially given the uncertainties related to these parameters used as input data for models. The radiatively important aerosol properties are determined at the most fundamental level by the aerosol composition and size distribution. However, for the purposes of direct forcing calculations and for assessment of uncertainties, aerosol-related or other surface and atmospheric properties can be subsumed into a number of parameters. Such parameters are the mass light-scattering efficiency, the dependence of light scattering and absorption on RH, the single-scattering albedo, the asymmetry parameter, the non-sphericity and the nature of the external mixture of aerosols, the particle composition, the albedo of the underlying surface, the cloud fraction, the vertical distribution of aerosol and cloud, the aerosol loading (optical depth), the solar zenith angle, the size distribution and the height of the boundary layer. The climatic role of some aerosol properties, surface and atmospheric parameters for aerosol radiative forcing is investigated in this paper. Even though there have been many studies on short-wave aerosol forcing (e.g. Koepke and Hess, 1988; Charlson et al., 1992; Kiehl and Briegleb, 1993; Penner et al., 1994; Boucher and Anderson, 1995; Nemesure et al., 1995; Hansen et al., 1997; Haywood et al., 1997; Haywood and Shine, 1997; Schult et al., 1997; West et al., 1998; Penner et al., 1998; Haywood and Ramaswamy, 1998; Myhre et al., 1998; Kay and Box, 2000; Ramanathan et al., 2001; Wendisch et al., 2001), they all agree that the magnitude of forcing is still uncertain, with the main cause of uncertainty being the aerosol properties used as input to the models.

In this study, the relative role of key climatic parameters on direct aerosol forcing is investigated through a series of sensitivity tests. With this aim, each parameter is modified and the resulting aerosol forcing is compared with that corresponding to a reference case with specific parameter values. The parameters examined are: RH, surface albedo (R_g), cloud cover (A_c), AOT, ω_{aer} and g_{aer}, as well as ozone (O_3) concentration, incoming solar radiation and the wavelength dependence of aerosol optical properties. The importance of each parameter was evaluated through comparison of the sign and magnitude of induced changes on aerosol forcing at the TOA, within the atmosphere and at the surface. Furthermore, possible climatic feedback, due to the presence of aerosols in the atmosphere, is assessed and discussed, based on the results of the sensitivity tests.

2. Methodology

A series of sensitivity tests were performed to investigate the relative role of the various key climatic parameters that determine the aerosol UV–VIS radiative forcing $\Delta F = F_i - F_{\text{clear},i}$ (where F_i and $F_{\text{clear},i}$ are the solar radiative fluxes with and without the presence of aerosols, respectively, Hatzianastassiou et al., 2004), namely: the aerosol-induced changes in the outgoing solar radiation at the TOA (ΔF_{TOA}), the solar atmospheric absorption (ΔF_{atmoab}) and the downward solar radiation at the surface (ΔF_{soldn}). The results of the sensitivity tests are given in terms of absolute and percentage (eqs. 1 and 2, respectively) changes, $\Delta\Delta F$ and $\Delta\Delta F(\%)$ respectively, in the aerosol radiative forcings as

$$\Delta\Delta F_i = \Delta F_i - \Delta F_{\text{ref},i} \tag{1}$$

$$\Delta\Delta F_i\% = \frac{\Delta F_i - \Delta F_{\text{ref},i}}{\Delta F_{\text{ref},i}} \cdot 100 \tag{2}$$

where $\Delta F_{\text{ref},i}$ are the aerosol radiative forcings for the reference case study and ΔF_i are the corresponding forcings for each sensitivity study, where i refers to the various aerosol forcings defined above.

The forcing computations were performed on a mean monthly basis and 2.5° × 2.5° longitude–latitude resolution, using a

deterministic spectral radiative transfer model described in detail by Hatzianastassiou et al. (2004). The model computes theoretically the incoming solar flux at the TOA at 115 wavelengths ranging from 0.20 μm to 0.85 μm. For each wavelength, a set of monochromatic radiative flux transfer equations is solved for an absorbing/multiple-scattering atmosphere, using the delta-Eddington method of Joseph et al. (1976) which is an extension of the Eddington method described in Shettle and Weinmann (1970). The atmosphere is divided into three layers allowing for scattering and absorption of aerosol particles, atmospheric molecular absorption and Rayleigh scattering. Reflection of incident solar radiation from the Earth's surface is accounted for in the model, while the dependence of aerosol optical properties on RH is taken into account in GADS (Shettle and Fenn, 1979; D'Almeida et al., 1991; Koepke et al., 1997) and in our model (Hatzianastassiou et al., 2004). The aerosol radiative forcing is computed by running the radiative transfer model with and without aerosols and the forcings are subsequently averaged over the UV–VIS range by weighting with the spectrally resolved incoming solar radiation. A full description of the model and input data is given in Hatzianastassiou et al. (2004).

3. Model sensitivity studies

3.1. Reference forcings

The global distribution of model computed UV–VIS aerosol forcings for the reference case are shown in Fig. 1 for January (a) and July (b) respectively (white areas in the figures correspond to areas with no sunlight during local winter or to missing ISCCP-D2 data). For brevity, the term "solar radiation" is used throughout the study, meaning the UV–VIS range of wavelengths. Reference forcings were taken to be the global distribution of aerosol forcings for the year 1992, computed by using realistic input data (RH, cloud cover, surface albedo, incoming solar radiation, atmospheric molecules) as described in detail by Hatzianastassiou et al. (2004). In this reference case, we used aerosol optical properties (AOT, ω_{aer}, g_{aer}) originally taken from GADS, given at 11 wavelengths ranging from 0.25 to 0.8 μm (UV–VIS) and at eight classes of RH ranging from 0% to 99%, that were used to calculate the aerosol properties for actual values of RH for the year 1992, based on NCEP/NCAR humidity data.

The effect of aerosols on the outgoing solar radiation at the TOA (ΔF_{TOA}, case i), on the solar atmospheric absorption (ΔF_{atmoab}, case ii) and on the downward solar radiation at the surface (ΔF_{soldn}, case iii) are given separately. Aerosols cause a cooling ($\Delta F_{TOA} > 0$) over oceans (low surface albedo) and a warming ($\Delta F_{TOA} < 0$) over deserts or ice (high surface albedo). They can modify the planetary reflection of solar radiation by up to 10 W m^{-2}. They can produce a planetary warming through absorption of solar radiation by mineral components over deserts (e.g. the Sahara, Arabia) or soot particles over continental areas (e.g. Europe, North Asia, North America). The importance of the reflectance of the surface underneath the aerosol layer, as well as of the aerosol particle composition (reflected in ω_{aer}), is shown by the computed negative ΔF_{TOA} values (planetary warming) over Antarctica and Greenland (Arctic haze) as well as by negative values over Siberia in winter, which are changed to positive values (cooling) in summer. The atmosphere is found to be heated by aerosols by as much as 25 W m^{-2}, with the highest values over areas characterized by strongly absorbing mineral particles and high surface albedo (e.g. the Sahara). Quite large values are also found over Europe, North America, South and Southeast Asia, the Amazon basin and Australia. The downward solar radiation at the surface is drastically reduced due to the presence of aerosols by up to about 30 W m^{-2}. The largest decreases are found over regions with high aerosol loadings (AOT values); therefore, ΔF_{soldn} values are generally larger over continents than oceans. A thorough discussion of patterns of spatial and seasonal variation of aerosol forcings is given in Hatzianastassiou et al. (2004).

3.2. Relative humidity

The RH is the single most important parameter that determines the direct aerosol forcing, since the increase in aerosol mass as a result of water uptake is the most important driving process (Pruppacher and Klett, 1997). RH significantly affects AOT, ω_{aer}, and g_{aer} by modifying the aerosol liquid water content, size and hence extinction coefficient and refractive indices. The effect of changing RH on aerosol forcing is a function of the size and number distribution, total concentration and composition of the aerosol particle spectrum, which are highly variable. The strong effect of RH on the microphysical, optical and radiative properties of the aerosol has also been reported by other investigators (e.g. Pilinis et al., 1996; Koepke et al., 1997; Penner et al., 1998; West et al., 1998; Kondratyev, 1999).

To examine the effect of RH on aerosol radiative forcing, the computed RH within the tropospheric aerosol layer, for the reference case, was decreased or increased by $\pm$10%, in relative terms, within the limits 0% and 90%, to avoid pre-condensation conditions, and the computed aerosol forcings were compared with those of the reference case (see Fig. 1). The sensitivities of the aerosol forcing are expressed here in terms of percentage modification of the reference forcing values, and they have to be combined with those forcing values to yield absolute forcing changes (in W m^{-2}). The changes in aerosol forcings, $\Delta\Delta F_i$, produced by changing the RH, were found to increase in a non-linear way with increasing RH values, as also reported by other investigators (e.g. Box and Trautmann, 1994; Pilinis et al., 1996; Haywood and Shine, 1997; Haywood et al., 1997, 1998; Penner et al., 1998). Increasing the RH by a certain relative percentage has in general the opposite effect on aerosol forcings to decreasing RH by the same amount, but with a larger magnitude. Increasing the RH by 10% of its reference value strongly modifies

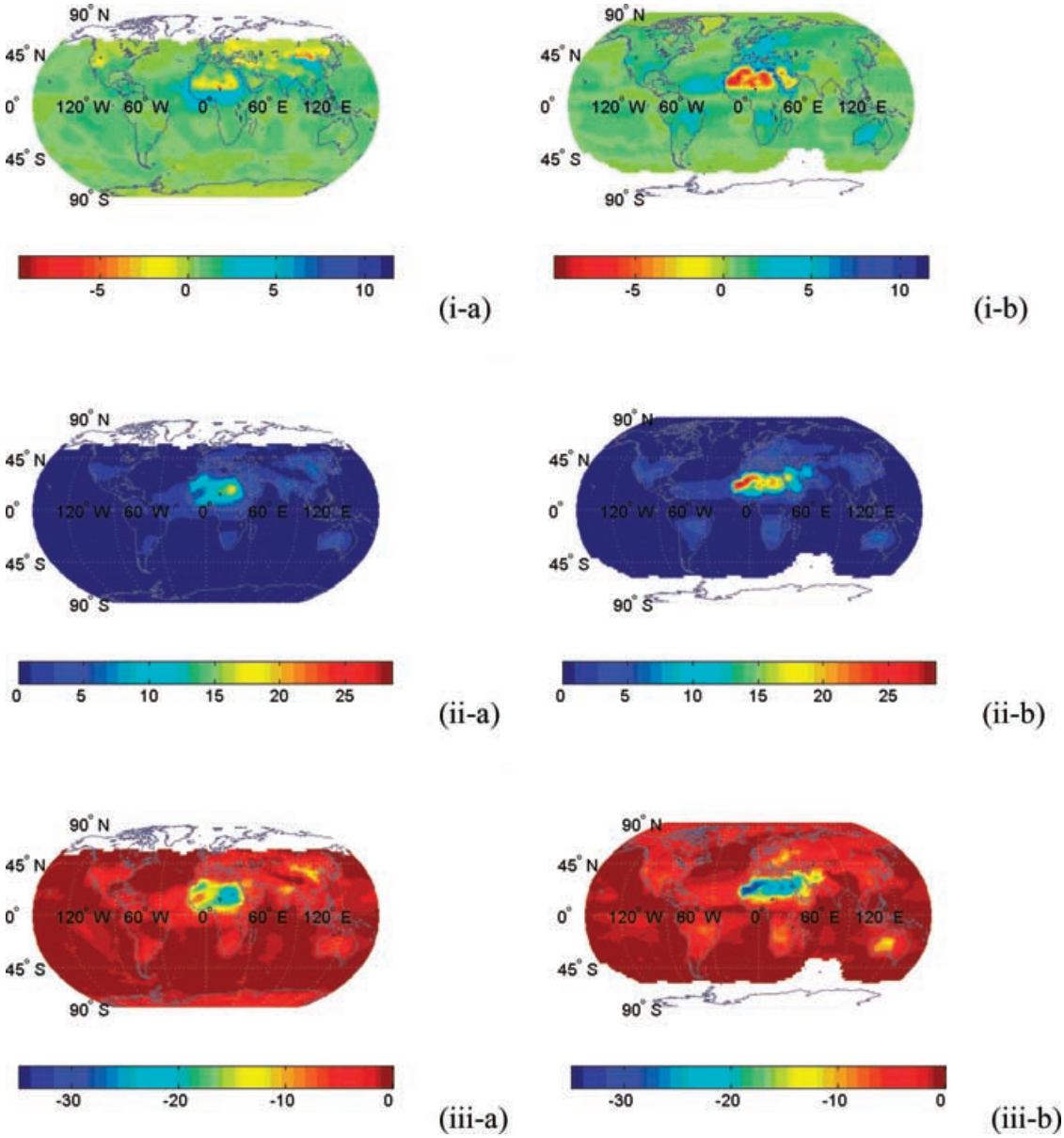

Fig 1. Global distribution of the reference case ultraviolet–visible (UV–VIS) aerosol radiative forcings (ΔF in W m^{-2}) computed using aerosol optical thickness, single-scattering albedo and the asymmetry parameter from the Global Aerosol Data Set for actual climatic conditions (cloud cover, surface albedo, RH) for (a) January, and (b) July of the year 1992: (i) Aerosol forcing on the outgoing UV–VIS solar radiation at TOA (ΔF_{TOA}), (ii) forcing on the absorption of UV–VIS solar radiation within the atmosphere ($\Delta F_{\mathrm{atmoab}}$) and (iii) forcing on the downward UV–VIS solar radiation at the surface ($\Delta F_{\mathrm{soldn}}$).

the aerosol forcing, as shown in Figs. 2a and b. The computed changes in forcing $\Delta\Delta F_i$, expressed as percentage changes of the reference forcings given in Fig. 1, are as large as ±200–300% for ΔF_{TOA}, larger than 150% for $\Delta F_{\mathrm{atmoab}}$ over extended regions, while they are equal up to about 50% for $\Delta F_{\mathrm{soldn}}$. In general, increasing the RH values increases (amplifies) the aerosol forcings in absolute terms (increase in magnitude regardless of sign). The increase in RH increases the outgoing solar radiation over oceans through increased aerosol scattering from about 1 W m^{-2} up to 2 W m^{-2}. Over highly reflecting polar regions the warming effect, i.e. reduced outgoing radiation, changes to a cooling effect. There is also amplification of the cooling of the ocean surface due to aerosols (cf. Figs. 2iii-a and iii-b). Thus, global warming and an associated increase in RH would lead to enhanced worldwide aerosol-induced radiative cooling which, however, might be overwhelmed by other effects such as convection and cloud radiative effects or infrared heating which are not considered here.

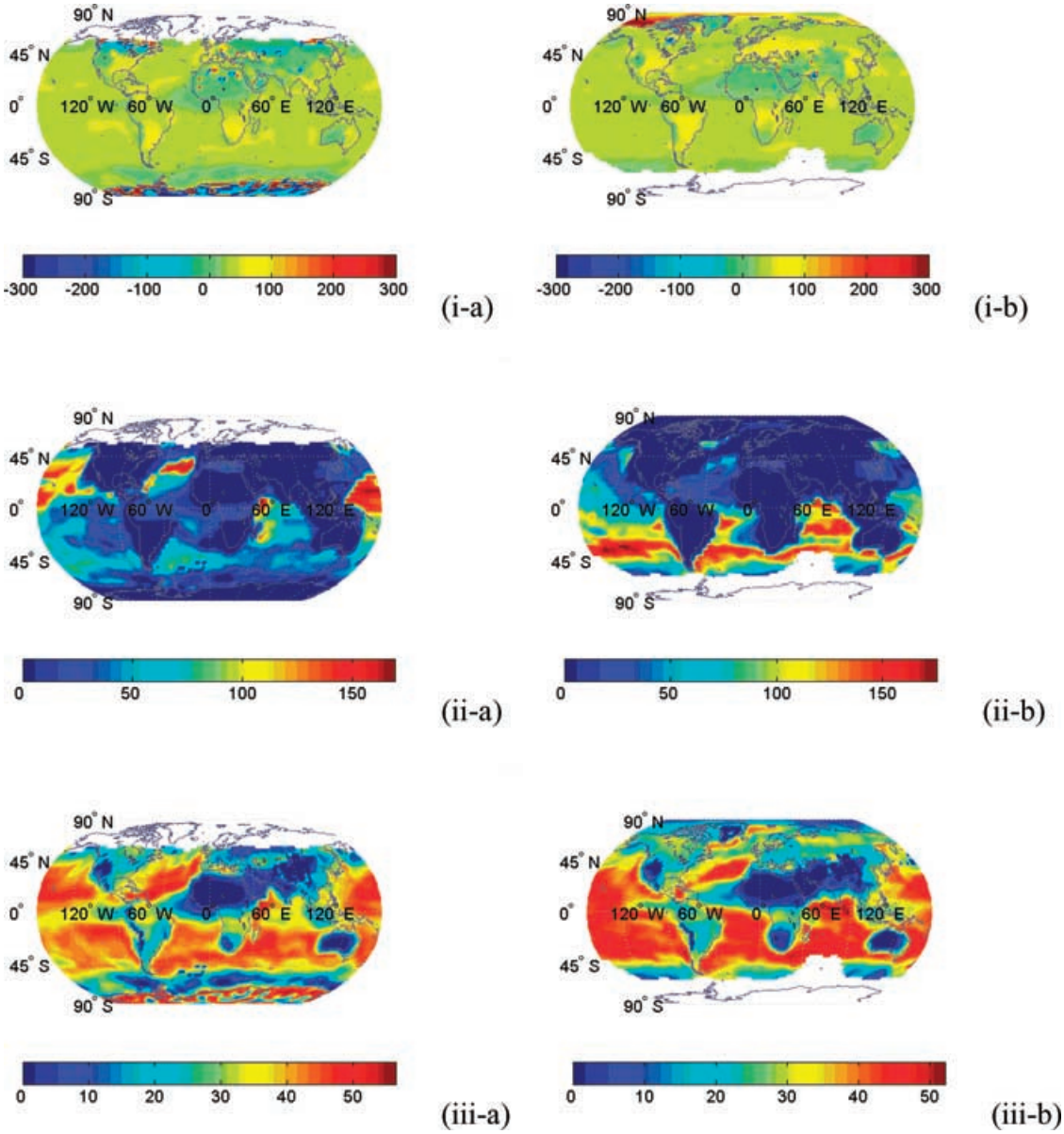

Fig 2. Global distribution of percentage differences ($\Delta\Delta F$ in %) in the reference aerosol forcings (Fig. 1) induced by increasing the RH by 10% of its reference value for (a) January and (b) July. The aerosol forcings are explained in Fig. 1.

The sensitivity of aerosol radiative forcing to changing RH differs from one region to another due to differences in AOT values, as a function of RH, which are in turn attributed to the different capacities of aerosols for water uptake (Flossmann, 1998) due to their differing composition, number concentration and size and number distribution of aerosol spectra. The differences from one region to another are also due to differences in the reference RH conditions in the aerosol layer, which range roughly between 20% and 90% worldwide, given that aerosol properties are mostly sensitive to higher humidity values. Thus, there are some regions (e.g. North Africa, Australia, Eurasia, North America) which show a systematically smaller sensitivity of aerosol forcing to changes in RH than others (e.g. the Indian Ocean, the windy oceanic zone of the Southern Hemisphere, the Pacific Ocean). Nevertheless, note that large forcing sensitivities expressed in percentage values do not necessarily involve large forcing modifications in absolute values. Thus, quite large absolute changes (in W m^{-2}) in aerosol forcings are found over areas such as Europe, the Mediterranean Basin, Central Africa, Central and South America, the Gobi desert or the Western Pacific.

It should be noted that changing the RH significantly affects AOT, ω_{aer} and g_{aer}, which is the case in this sensitivity test. For example, an increase in RH results in an increase in particle size, and hence in more forward scattering associated with higher g_{aer}

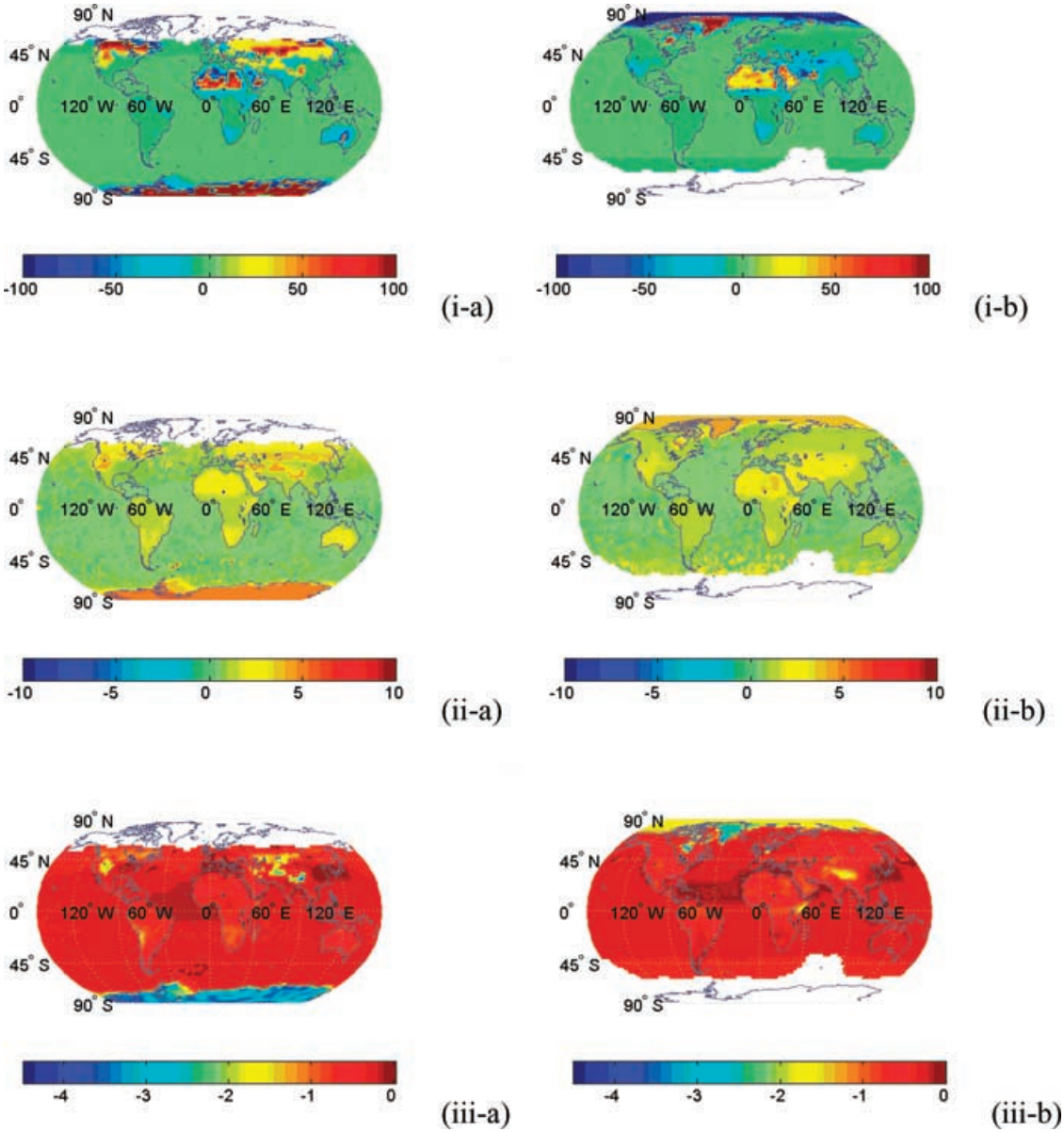

Fig 3. Global distribution of percentage differences ($\Delta\Delta F$ in %) in the reference aerosol forcings (Fig. 1) induced by increasing the surface albedo by 10% for (a) January and (b) July. The aerosol forcings ΔF are explained in Fig. 1.

values, as shown in the GADS data. Thus, the sensitivity tests that are presented in Sections 3.5 and 3.6, which deal with the effects of modified AOT, ω_{aer} and g_{aer} conditions on the aerosol forcing, for example due to anthropogenic or natural activity, are independent single assumptions, and part of their effect is shown in this sensitivity test.

3.3. Surface albedo

To examine the effect of surface albedo, R_g, on aerosol radiative forcing R_g was increased by 10%, in relative terms, up to a limit of 100%. This decreased the magnitude of aerosol forcing at the TOA by up to 100% (e.g. in Arctic summer, Fig. 3(i-b)), but in some cases it increased the forcing magnitude by up to 100% (e.g. over Antarctica in summer, Fig. 3(i-a)). Increasing R_g by 10% resulted in a general increase of aerosol forcing ΔF_{atmoab} by up to 10% (enhanced atmospheric warming), while it decreased the aerosol forcing at the surface ΔF_{soldn} (i.e. less surface cooling) by less than 5%. Therefore, aerosol forcing at the TOA is found to be more sensitive to changes in R_g, especially over regions with high surface albedo, than the forcing within the atmosphere or at the surface. Over Antarctica and the Arctic there is enhanced planetary warming due to aerosols under conditions of increased surface albedo, arising from enhanced

multiple scattering between the aerosol layer and the surface. According to our model sensitivity test, there is enhanced aerosol-induced planetary warming over the Sahara if the surface albedo is increased. This would enhance desertification of the surrounding regions. The model sensitivity demonstrates that, over areas threatened by desertification, an increase in surface albedo due to a reduction in land vegetation cover would lead to enhanced atmospheric warming and surface cooling due to aerosols, resulting in a reduction in cloud formation (Kaufman et al., 2002) and an enhancement of the desertification process. The results of the sensitivity test given here assume an increase in surface albedo. Nevertheless, sensitivity tests were also performed, assuming decreasing surface albedo values, and their results have shown that the effect of decreasing R_g is equivalent, but opposite in sign, to that of increasing R_g by the same amount (e.g. 10% in relative values). Thus, a decrease in surface albedo over polar areas due to, for example, ice-melting associated with global warming would induce decreased planetary warming over polar areas due to aerosols.

3.4. Cloud cover

The effect of cloud cover, A_c, was examined by decreasing the total A_c by 5%, in relative values, to a limit of 0%. In general this increased the magnitude of aerosol forcing, ΔF_i, by up to 100% locally, as expected (Figs. 4i, ii, iii), since this implies an increased clear-sky fraction and hence clear-sky direct aerosol forcing at the TOA, within the atmosphere and at the surface. Thus, for decreased cloud cover there is a larger clear-sky effect of aerosols on the outgoing solar radiation at the TOA (higher planetary cooling), and more atmospheric warming and surface cooling, i.e. reduced downward solar radiation at the surface. This mechanism can exacerbate desertification processes in aerosol-polluted semi-arid regions, like the Mediterranean Basin, through reduced evaporation and precipitation. Nevertheless, other conditions must also be taken into account, and they have to be combined in order to draw definite conclusions on climatic issues. For example, increased RH would lead to more cloud, with opposite effects to the current sensitivity test. Thus, in a sensitivity test, increasing the total A_c by 5%, with a limit of 100%, was found to produce an equivalent, but opposite in sign, effect to decreasing A_c by 5%, i.e. a decrease in magnitude of aerosol forcing.

3.5. Aerosol optical thickness

Increasing the extinction aerosol optical thickness (AOT) by 10% in relative terms generally increased the magnitude of aerosol forcing at the TOA by up to 30% (Fig. 5-i), thus producing enhanced planetary cooling or warming at the TOA. Thus, over areas characterized by aerosol-induced planetary warming (e.g. the Sahara, the Antarctic plateau, Siberia in winter), the magnitude of ΔF_{TOA} is found to increase by up to 30%. This involves a further decrease in outgoing solar radiation at the TOA in these regions compared with the reference case, i.e. the warming effect of aerosols is found to be amplified, which might seem initially paradoxical since an increase in AOT is usually thought to induce more light scattered upwards. However, for a fixed ω_{aer}, increased extinction AOT involves a corresponding increase in aerosol scattering and absorption optical thickness values, which in the case of strongly absorbing aerosols over highly reflecting surfaces underneath can result in an even stronger aerosol warming effect at the TOA. There are some limited areas over which the forcing ΔF_{TOA} changes sign, for example over the Antarctic coast in January or over the Beaufort Sea in July. This indicates that the sign of aerosol forcing is determined not only by the aerosol composition (particle absorptivity) and the albedo of the surface underneath, but also by the amount of aerosol load. Increasing AOT by 10% amplified both the solar atmospheric absorption due to aerosols and the reduction in the downward solar surface radiation due to scattering. Over the Sahara, the 30 W m^{-2} decrease in downwelling flux due to aerosols was increased by 3 W m^{-2}. In Antarctica, the aerosol-induced reduction in downwelling flux by 3 W m^{-2} was increased by 0.5 W m^{-2}. In general, the dependence of aerosol forcing on AOT was found to be linear. It should be noted that, in our sensitivity studies, the effect of modified aerosol optical properties (AOT, ω_{aer}, g_{aer}) on the aerosol forcing was attributed to the total aerosol population. Nevertheless, it is possible to obtain the modification of forcing by anthropogenic aerosols separately, by using the GADS data.

3.6. Aerosol single-scattering albedo and asymmetry parameter

The largest effect of increasing ω_{aer} by 10% (with an upper limit of 1) is found to be on ΔF_{atmoab} (Fig. 6), as expected, since it corresponds to decreased particle absorption. The forcing ΔF_{atmoab} decreased through latitudes and seasons, since increased ω_{aer} values involve smaller particle absorption, leading to less absorption of solar radiation within the atmosphere, ranging mainly from 60% to 100%. The forcing ΔF_{TOA} values increased (more planetary cooling) by up to 100%. In areas with planetary warming in the reference case, such as the Sahara, Antarctica and northern mid-latitudes in winter, increasing ω_{aer} decreased the magnitude of this warming by up to 100%. The forcing ΔF_{soldn} is found to decrease in magnitude, i.e. the downward solar radiation at the surface increases, because of decreased solar atmospheric absorption. The larger changes in ΔF_{TOA}, ΔF_{atmoab}, and ΔF_{soldn} are found over continental rather than maritime regions.

An increase in g_{aer} by 10%, corresponding to a decrease/increase in aerosol backward/forward scattering, generally decreased the magnitude of aerosol forcing (Fig. 7). Over oceans (low surface albedo) this leads to a decrease in outgoing radiation at the TOA (planetary warming) by as much as 30%, as more forward scattering results in higher ocean absorption. Above polar regions or desert areas (high surface albedo),

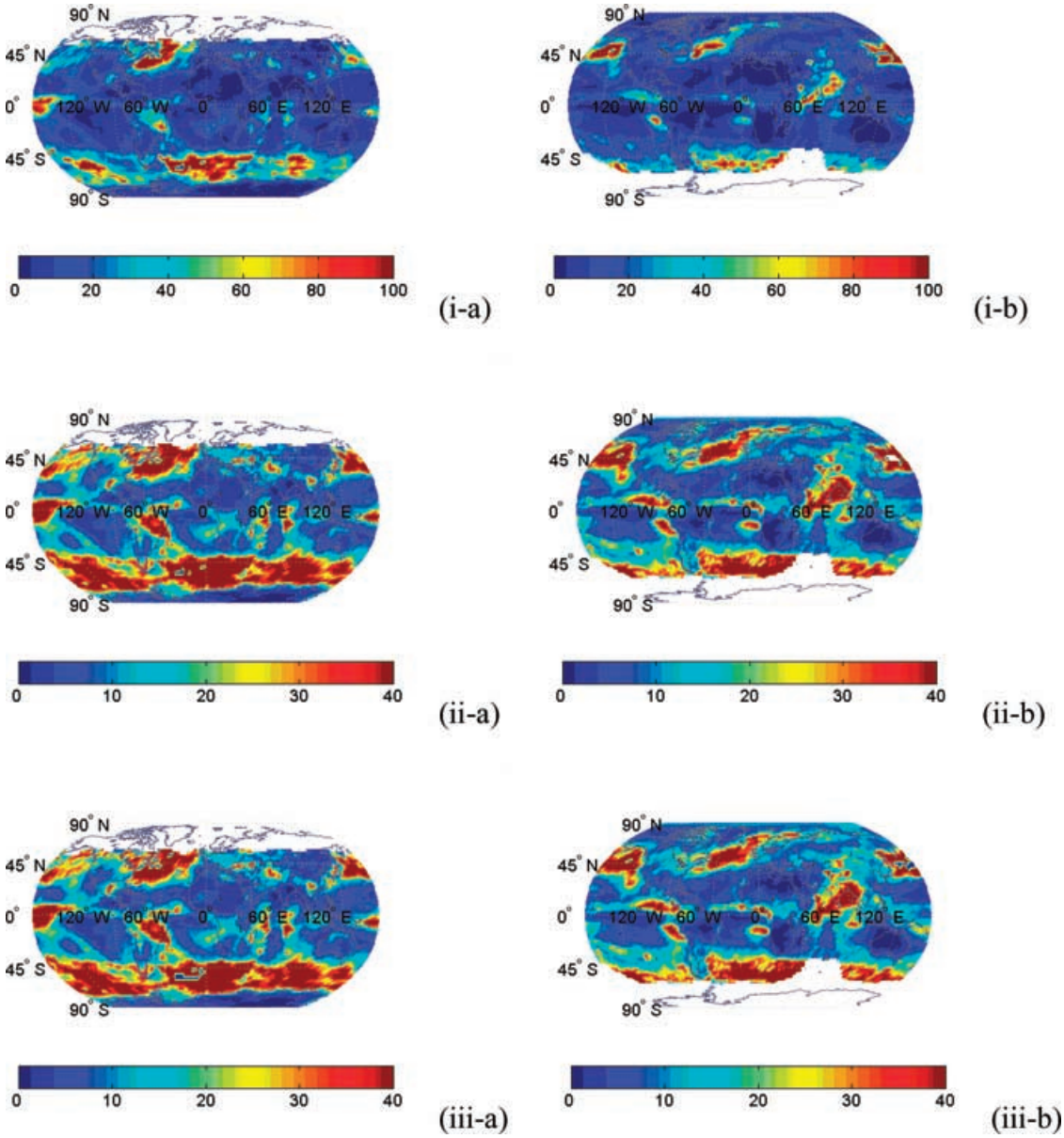

Fig 4. Global distribution of percentage differences ($\Delta\Delta F$ in %) in the reference aerosol forcings (Fig. 1) induced by decreasing the cloud cover by 5% for (a) January and (b) July. The aerosol forcings ΔF are explained in Fig. 1.

planetary warming is enhanced by more than 50% because of increased surface absorption. The magnitude of forcings ΔF_{atmoab} and ΔF_{soldn} generally decreased by up to 10% and 25%, respectively. Nevertheless, an increase in the magnitude of ΔF_{atmoab}, for a g_{aer} increased by 10%, is found over areas of high surface albedo such as polar regions, or areas which are characterized by significantly absorbing aerosols.

3.7. Other parameters

The sensitivity of aerosol forcing to ozone (O_3) concentration and to incoming solar radiation was also investigated by increasing the concentration of O_3 by 10%, and the incoming UV–VIS radiation by 1%, in relative terms. In both cases the magnitude of aerosol forcing was found to change within 5%. Only the forcing ΔF_{atmoab} changed (decreased) by up to 10%, even reaching values of 100% over limited areas for an O_3 concentration increased by 10%. Aerosol forcing was found to be much more sensitive to wavelength. In a sensitivity test, the spectral GADS aerosol properties were averaged over the UV–VIS range of wavelengths, weighted by the spectrally resolved incoming solar flux, and the resulting mean aerosol optical properties were used in the model instead of the spectral ones (cf. Section 2). This modified locally the forcings ΔF_{TOA}, ΔF_{atmoab} and ΔF_{soldn} by as

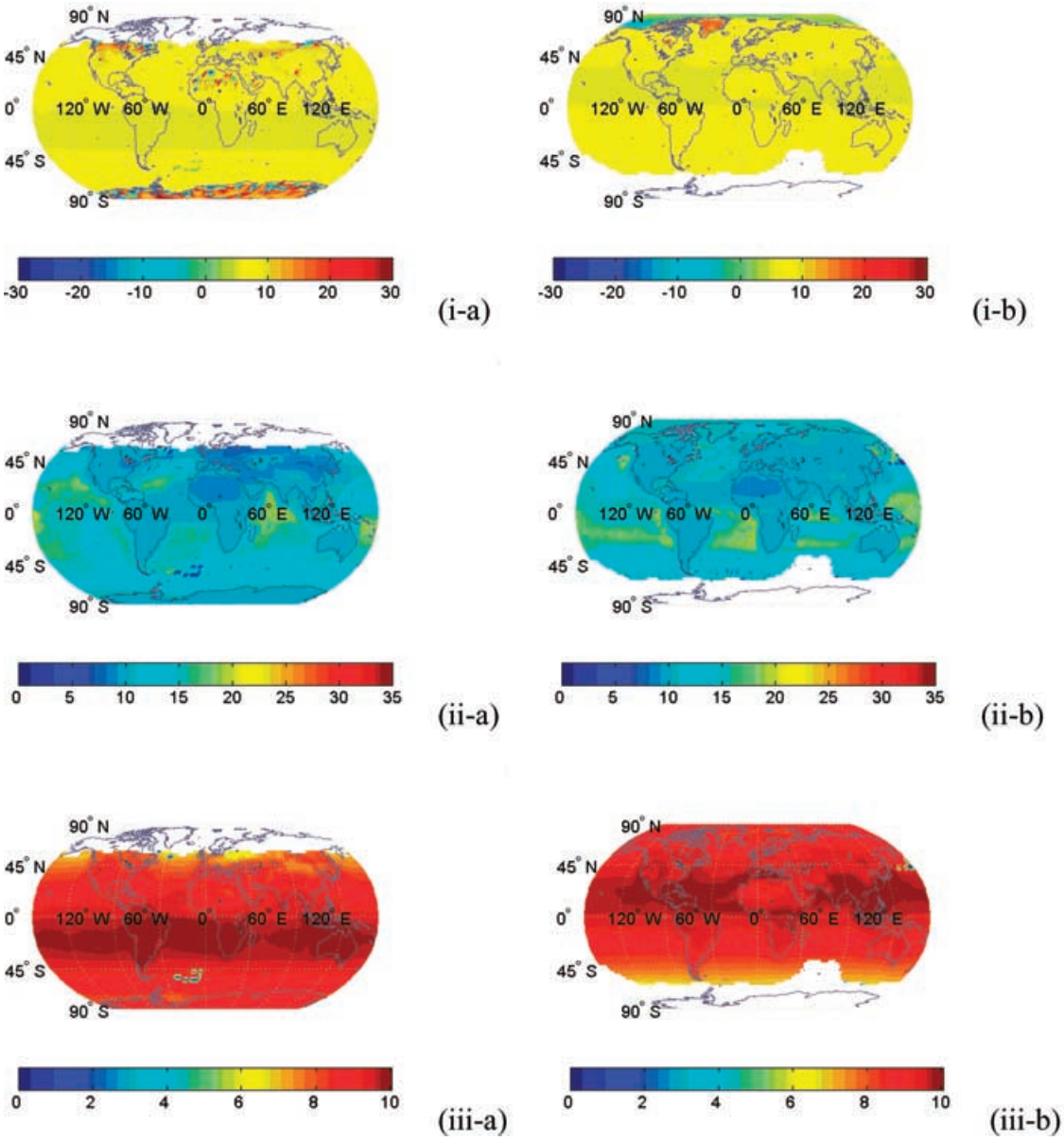

Fig 5. Global distribution of percentage differences ($\Delta\Delta F$ in %) in the reference aerosol forcings (Fig. 1) induced by increasing the extinction aerosol optical thickness (AOT) by 10% (see Section 3.5) for (a) January and (b) July. The aerosol forcings ΔF are explained in Fig. 1.

much as 100%, or by as much as 3 W m^{-2} in absolute terms. This highlights the importance of performing detailed spectral model computations using spectrally resolved aerosol properties to study aerosol radiative forcing, due to the strong spectral variation of aerosol optical properties, i.e. AOT, ω_{aer}, g_{aer}, especially in the UV–VIS range of wavelengths for most aerosol types.

4. Mean annual hemispherical and global results

Table 1 gives the mean hemispherical and global average values for the reference aerosol forcings at the TOA (ΔF_{TOA}), in the atmosphere (ΔF_{atmoab}) and at the surface (ΔF_{soldn}), on a mean annual basis. The reference aerosol radiative forcings were computed for conditions described by the GADS aerosol data in combination with data on surface albedo, RH and amount of cloud valid for the year 1992. Globally, aerosols increase the outgoing solar radiation at the TOA by 0.72 W m^{-2}, while they increase the solar atmospheric absorption by 0.83 W m^{-2}, and decrease the downward solar radiation at the Earth's surface by 2.0 W m^{-2}. There are significant interhemispherical differences for ΔF_{atmoab} and ΔF_{soldn} and larger values for the Northern than the Southern Hemisphere. The largest effect of aerosols on solar radiation is found at the Earth's surface, where it is about

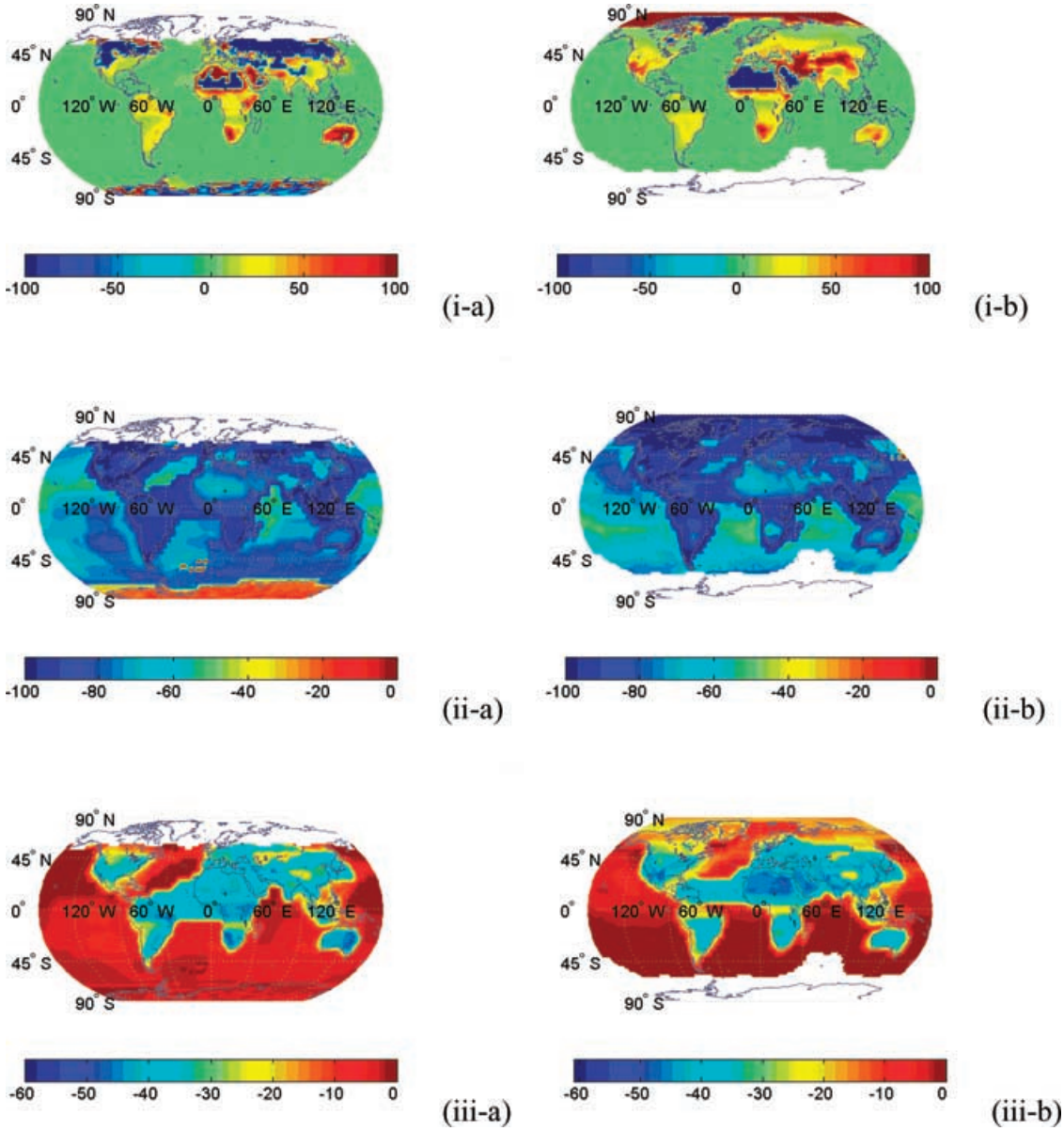

Fig 6. Global distribution of percentage differences ($\Delta\Delta F$ in %) in the reference aerosol forcings (Fig. 1) induced by increasing the aerosol single-scattering albedo by 10% for (a) January and (b) July. The aerosol forcings ΔF are explained in Fig. 1.

three times larger than at the TOA and about twice that in the atmosphere.

The computed changes $\Delta\Delta F$ in the above forcings that resulted from the various model sensitivity tests are given in Table 2. Increasing the RH by 10%, with a limit of 90% (cf. Section 3.2), significantly affected the aerosol forcings (by up to 40%), mainly at the TOA and surface. Our model results show that increasing the RH by 10% increases the aerosol forcings by factors of 1.42 for ΔF_{TOA}, 1.02 for $\Delta F_{\mathrm{atmoab}}$ and 1.17 for $\Delta F_{\mathrm{soldn}}$. Jacobson (2001) reported a factor of 1.9 increase for ΔF_{TOA}, corresponding to a general increase of 15% in their model-predicted RH. Our values, and those of others, indicate that the largest effect of RH on aerosol forcings appears at the TOA, and secondarily at the surface, while the effect within the atmosphere is smaller. Given that aerosols have a two-fold effect, namely surface cooling and atmospheric warming which result in a net planetary cooling, our sensitivity studies show that increasing the RH of the tropospheric aerosol layer would lead to enhanced aerosol cooling of the Earth–atmosphere system, in opposition to the increased atmospheric temperature and water-holding capacity values from global warming due to anthropogenically added greenhouse gases (IPCC, 2001). The results of our model sensitivity tests indicate that increasing RH can induce a change in atmospheric dynamics by reducing the vertical atmospheric

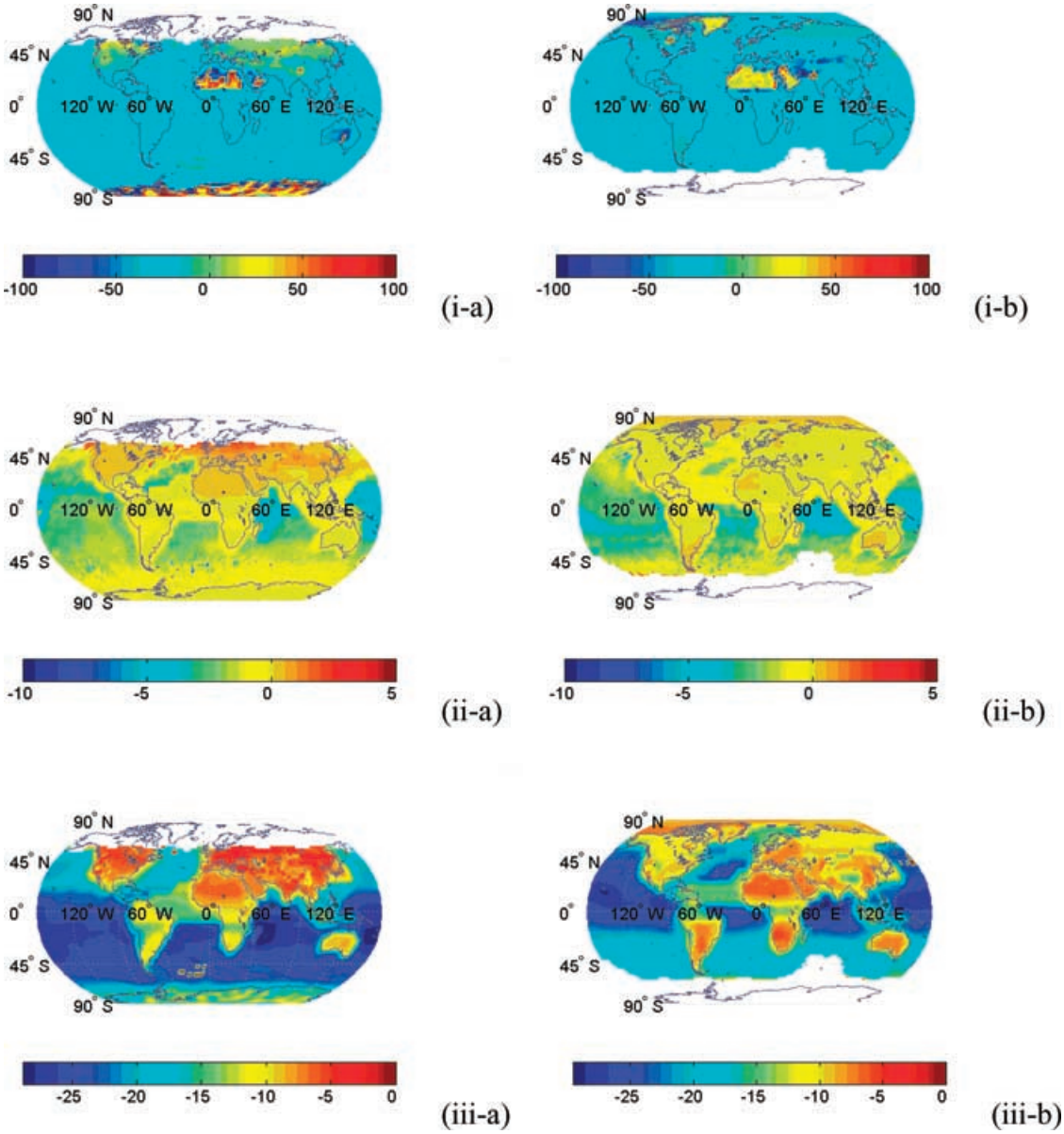

Fig 7. Global distribution of percentage differences ($\Delta\Delta F$ in %) in the reference aerosol forcings (Fig. 1) induced by increasing the aerosol asymmetry parameter by 10% for (a) January and (b) July. The aerosol forcings ΔF are explained in Fig. 1.

temperature gradient and thus causing a decline in evaporation and thus cloud formation and precipitation (e.g. Satheesh and Ramanathan, 2000; Kaufman et al., 2002).

Increasing AOT by 10% was found to increase ΔF_{TOA} globally by 10%, and the forcings $\Delta F_{\mathrm{atmoab}}$ and $\Delta F_{\mathrm{soldn}}$ by 11% and 9% respectively. Thus, increased AOT values, for example due to increased dimethyl sulfide emission from oceans due to increased surface temperature associated with global warming (scenario of Charlson et al., 1987, 1991, 1992) or due to enhanced particulate matter in the atmosphere owing to increased anthropogenic emissions (the "whitehouse" effect of aerosols), would result in planetary cooling associated with a larger surface cooling and a smaller atmospheric warming. Nevertheless, specific regional conditions must be known, since they determine the type and distribution of aerosols as well as their interaction with radiation. In addition, aerosols are not inert but undergo transport, which may even be long-range, thus complicating any investigation of climatic feedback.

The strongest effect of increasing ω_{aer} by 10% was found on $\Delta F_{\mathrm{atmoab}}$ ($\Delta\Delta F_{\mathrm{atmoab}}$ equal to about -80%), while the effects at the TOA and surface are equal to about $\pm 30\%$. Thus, natural or anthropogenic changes in the composition and chemical properties of atmospheric aerosols, and hence in ω_{aer}, for example smaller ω_{aer} values due to increased burning of biomass or

Table 1. Mean annual hemispherical and global average aerosol radiative forcings (in W m^{-2}) used as reference forcings in this study. NH is Northern Hemisphere, SH is Southern Hemisphere. The forcings are given in terms of outgoing solar radiation at the TOA (ΔF_{TOA}), solar atmospheric absorption ($\Delta F_{\mathrm{atmoab}}$) and downwelling solar radiation at the surface ($\Delta F_{\mathrm{soldn}}$)

	ΔF_{TOA}	$\Delta F_{\mathrm{atmoab}}$	$\Delta F_{\mathrm{soldn}}$
NH	0.77	1.44	−2.91
SH	0.68	0.23	−1.08
Global	0.72	0.83	−2.0

Table 2. Differences ($\Delta\Delta F$) in model mean annual reference aerosol radiative forcings ΔF (in W m^{-2}) for the globe, induced by variation (ΔV) of the variable V (RH, relative humidity, in %; AOT, extinction aerosol optical thickness; ω_{aer}, aerosol single-scattering albedo; g_{aer}, aerosol asymmetry parameter; R_g, surface albedo; A_{c}, cloud cover, in %; O_3, ozone concentration; F_{in}, incoming UV–VIS solar radiation at the top of the atmosphere; λ, the wavelength dependence of aerosol optical properties). The forcings are explained in Table 1. The percentage variation ΔV is expressed in relative terms. The forcing changes $\Delta\Delta F$ in parentheses are given as percentage changes of the reference case forcings as given in Table 1. Reference forcings (defined in Table 1) are also given in W m^{-2}

V	ΔV	$\Delta\Delta F_{\mathrm{TOA}}$	$\Delta\Delta F_{\mathrm{atmoab}}$	$\Delta\Delta F_{\mathrm{soldn}}$
RH	+10%	0.3(41.7)	0.02(2.4)	−0.34(17.0)
AOT	+10%	0.07(9.7)	0.09(10.8)	−0.17(8.5)
ω_{aer}	+10%	0.21(29.2)	−0.66(−79.5)	0.62(−31.0)
g_{aer}	+10%	−0.20(−27.8)	0(0)	0.26(−13.0)
R_g	+10%	−0.06(−8.3)	0.02(2.4)	0.02(−1.0)
A_{c}	−5%	0.09(12.5)	0.05(6.0)	−0.15(7.5)
O_3	+10%	0.02(2.8)	−0.03(−3.6)	−0.03(1.5)
F_{in}	+1%	0.01(1.4)	0.01(1.2)	−0.01(0.5)
λ	[a]	−0.01(−1.4)	0.13(15.7)	−0.15(7.5)
Reference forcings		$\Delta F_{\mathrm{TOA}} = 0.72$	$\Delta F_{\mathrm{atmoab}} = 0.83$	$\Delta F_{\mathrm{soldn}} = -2.0$

[a]The sensitivity of aerosol forcings to the wavelength dependence of aerosol optical properties was investigated by using in the model the averaged UV-visible GADS aerosol optical properties instead of the spectral ones (cf. 3.7).

emission of black carbon aerosols from burning of fossil fuels or a change in naturally produced mineral-dust aerosols, could lead to a decoupling of the Earth's climate; this may heat the atmosphere, simultaneously cooling the surface. These effects are opposite and they occur together, even if they may give a local net zero forcing at the TOA (Sinha and Harries, 1997). Besides, although global surface warming due to greenhouse gases should exceed the global cooling effect of aerosols, on a regional scale the aerosol cooling effect may exceed the greenhouse warming (Ramanathan et al., 2001). This could disturb the hydrological cycle of a semi-arid region with inadequate water resources such as the Mediterranean Basin, which now acts as a sink for aerosols from industrial areas to the north and Saharan dust from the south (Lelieveld et al., 2002). Furthermore, a change in the water salinity, associated with global warming (IPCC, 2001), would probably result in a more hydrophilic composition of aerosols in the atmosphere (e.g. NaCl) involving higher ω_{aer} values and hence less heating of the lower troposphere.

Increasing the aerosol asymmetry parameter by 10% in relative values was found to decrease the aerosol forcings at the TOA and at the surface by 28% and 13%, respectively, on a global scale, while it did not affect the forcing $\Delta F_{\mathrm{atmoab}}$. Increasing R_g by 10% in relative values reduced the aerosol planetary cooling by 8%, and amplified the aerosol atmospheric heating by 2%, while it did not significantly affect the aerosol forcing at the surface. According to our model sensitivity tests, a decrease in cloud cover by 5% increased all aerosol forcings by 6–12%, with a larger effect at the TOA. A 1% increase in incoming solar UV–VIS radiation affected the aerosol forcings by up to 1.5%. Slightly larger changes in forcing (up to 4%) were computed for a 10% increase in O_3 concentration. A much larger sensitivity, especially for the aerosol forcings $\Delta F_{\mathrm{atmoab}}$ and $\Delta F_{\mathrm{soldn}}$, was found on the wavelength dependence of aerosol optical properties, equal to up to 16% on a global scale.

5. Summary and conclusions

A series of sensitivity tests were performed with a radiative transfer model to investigate the role of several key parameters of the Earth–atmosphere climate system on aerosol radiative forcings at the TOA, within the atmosphere and at the Earth's surface. RH and aerosol single-scattering albedo are found to be the most important climatic parameters, strongly affecting aerosol forcings. RH affects the forcings in a non-linear way, primarily at the TOA and secondarily at surface. The computed changes in aerosol forcings due to a change in RH by 10% in relative terms are found to reach 300% on a local scale and 40% on global scale. Increasing the RH generally increases the magnitude of aerosol forcings, inducing more outgoing solar radiation at the TOA (cooling effect). Thus, over the polar regions the aerosol warming effect, i.e. reduced outgoing radiation, changes to a cooling effect. On a global scale, increasing RH by 10% increases the aerosol forcings by factors of 1.42 at the TOA, 1.02 in the atmosphere and 1.17 at the Earth's surface. The effect of aerosol single-scattering albedo on aerosol forcing is also very important, involving forcing changes of up to 100% and 80% on local and global scale, respectively. Increasing the aerosol single-scattering albedo by 10%, in relative terms, increased the aerosol forcing at the TOA by 1.29, while it decreased the forcings in the atmosphere and at the surface by factors of 0.2 and 0.69 respectively. The role of AOT and the asymmetry parameter (or other climatic parameters such as surface albedo or cloud cover) remains significant for aerosol forcings, although of less importance, involving changes in forcing of up to 10% on a global scale. The aerosol forcings were found to be quite

sensitive to the wavelength dependence of aerosol optical properties, whereas they do not show a large sensitivity to ozone concentration or to incoming UV–VIS solar radiation.

According to the model sensitivity studies, an increase in RH associated with global warming can result in increased reflected solar radiation at the TOA, increased solar atmospheric absorption and decreased surface downward solar radiation due to aerosols, thus decreasing the atmospheric temperature gradient and possibly reducing cloud formation and precipitation. Increased aerosol loads in the atmosphere, due to natural or anthropogenic activity, can provide a global cooling effect, with important variability and contrasts on a regional scale. In addition, changes in aerosol single-scattering albedo, possibly resulting from anthropogenic modifications of aerosol composition and properties (e.g. from burning of fossil fuel or biomass) can have a significant impact, since a 10% increase/decrease in ω_{aer} was found to decrease/increase the atmospheric solar absorption by 80% on a global scale. Our model sensitivity tests have shown enhanced planetary warming over the Sahara desert under conditions of increased surface albedo, implying that reduced land-vegetation cover would exacerbate desertification processes. This would enhance desertification of the surrounding regions through reduction in cloud cover and precipitation, constituting a positive aerosol feedback. Under decreased cloud cover, aerosols were found to produce higher planetary cooling, atmospheric warming and surface cooling, which can enhance desertification processes in aerosol-polluted semi-arid regions like the Mediterranean Basin through reduced evaporation and precipitation. Nevertheless, more detailed studies are necessary in order to investigate aerosol-related climatic issues in which combined conditions and balancing effects of the climatic parameters examined here can be accounted for.

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