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- Surface radiation trends in Europe scale with aerosol forcing in climate models
- All-sky surface solar radiation trends analyzed from GEBA stations
- Observed trend used as constraint on TOA total aerosol forcing

Supporting Information:
- Readme
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Pollution trends over Europe constrain global aerosol forcing as simulated by climate models

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Abstract An increasing trend in surface solar radiation (solar brightening) has been observed over Europe since the 1990s, linked to economic developments and air pollution regulations and their direct as well as cloud-mediated effects on radiation. Here, we find that the all-sky solar brightening trend (1990–2005) over Europe from seven out of eight models (historical simulations in the Fifth Coupled Model Intercomparison Project) scales well with the regional and global mean effective forcing by anthropogenic aerosols (idealized "present-day" minus "preindustrial" runs). The reason for this relationship is that models that simulate stronger forcing efficiencies and stronger radiative effects by aerosol-cloud interactions show both a stronger aerosol forcing and a stronger solar brightening. The all-sky solar brightening is the observable from measurements (4.06 ± 0.60 W m^{-2} decade^{-1}), which then allows to infer a global mean total aerosol effective forcing at about −1.30 W m^{-2} with standard deviation ±0.40 W m^{-2}.

1. Introduction

Historically, a widespread and strong reduction in aerosol and aerosol-precursor gas emissions has taken place after the "fall of the wall" in 1989 in eastern Europe coincident with air quality control policies in western Europe [*Vestreng et al.*, 2007; *Berglen et al.*, 2007]. Emission reduction over Europe has mainly come from the economic restructuring after the fall of the Berlin Wall, from switching coal to gas and from more desulfurization of emissions from power plants. The increasing trends in surface solar radiation (brightening) have been linked to the decrease in anthropogenic aerosol emissions over the European region [*Wild et al.*, 2005, 2007; *Norris and Wild*, 2007; *Ruckstuhl and Norris*, 2009; *Ruckstuhl et al.*, 2008; *Wild and Schmucki*, 2011; *Chiacchio et al.*, 2011]. These trends subsequently have likely influenced changes in the Earth's surface temperature [*Wild et al.*, 2007; *Ruckstuhl et al.*, 2008]. Recent studies showed that reduced haze, mist, and fog conditions over the past 30 years may have contributed on average up to about 50% of observed eastern European warming [*Vautard et al.*, 2009]. Emission reduction has also been linked to an increase in horizontal visibility of 15 km from 1983 to 2008 in a heavily industrialized area over Europe [*Stjern et al.*, 2011]. Prior to the solar brightening trend, a strong increase in aerosol emissions since the midtwentieth century took place over the same region.

In this study, we aim to quantitatively constrain the total (direct + indirect) aerosol radiative effect. This forcing, in particular the "indirect" effect by the modification of clouds, continues to constitute one of the main uncertainties in climate research [*Boucher et al.*, 2013]. Even if aerosol emissions are harmonized, climate models show a wide range in simulated forcings due to the different parameterizations of forcing efficiency and aerosol-cloud interactions [*Textor et al.*, 2007; *Quaas et al.*, 2009]. Previous studies using observations to constrain the indirect effect suggested smaller values compared to climate model results [*Quaas et al.*, 2008, 2009] but have been challenged recently [*Penner et al.*, 2011; *Quaas et al.*, 2011].

The main focus of this study is to exploit the trends in solar brightening over Europe to infer clues about the global climate forcing by anthropogenic aerosols. For this, we have used observations and the climate model simulations submitted to the Fifth Coupled Model Intercomparison Project (CMIP5) archive [*Taylor et al.*, 2012]. We first discuss model-simulated all-sky solar brightening trend (1990–2005) over the European region. We then discuss the relationship between regional solar brightening and regional and global anthropogenic aerosol radiative forcing. Finally, we use the observed solar brightening over the European region to constrain the model-simulated regional and global aerosol radiative forcing.

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Section 2 discusses the observation data sets used in this study. The model data sets and methodology are described in section 3. The trends in solar brightening and its relationship to aerosol radiative forcing is described in section 4. Main findings are presented in section 5.

2. Data Sets

Surface solar radiation observational data are taken from the Global Energy Balance Archive (GEBA) [*Ohmura et al.*, 1989; *Gilgen et al.*, 1998; *Wild et al.*, 2005]. The surface solar radiative fluxes are measured using well-calibrated instruments at around 300 GEBA sites over Europe [*Wild et al.*, 2005]. These values underwent rigorous quality checks to assure high accuracy in the measurements [*Wild et al.*, 2005; *Gilgen et al.*, 1998]. The relative random error of GEBA data was found to be approximately 2% of the annual mean values [*Gilgen et al.*, 1998]. In this study, 52 GEBA stations with near-complete record of observations were used. Monthly mean 1° gridded data from the Climate Research Unit (CRU) archive was used to examine the observational trends in surface temperature [*Mitchell and Jones*, 2005]. The uncertainty in the annual mean observed CRU temperature values is reported as $\pm$0.05° C (2 standard errors) for the period since 1951 [*Brohan et al.*, 2006]. The model data sets and methodology are further described in section 3.

3. Methodology

CMIP5 experiments are designed to explore the effects of changes in aerosols, greenhouse gases, and natural forcings. In this study, the analysis is performed for the three historical scenarios ("historical," "historicalGHG," and "historicalNat") from the late nineteenth century to 2005. The "piControl" simulation is run to equilibrium with all natural and anthropogenic forcings held fixed at the condition corresponding to 1850 (preindustrial (PI) forcing). The historical experiments take into account effects of all natural and anthropogenic forcings following time-varying boundary conditions, e.g., for the emissions. In turn, the historicalGHG experiment includes only the effects of long-lived greenhouse gases, varying from 1850 to present-day conditions. The historicalNat includes only the effects of solar and volcanic forcing in a time-dependent manner for the same period [*Taylor et al.*, 2012]. Each of these perturbation experiments was initialized in January 1850 or 1860 from states taken from PI control run. Each ensemble members of these perturbation experiments are simulated by using different initialization time states taken from the PI control run. An addition to this, the two 30 year atmospheric runs ("sstClim" and "sstClimAerosol") with prescribed sea surface temperature and sea ice taken as a long-term average from the PI control run was used for the radiative forcing calculations. The sstClim experiment uses 1850 anthropogenic aerosol emissions, whereas the sstClimAerosol uses anthropogenic aerosol emissions for the year 2000. In this analysis, we mainly focused on the trends in anthropogenic aerosol emissions, model-simulated trends in aerosol optical depth and their effects on radiation and surface temperature.

Within the CMIP5 archive, 10 models contributed to the historical and eight models contributed to sstClim and sstClimAerosol experiments and have been analyzed in this study. All the CMIP5 models that used interactive aerosol schemes and performed all the three experiments (historical, sstClim, and sstClimAerosol) are considered for the present analysis. All models considered here used interactive aerosol schemes rather than prescribed aerosol burdens or forcing. Fifty ensemble runs from 10 models in the historical experiment are analyzed. For each ensemble member of each model, the simulated downwelling surface solar radiation trends are estimated by a least squares regression over the 52 GEBA stations points in the historical experiment for the period from 1990 to 2005. The effective forcing (EF) is estimated as the difference between sstClimAerosol and sstClim for the 10 year mean period for each model. The effective forcing as defined by the fifth assessment report of the Intergovernmental Panel on Climate Change [*Boucher et al.*, 2013] includes the pure radiative forcing but also adjustments that occur on fast (i.e., up to months) time scales. The uncertainty is computed by shifting the averaging period by $\pm$ 2 years. This results in about $\pm$ 0.018 W m^{-2}.

The observed solar brightening is estimated using the GEBA data [*Ohmura et al.*, 1989]. The annual mean observed surface solar radiation is calculated by averaging the global (i.e., direct plus diffuse) radiation fluxes (W m^{-2}) at 52 different stations that fall inside the continental European region (25°W to 45°E and 35°N to 70°N) for the period 1990 to 2005. The annual mean observational solar radiation data are used to calculate the brightening trends during the 1990 to 2005 period. A least squares linear regression method was applied for the trend analysis and the uncertainty range in the observations is estimated by varying

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for the linear trends the start year by one, two, or three (i.e., using the 1988–2005, 1989–2005, 1990–2005, 1991–2005, and 1992–2005 periods).

4. Results and Discussion

All the CMIP5 models in the historical experiments, analyzed in this study, broadly simulate an all-sky solar brightening trend from 1990 to 2005, albeit smaller compared to the mean observed trend (4.06 ± 0.6 W m^{-2} $decade^{-1}$) estimated from 52 GEBA measurement sites over the continental Europe region (Figures 1 and S7 in the supporting information). The Model for Interdisciplinary Research on Climate (MIROC5), Geophysical Fluid Dynamics Laboratory-CM3, Hadley Centre Global Environment Model (HadGEM2)-ES, and Commonwealth Scientific and Industrial Research Organisation-Mk3.6.0 models are close to, but are slightly higher than, the observed trends. In total 48 out of 50 ensemble members, each initialized slightly differently and as such simulating different weather conditions, yield a solar brightening trend for the historical experiment. The model-simulated solar brightening is in line with the strong simulated reduction (43%) in aerosol optical depth (AOD) at 550 nm over the continental Europe region (Figures S3 and S7). The AOD reduction in the models mainly stems from a strong decline (4 Tg yr^{-1} $decade^{-1}$, Figures S1 and S7) of sulfur dioxide and coemitted anthropogenic aerosols due to more effective air pollution regulations and economic restructuring after the fall of the Berlin Wall in the eastern European region since 1989 and imposed to the CMIP5 models as decadally changing anthropogenic emissions [*Lamarque et al.*, 2010]. The model-simulated AOD trends are also comparable with the observed AOD reduction of about 62% from the surface point measurements over six sites 1986–2005 [*Ruckstuhl et al.*, 2008]. However, for the entire European domain, no long-term AOD observations are available.

For each model a fairly large spread in the simulated brightening by individual ensemble members is found. Several individual simulations closely capture the magnitude of the observed brightening (Figure S6). However, for most models the ensemble average yields a somewhat lower or higher than observed trend (Figure S6). The difference between ensemble average trend and the trend that found for individual ensemble members can be considered as the contribution of simulated internal variability to the trend found for that ensemble member. This suggests that internal variability might have contributed to the solar brightening trend. None of the ensemble members of the historicalGHG and only one-ensemble member of the historicalNat experiment are able to quantitatively reproduce the brightening trend (Figure S6).

The CMIP5 model-simulated mean effective forcing (EF = idealized sstClimAerosol (present-day aerosol emissions at year 2000) minus sstClim (preindustrial aerosol emissions at year 1850)) is used to examine the overall climatic effect of anthropogenic aerosols. For this, mean all-sky solar radiation EF over the entire continental European region along with the mean all-sky solar brightening trends from historical experiments over 52 GEBA stations are analyzed. The scatterplot of EF on solar radiation at surface and top-of-the-atmosphere (TOA) versus 1990–2005 all-sky solar brightening trends show that in the CMIP5 models, the EF not only at the surface but also at TOA over the continental European region are closely linked to the surface brightening trends (Figures 1a and 1b). Such a relationship of an unknown, climate change-related quantity and an observable has been used previously, e.g., to quantify the snow-albedo feedback from the seasonal cycle in snow cover extent [*Hall and Qu*, 2006]. Observed brightening trends thus may be used to constrain the regional anthropogenic aerosol radiative effect. Given the mean value and uncertainty in surface radiation trends, and in the scaling between surface radiation trend and regional EF, we estimate the constrained regional EF at the surface (−5.17 ± 1.62 W m^{-2}) and at the TOA (−3.56 ± 1.41 W m^{-2}). The difference between surface and TOA EF of about +1.61 W m^{-2} suggests a considerable atmospheric warming due to aerosol absorption of sunlight. More importantly still, the solar brightening trend 1990–2005 over Europe from seven out of eight models (except MRI-CGCM3) also scales well with the global mean effective forcing (Figure 1c). This is because MRI-CGCM3 model simulates exceptionally strong atmospheric absorption of black carbon aerosols over the entire European continent. The all-sky solar brightening is the observable from GEBA, which then allows to infer a total global aerosol effective forcing at −1.30 ± 0.40 W m^{-2} (Figure 1, red lines, center and error bar).

Finally, we are interested in examining how the observed solar brightening relates to trends in surface temperature over Europe. All the CMIP5 models in the historical experiment simulate a warming trend from 1990 to 2005 except MRI-CGCM3, albeit slightly higher compared to the mean CRU observed trend (0.5 K $decade^{-1}$) over the Europe region (Figures 2, S5, and S6). In total 48 out of 50 ensemble runs yield a

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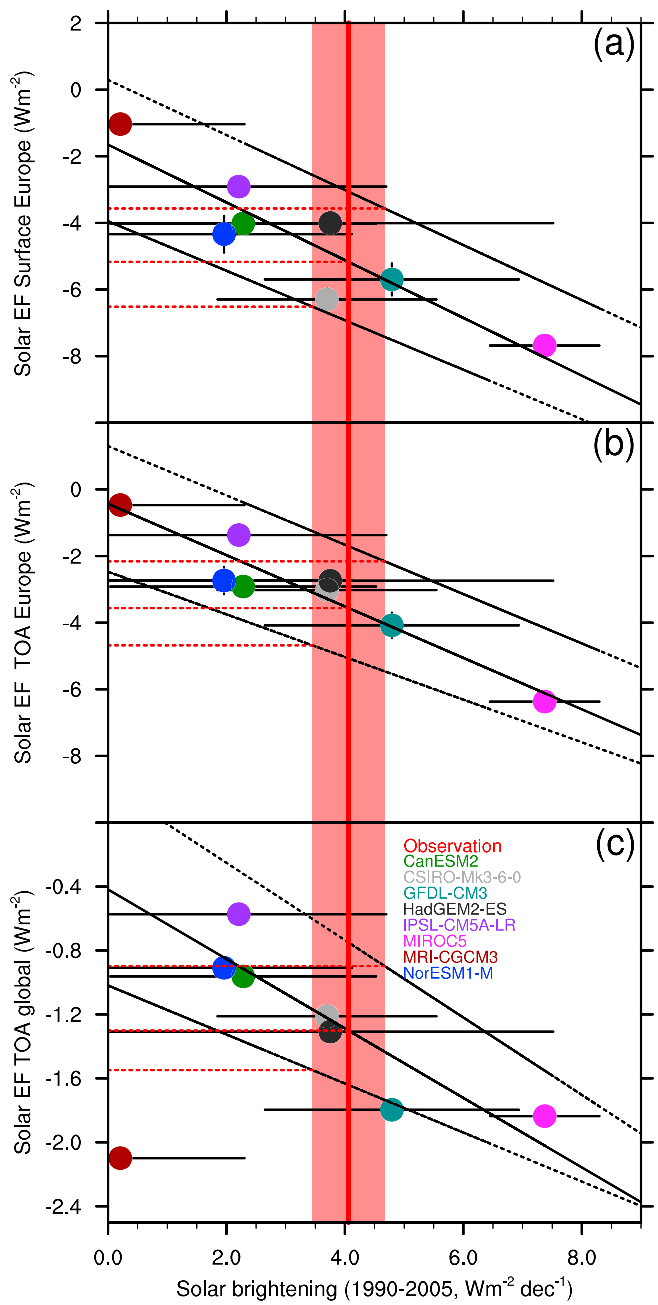

Figure 1. Solar brightening trend for the years 1990 to 2005 over 52 GEBA observational sites (Figure S11) in the European region for the historical experiment versus aerosol effective forcing (EF = sstClimAerosol − sstClim) at (a) surface, (b) top-of-the-atmosphere (TOA) over the continental European region, and (c) global TOA. Please note the different scales on the *y* axis. The Meteorological Research Institute (MRI)-coupled (ocean-atmosphere) general circulation model CGCM3 model is not included in the regression analysis in Figure 1c. The different colors are for observations and different models. The black lines indicate the least squares trend line (average and 1 standard deviation considering the uncertainties for each model and the observations). The red lines indicate the constrained forcing value line and light red shade indicates the uncertainty range in the GEBA observed trend.

warming trend for the historical experiment. The scatterplot between solar brightening and the temperature trend difference from 1990 to 2005 and 1960 to 1980 shows that the solar brightening trend simulated by CMIP5 models scales with the change in warming trend over the continental European region (Figure 2a). The temperature trend difference between the two periods is used to single out to a first order the greenhouse gas and natural forcing influence from aerosol forcing. The models that quantitatively reproduce the brightening trend show a stronger warming trend than the observed one (Figure 2a). From the

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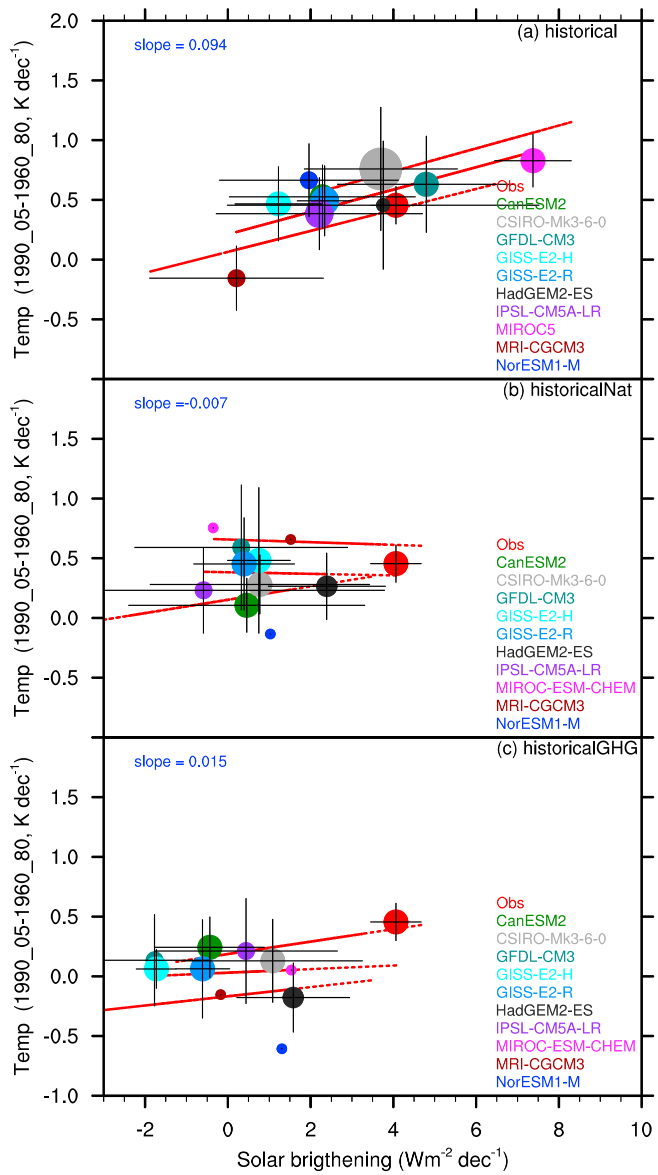

Figure 2. Temperature trend difference between 1990–2005 and 1960–1980 versus solar brightening trend for the years 1990 to 2005 over the 52 GEBA observational sites in the continental European region for the (a) historical, (b) historicalNat, and (c) historicalGHG experiments. The different colors are for observations and different models. The red line indicates the least squares trend line. The size of the symbols denotes the number of ensemble runs for each model.

analysis of the historicalGHG (GHG forcing only) and historicalNat (natural forcing only) simulations, we conclude that the reason is an overestimation of the effect of natural forcings, related to the recovery after a too strong cooling due to volcanic eruption of Mount Pinatubo in 1991 in the European region compared to what has been observed (Figure 2b). The warming due to greenhouse gases, though, did increase only slightly from the 1960–1980 to the 1990–2005 period, in line with expectations (Figure 2c).

The actual observational constraint in this approach is on the trends for the recent history. Then the method makes use of the climate modeling for the total aerosol radiative forcing for the present day with respect to the preindustrial period. The evaluation of the recent brightening trend effectively is a constraint on the strength of the simulated aerosol effects on radiation and clouds. It should be noted, however, that simulations of the global aerosol loading by different models, in general, show good agreement with satellite and in situ observations, leading to better confidence in aerosol emission estimates. But there are still

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uncertainties in the model-simulated anthropogenic contribution to aerosol burden [*Textor et al.*, 2007; *Bellouin et al.*, 2013].

5. Conclusions

In conclusion, the models, driven by a decline in anthropogenic aerosol emissions, simulate a decrease in aerosol burden and thereby a solar brightening trend over Europe (for most of the models), for the 1990–2005 period. The close link between the simulated brightening trends and the global mean total aerosol effective forcing allows for a constraint of the anthropogenic aerosol effect at -1.30 ± 0.40 W m^{-2}, which is stronger than the recent Intergovernmental Panel on Climate Change estimate [*Boucher et al.*, 2013] but consistent with previous estimates [e.g., *Bellouin et al.*, 2013]. In addition to the effect of greenhouse gases, the solar brightening has strongly contributed to the observed 0.5 K decade^{-1} European warming since the mid-1980s.

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