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Atmospheric Chemistry and Physics Discussions

Robust evidence for reversal in the aerosol effective climate forcing trend

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Abstract. Anthropogenic aerosols exert a cooling influence that offsets part of the greenhouse gas warming. Due to their short tropospheric lifetime of only up to several days, the aerosol forcing responds quickly to emissions. Here we present and discuss the evolution of the aerosol forcing since 2000. There are multiple lines of evidence that allow to robustly conclude that the anthropogenic aerosol effective radiative forcing – both aerosol-radiation and aerosol-cloud interactions – has become globally less negative, i.e. that the trend in aerosol effective radiative forcing changed sign from negative to positive. Bottom-up inventories show that anthropogenic primary aerosol and aerosol precursor emissions declined in most regions of the world; observations related to aerosol burden show declining trends, in particular of the fine-mode particles that make up most of the anthropogenic aerosols; satellite retrievals of cloud droplet numbers show trends consistent in sign, as do observations of top-of-atmosphere radiation. Climate model results, including a revised set that is constrained by observations of the ocean heat content evolution show a consistent sign and magnitude for a positive forcing relative to 2000 due to reduced aerosol effects. This reduction leads to an acceleration of the forcing of climate change, i.e. an increase in forcing by 0.1 to 0.3 $\mathrm{W\,m^{-2}}$, up to 12% of the total climate forcing in 2019 compared to 1750 according to IPCC.

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1 Introduction

Anthropogenic pollution particles, aerosols, exert an effective radiative forcing on climate due to aerosol-radiation interactions (ERFari, also known as "aerosol direct effect") and aerosol-cloud interactions (ERFaci, "aerosol indirect effect") (Boucher et al., 2013; Forster et al., 2021; Szopa et al., 2021). ERFari occurs through the scattering and absorption of sunlight by aerosols while for ERFaci aerosols act as cloud condensation nuclei (Twomey, 1974). Both entail rapid adjustments that tend to enhance the radiative forcing. A recent assessment provided an estimated total ERF due to aerosols (ERFaer) in the range of -2.0 to $-0.35\,\mathrm{W\,m^{-2}}$ (5 to 95% confidence interval; 2005 to 2015 compared to 1850) (Bellouin et al., 2020b). The latest assessment report by the Intergovernmental Panel on Climate Change (IPCC) concluded that the 2019 vs. 1750 ERFaer has a best estimate of –1.1 $\mathrm{W\,m^{-2}}$ and 5 to 95% confidence interval of –1.7 to –0.4 $\mathrm{W\,m^{-2}}$ (Forster et al., 2021). This negative forcing offsets a sizeable fraction of the current CO_2 ERF. Forster et al. (2021) quantify a temperature increase in 2019 relative to 1750 of +1.01°C due to the ERF by CO_2, and a temperature change by –0.51°C due to aerosols in that period. This implies that without the cooling effect of aerosols, the world would already have reached the 1.5°C temperature threshold of "dangerous" climate change as set out by the Paris agreement.

A fundamental difference between radiative forcing by aerosols and long-lived greenhouse gases is tied to their atmospheric lifetimes: greenhouse gases have lifetimes of decades to millennia (Solomon et al., 2009), while the lifetime of tropospheric aerosols is only up to several days. Climate thus responds to long-lived greenhouse gases such as CO_2 largely in terms of their cumulative emissions, but to aerosols in direct link to its current emissions rate. Shorter lived greenhouse gases such as methane have an intermediate effect, whereby deep reductions in emissions can have substantial effects on temperature within a few decades (Shindell and Smith, 2019; Smith et al., 2021b; Allen et al., 2022). A further reduction in aerosol emissions – motivated by their environmental and health impacts (Lelieveld et al., 2015; Cohen et al., 2017) – thus takes out the negative aerosol forcing and leads to a warming relative to the period prior to emission reduction (Brasseur and Roeckner, 2005; Dufresne et al., 2005), an effect also known as climate penalty of air quality improvements (Ekman et al., 2020; Hong et al., 2020). Also, the importance of the aerosol forcing relative to the CO_2 forcing, was largest in the early industrial period (Stevens, 2015). It will continue to decrease, since anthropogenic aerosol emissions will likely decrease at the global level (Myhre et al., 2015; Szopa et al., 2021).

At what point did the aerosol forcing became substantially less negative at a scale relevant for global forcing? There are suggestions that decreasing started in the last decades in different regions, and for several regions this trend reversal has been documented (e.g., Cermak et al., 2010). This decrease stems in particular from reductions of SO_2 emissions from coal use in residential sector, power plants, and industry. For other regions, evidence is lacking or more anecdotal. However, for understanding of global climate change, it is thus relevant to ask to which extent the aerosol climate forcing has become less negative at the global scale.

Here we propose that aerosol trends and their effects can be best investigated in the satellite era since the turn of the century. We analyse multiple observational and model datasets to demonstrate that in regions affected by anthropogenic aerosol and

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aerosol precursor emissions, there are multiple lines of evidence that show that both, ERFari and ERFaci, show reduced trends since 2000, in regions which demonstrate a robust and substantial aerosol ERF trend in models.

2 Changes in aerosol emissions

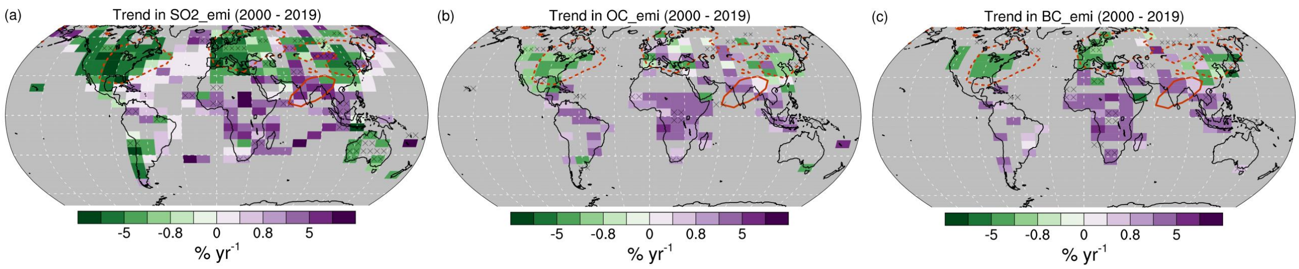

Figure 1. Linear trends (2000 to 2019) of (a) anthropogenic emissions in sulfur dioxide (SO_2) from the Community Emissions Data System (CEDS v_2021_04_21; Hoesly et al., 2018); (b) and (c) as (a), but for anthropogenic emissions in organic carbon (OC) and black carbon (BC), respectively. Regions with small absolute trends (less than 7 μg m^{-2} day^{-1} yr^{-1}) are masked by grey shading. Isolines enclose regions with trends in clear-sky solar ERF (see later, Fig. 4) larger than 0.05 W m^{-2} yr^{-1} in absolute terms. The average values in these regions as listed in Table 1 and discussed in Section 7. A figure that combines the panels of Fig. 1 to 4 is provided as Supplementary material.

Despite substantial differences in their absolute magnitude especially at the regional level (Elguindi et al., 2020), the different emission inventories agree in general on the sign of the historical trends at regional and global levels (Granier et al., 2011; Klimont et al., 2017; Hoesly et al., 2018; Aas et al., 2019; Elguindi et al., 2020), especially over Europe and North America (Elguindi et al., 2020). A number of clear conclusions have thus been drawn in the literature for aerosol emissions in specific regions. Aerosol emissions have seen a steep increase since the beginning of the industrial period (e.g., Szopa et al., 2021). In several regions, declines after a peak are documented. An example is Europe, where since the 1980s, aerosol emissions declined strongly following air quality policies (Krüger and Graßl, 2002; Vestreng et al., 2007; Tørseth et al., 2012; Cherian et al., 2014; Crippa et al., 2016; Costa-Surós et al., 2019). A similar behaviour is documented for North America (e.g., Streets et al., 2009; Aas et al., 2019; Elguindi et al., 2020). Sulphur and Nitrogen deposition over the USA, reflecting anthropogenic emissions, have been declining by between 1 and 3% yr^{-1} in the period 1989–2010 (Sickles II and Shadwick, 2015). In contrast, anthropogenic aerosol emissions over China have been increasing until around 2010, and decreasing thereafter (Klimont et al., 2017; Zheng et al., 2018; Aas et al., 2019; Wang et al., 2021). The exact temporal evolution of aerosol emissions over the past 20 years especially over China was erroneously represented (a too weak decline since 2010) in some emission datasets, leading to some incompatibility of aerosols in the 6th Coupled Model Intercomparison Project (CMIP6 Eyring et al., 2016; Hoesly et al., 2018; Elguindi et al., 2020) in comparison to observations (Paulot et al., 2018; Wang et al., 2021). Aerosol emissions over India continued to rise throughout the period 2000 to 2019 (Klimont et al., 2013; Wang et al., 2021). Over remote oceanic regions, ship emissions played a substantial, increasing role in the first part of the time period of interest (Smith et al., 2011). Since

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2010, they have declined first in emission control areas (IMO, 2008) and since 2020 over much of the global oceans (IMO, 2019); this declining signal is visible also in cloud properties (Gryspeerdt et al., 2019).

Here we consider more specifically emissions from the the newest version of the Community Emissions Data System (CEDS; O'Rourke et al., 2021), as also used in CMIP6 and as described by Hoesly et al. (2018) and are shown in Fig. 1. Sulfur emissions were mostly declining since 2000, in particular there were substantial declines over North America and Europe, continuing decreasing trends that started in the last decades of the 20th century. Also over East Asia, due to reductions after 2010, the overall trend is negative, despite the fact that in the first decade of the 21st century, emissions did still increase. Over Southeast Asia, including India, and also over substantial parts of Africa, sulfate precursor emissions showed increasing trends. Some shipping routes over ocean show increasing trends in this period. OC and BC emissions mirror this image broadly, but with substantially more widespread increases especially over more regions in East Asia, Africa, and also South America. All considered aerosol species show increasing trends in emissions for high latitudes of both hemispheres. The updates of CEDS emissions (Elguindi et al., 2020) show that more recent evidence points to even stronger decline in SO_2 emissions in the second part of the last decade and the BC and OC trends are showing a decline rather than increase, especially in China (see also Kanaya et al., 2020). These are further discussed in Elguindi et al. (2020).

3 Changes in aerosol abundance

The emission trends are reflected in observations of aerosol abundance. Due to their short lifetime, it is expected that regional trends in emissions are also reflected by regional trends in concentrations that are somewhat smoothed out spatially, in case of typically prevailing wind directions mostly leewards. Trends in surface concentrations from in situ observations were found to show the expected trends in a global compilation (Collaud Coen et al., 2020) for sulfate and PM2.5, and specifically for the declining trends over Europe (Stjern et al., 2011; Aas et al., 2019) and North America (Jongeward et al., 2016; Aas et al., 2019), and the first increasing, then decreasing behaviour over China (Zheng et al., 2018; Aas et al., 2019).

The analysis of trends from remote sensing, especially from satellite remote sensing, is challenging, because datasets may not be homogeneous over the lifetime of a satellite instrument, due to changing instrument response and satellite orbit. However, for NASA's Earth Observation Satellites Terra and Aqua, care has been taken to avoid many of the issues that hamper satellite trend analysis such as orbital drift (Levy et al., 2013). Studies show that trends from various satellites are, at least qualitatively, consistent (Wei et al., 2019). Declining trends in aerosols in certain regions, such as over Europe (Stjern et al., 2011; Cherian et al., 2014; Li et al., 2014; Georgoulias et al., 2016; Cherian and Quaas, 2020) and over the USA (Li et al., 2014; Jongeward et al., 2016; Cherian and Quaas, 2020), as seen from satellite analysis, have been documented earlier. The changes in aerosols over East Asia, especially China, are not monotonic over the period of interest. Rather, the trends reversed from positive (2000–2010) to negative (since 2010), and this is seen in satellite observations of aerosols (Paulot et al., 2018; Sogacheva et al., 2018; Filonchyk et al., 2019; Ma et al., 2019; Samset et al., 2019). In contrast, over Southeast Asia, especially India, aerosol retrievals from satellites show continuing increases throughout the period (Li et al., 2014; Zhao et al., 2017; Dahutia et al., 2018; Hammer et al., 2018; Cherian and Quaas, 2020). Model-data synergy allowed to attribute these satellite-derived trends

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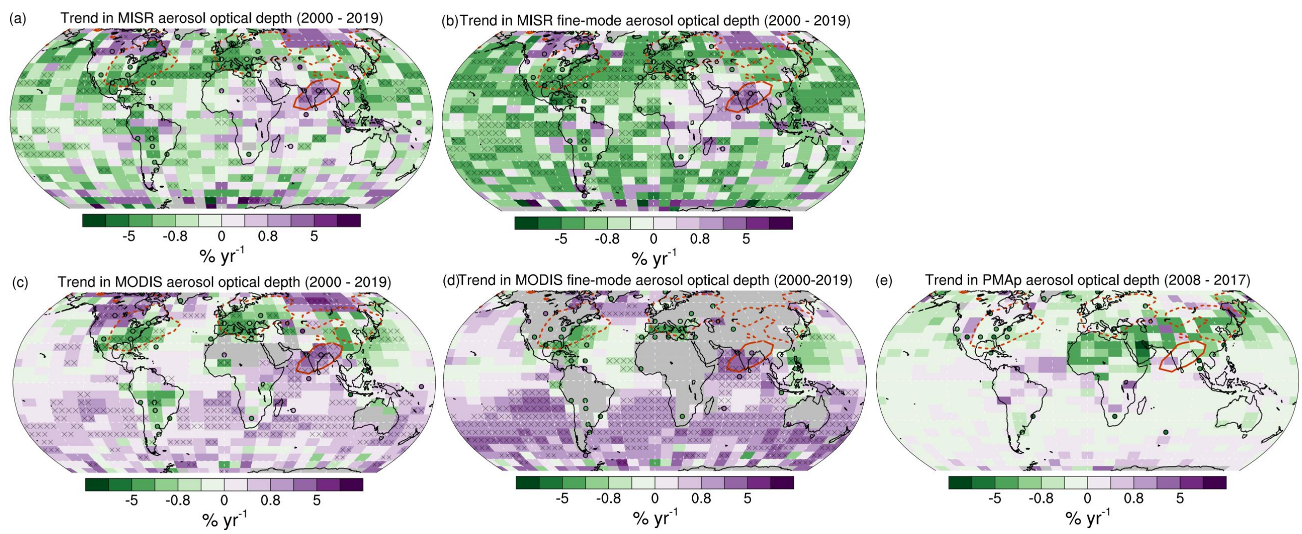

Figure 2. Linear trends (2000 to 2019) of (a) aerosol optical depth (AOD) as retrieved from the Multi-angle Imaging Spectroradiometer (MISR; Garay et al., 2017) on board the Terra satellite, the coloured circles show the AOD trends from the AERONET ground-based sunphotometer network (Holben et al., 1998; Giles et al., 2019) where data since 2000 are available; (b) as (a) but for the fine-mode AOD, i.e. the AOD due to aerosols with radius smaller than 1 μm. (c) and (d) as for (a) and (b), but retrievals from the MODerate Resolution Imaging Spectroradiometer (MODIS; Levy et al., 2013) retrievals (fine-mode AOD unavailable over land) from the Terra satellite averaged (starting 2002) with MODIS retrievals from the Aqua satellite; (c) PMAp aerosol optical depth as retrieved from the Global Ozone Monitoring Experiment–2 (GOME-2) instrument on-board EUMETSAT's Metop-A satellite that is available only for 2008 to 2017. Isolines as in Fig. 1.

to the specific emission changes (Bauer et al., 2020; Yu et al., 2020), and to quantify the changes at between -3.1 to -1.2% yr^{-1} for the different regions affected by the declines in anthropogenic aerosol emissions from 2000 to 2014 (Mortier et al., 2020). Mortier et al. (2020) further documented that climate models were able to reproduce these trends quantitatively.

We report AOD trends from various satellite datasets on a common scale in Fig 2. It specifically shows aerosol optical depth (AOD) and fine-mode AOD (AODf) from the MODerate Resolution Imaging Spectroradiometer (MODIS, Levy et al., 2013) instrument on board the EOS Terra and Aqua satellites, and the Multi-Angle Imaging Spectro-Radiometer (MISR, Garay et al., 2017) instrument on board the EOS Terra satellite. Also the – presumably more stable – ground-based retrievals from the AERONET network (Holben et al., 1998, 2001; Giles et al., 2019) are analysed for the stations for which the time series since 2000 are available. The trends in both AOD and AODf from the different satellite instruments in the Southern hemisphere oceanic, and also in the Northern hemisphere high-latitude oceanic regions differ – MODIS shows increases and MISR, decreases or scattered results in both quantities. As a third estimate, the EUMETSAT Polar Multi-sensor Aerosol optical properties product (PMAp, Grzegorski et al., 2021) climate data record derived using the Global Ozone Monitoring Experiment–2 (GOME-2) instrument on board EUMETSAT's Metop-A satellite are used. These are available for a shorter, 10-year period, for 2008 to 2017. In the Southern hemisphere oceanic regions, MetopB shows very small trends for this shorter

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period; in the Northern hemisphere high latitude oceans, it tends to confirm the decreases shown by MISR. However, in the regions discussed above with pronounced trends in anthropogenic aerosol (precursor) emissions, the satellite trends show the expected behaviour qualitatively in all three datasets. These trends are largely consistent with those from AERONET data. The decreasing trends over North America, Europe, and East Asia are clearly seen and at many grid points statistically significant (at 5% significance level), as are the increasing trends over India. It is particularly interesting to note that the trends in AODf are more consistent still in spatial extent to the changes in sulfate (precursor) emissions. These smaller particles, with radii $<1\,\mu$m, contain the bulk of the anthropogenic contribution to the aerosol (Bellouin et al., 2005; Kaufman et al., 2005; Kinne, 2019).

4 Changes in cloud properties

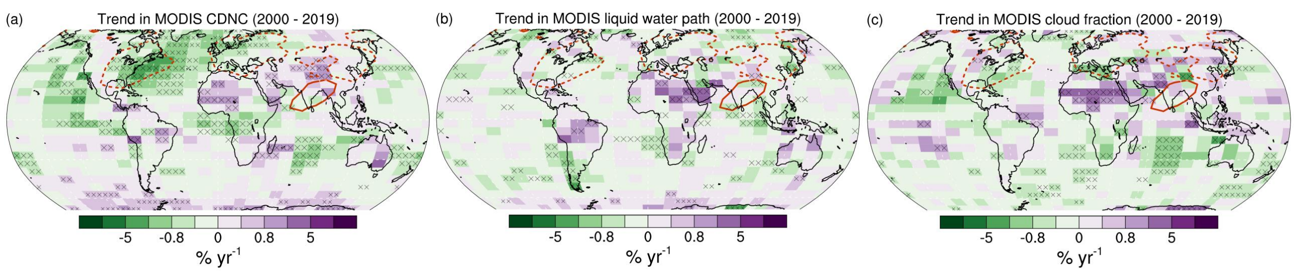

Figure 3. Linear trends (2000 to 2019) in cloud properties retrieved for liquid-water clouds from MODIS (Platnick et al., 2017) where cloud droplet number concentration (CDNC, panel a) and cloud liquid water path (LWP, panel b) are computed assuming adiabatic clouds (Quaas et al., 2006; Grosvenor et al., 2018); (c) liquid cloud fraction. Isolines as in Fig. 1.

Clouds are a key determinant for variability and trends of the Earth's energy budget. Due to their large spatio-temporal variability it is not easy to distinguish long-term signals from weather noise. Clouds respond not only to aerosols, but also to global warming and interannual as well as decadal internal climate variability (Forster et al., 2021). Overall, satellite analysis documented changes in clouds that are consistent with several of the hypotheses relevant for cloud-climate feedbacks (Norris et al., 2016), but little evidence for patterns of cloud cover or cloud-top altitude trends that would be expected due to aerosol-cloud interactions (Norris et al., 2016). The most immediate impact of aerosols is on cloud droplet number concentration (Bellouin et al., 2020b; Quaas et al., 2020). For this microphysical quantity, some clear and significant trends were identified in satellite observations for the outflow region east of East Asia (Bennartz et al., 2011), albeit the declining trends in cloud water path and cover are not what is expected in relation to aerosol-cloud interactions (Benas et al., 2020).

The trends in satellite-derived cloud droplet number concentrations are consistent with the aerosol trends (McCoy et al., 2018; Cherian and Quaas, 2020). Trends in cloudiness and cloud radiative properties are, however, less conclusive possibly due to their large variability (Norris et al., 2016; Cherian and Quaas, 2020). The trends in MODIS retrievals of cloud properties (Platnick et al., 2017) are shown in Fig. 3. MODIS Terra (10.30 h overpass time) is combined with MODIS Aqua (13.30 h

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overpass time) from 2002 onwards. Cloud droplet number concentration is derived from the MODIS retrievals as discussed in Grosvenor et al. (2018). For all three cloud quantities presented, only liquid-water clouds, as determined by the retrieval algorithm, are selected. Cloud droplet concentrations show declines especially over the oceans of the Northern hemisphere mid-latitudes, in particular downwind of the regions where aerosol emissions declined. The signal is much weaker over the continents, though (as also discussed by Ma et al., 2018). Cloud liquid water path (related to cloud thickness; cloudy- rather than all-sky) does not show trend patterns that would be strongly related to the pattern of trends in droplet concentration. It was documented earlier that the adjustment of liquid water path to cloud droplet concentration perturbation appears to be weak in comparison to natural variability (Malavelle et al., 2017; Toll et al., 2019; Haghighatnasab et al., 2022). In contrast, there are some hints at a change in cloud fraction consistent in pattern and sign with the trends in droplet concentration, as also suggested by satellite correlation studies (Gryspeerdt et al., 2016; Rosenfeld et al., 2019; Christensen et al., 2020). It is to be noted that cloud properties, especially outside the regions with strong aerosol changes, also respond to global warming (in particular, sea surface temperature trends under stratocumulus regions such as the Eastern Pacific) and natural variability.

Nevertheless, the conclusion of this review of trends in cloud quantities is that cloud droplet concentrations show trends that are spatially consistent with the expectation of declining anthropogenic aerosol emissions, and that also liquid-cloud fraction trends show pattern consistent with the aerosol declines. Since these retrievals are independent of the aerosol retrievals discussed earlier, this is a strong corroboration of the earlier conclusion that satellites show a declining trend in aerosols in regions of anthropogenic emissions.

5 Changes in radiation

Previous analysis of model simulations suggested that between 2000 and 2015, ERFaer was reduced in absolute magnitude, i.e. increased, by about 0.003 $\mathrm{W\,m^{-2}\,yr^{-1}}$ at a global scale (Myhre et al., 2017), mainly over the Northern hemisphere mid-latitudes, especially over North America, the North Atlantic ocean, Europe and adjacent Asia. In Fig. 4, the trends in ERFaer as simulated by models contributing to CMIP6 are analysed. This makes use of the dedicated simulations of the Radiative Forcing Model Intercomparison Project (RFMIP, Pincus et al., 2016) that trace the aerosol ERF over time (the piClim-histaer simulations). For seven Earth System Models, the relevant diagnostics for these simulations were submitted, namely for the one by the Canadian Centre for Climate Modelling and Analysis (CCMA, Swart et al., 2019), for the US National Aeronautics and Space Administration Goddard Institute for Space Studies Earth system model (GISS-E2-1-G, Kelley et al., 2020), for the UK Hadley Centre Global Environment Model (HadGEM3-GC31-LL, Andrews et al., 2019), for the Institut Pierre Simon Laplace Climate Model (IPSL-CM6A-LR, Boucher et al., 2020), for the Model for Interdisciplinary Research on Climate (MIROC6, Hajima et al., 2020), for the National Oceanic and Atmospheric Administration / Geophysical Fluid Dynamics Laboratory Climate Model (CM4, Held et al., 2019), and for the Norwegian Earth System Model (NorESM2-LM, Seland et al., 2020). The results show that the pattern in the clear sky solar ERFaer trends is closely related to the pattern in the trends in sulfate precursors. It reflects the strong declines in the main source areas over North America, Europe, and East Asia, along with the increases over India and surrounding areas. The patterns in all-sky ERFaer, i.e. including the cloud effects, both in solar and

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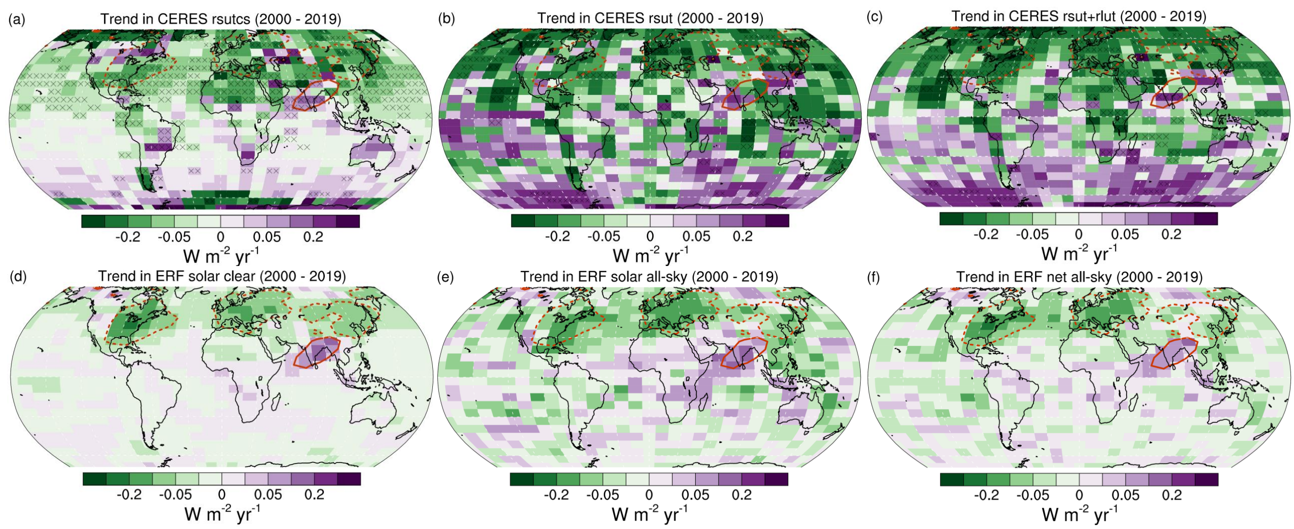

Figure 4. Linear trends (2000 to 2019) in (a) net broadband solar flux for clear-sky situations as retrieved from the Clouds and the Earth's Radiant Energy System (CERES) Energy Balanced and Filled (EBAF) product (Loeb et al., 2018a) from Terra, averaged for years after 2002 with retrievals also from Aqua; (b) and (c) as (a) but for all-sky solar radiation fluxes and all-sky solar plus terrestrial net fluxes, respectively; (d – f) effective radiative forcings due to aerosols as computed from the dedicated RFMIP (Pincus et al., 2016) / AerChemMIP (Collins et al., 2017) for which output was available from the CanESM5 (Swart et al., 2019), GISS-E2-1-G (Kelley et al., 2020), HadGEM3-GC31-LL (Andrews et al., 2019), IPSL-CM6A-LR (Boucher et al., 2020), MIROC6 (Hajima et al., 2020), NOAA-GFDL (Held et al., 2019), and NorESM2-LM (Seland et al., 2020) of which the ensemble average is shown for (d) clear sky solar, (e) all-sky solar and (f) all sky net (solar plus terrestrial) spectra. Signs are inverted for consistency with the CERES results (negative trends in ERFaer mean decreases in absolute magnitude). For the emissions, regions with small absolute trends (less than 7 μg m^{-2} day^{-1} yr^{-1}) are masked by grey shading. Isolines enclose regions with trends in clear-sky solar ERF (panel d) larger than 0.05 W m^{-2} yr^{-1} in absolute terms. The average values in these regions as listed in Table 1 and discussed in Section 7.

terrestrial spectra, are noisier but also show trends that are consistent with the pattern seen for the clear-sky, solar ERFaer and in aerosols and droplet concentrations also in the observations. In the multi-model mean, global mean changes show a decline of the clear-sky, solar ERFaer by 0.0117 W m^{-2} yr^{-1}, of the all-sky solar ERFaer a decline by 0.0172 W m^{-2} yr^{-1} and of the all-sky terrestrial ERFaer a (compensating) increase by 0.0013 W m^{-2} yr^{-1}. The integral, net decline over the 20-year period according to these models thus was year0.32 W m^{-2}. The IPCC AR6 assessment is based on multiple lines of evidence; the time series of the diagnosed ERFaer is available via the IPCC web site and at https://doi.org/10.5281/zenodo.5705391. Computing the linear trend between 2000 and 2019 on the basis of the emulator ensemble yields an increase by +0.0145 W m^{-2} yr^{-1} between 2000 and 2019 (5 to 95% confidence interval of +0.0068 to +0.0253), i.e. by +0.29 (+0.14 to +0.51) W m^{-2} over the full period (Gulev et al., 2021; Forster et al., 2021).

The ERFaer may be inferred from the Earth radiation budget which is measurable at the top of the atmosphere. Several studies have investigated the retrievals of this quantity from the Clouds and the Earth's Radiant Energy System (CERES)

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Energy instrument that is also on the EOS Terra and Aqua satellites. CERES shows patterns for clear-sky broadband radiation that are consistent with the aerosol spatio-temporal changes (Loeb et al., 2018b; Paulot et al., 2018; Loeb et al., 2021b). Further, Loeb et al. (2021a) document an increase in the Earth's energy imbalance, seen in both Earth radiation budget satellite observations and ocean heat content. It is split into a strongly decreasing trend in reflected solar radiation and a declining trend in emitted terrestrial radiation (defined positive downwards, so the trend implies more emission to space). Using partial radiative perturbation analysis, Loeb et al. (2021a) attribute the trend in solar radiation mostly to changes in clouds, with a very small contribution only due to the effect by aerosol-radiation interactions. This is also a result of a new study by Jenkins et al. (2022). CERES observations were also analysed by Raghuraman et al. (2021). They find for the period 2001 to 2020 an increasing trend by $0.038\pm0.024\,\mathrm{W\,m^{-2}\,yr^{-1}}$, which they attribute about one third of this trend to the reduction in aerosol ERF.

Kramer et al. (2021) disentangle the trends in satellite-retrieved radiation fluxes using radiative kernels, notably isolating the impact of radiative forcings. They quantify the change in absorbed solar radiation over the 2003 to 2018 period at $0.044\pm0.02\,\mathrm{W\,m^{-2}\,yr^{-1}}$. Singling out the instantaneous radiative forcing in the solar spectrum, they obtain a change of $0.006\pm0.003\,\mathrm{W\,m^{-2}\,yr^{-1}}$ which they largely attribute to aerosol changes. Paulot et al. (2018) constrained the radiative forcing due to aerosol-radiation interactions (aerosol direct effect) in the GFDL climate model obtained an almost negligible trend in ERFaer of $0.0002\,\mathrm{W\,m^{-2}\,yr^{-1}}$. Their study, however, considered the period from 2001 to 2015 only and thus a time when then increasing emissions over China were much more relevant.

Bellouin et al. (2020a) used the Copernicus reanalysis of atmospheric composition, which assimilates MODIS AODs, to estimate RFaer and found statistically significant decreasing (less negative) trends over North and South America, Europe, and China, and increasing (more negative) trend over India for the period 2003 to 2017. Their globally averaged trend in RFaer is $0.00\,\mathrm{W\,m^{-2}\,yr^{-1}}$, but limitations in their estimate may imply that the real trend is positive.

Surface measurements of radiation also show increasing trends over large regions (Wild, 2009, 2012; Cherian et al., 2014; Hatzianastassiou et al., 2020). Trends in aerosol effects become particularly apparent in surface radiation records under cloud-free conditions. Such records indicate in Europe increasing clear-sky surface solar radiation and thus decreasing aerosol effects throughout the 2000s with some tendency for saturation (leveling off) after 2010 (Manara et al., 2016; Wild et al., 2021). Surface radiation records in China suggest a trend reversal in clear sky surface solar radiation from decrease to increase in the late 2000s (Yang et al., 2019), in line with anthropogenic aerosol emission trends (Section 2). It has been shown for Europe (Pfeifroth et al., 2018) and China (Wang et al., 2019) that solar radiation consistently increases in both surface and satellite observations.

The CERES data are also shown in Fig. 4. The clear-sky solar radiation changes in the areas where the models show decreases in the clear-sky solar ERFaer, with a pattern consistent in sign and magnitude with the model results. For all-sky radiation, the data show larger trends and also much more noise in the patterns. However, the sign of the changes in the regions where an aerosol signal is expected is consistent between the models and the data.

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6 Ocean heat uptake and surface temperatures as constraints for the simulated ERFaer evolution

The temporal evolution of observed climate change – specifically, surface temperature changes and their pattern – has been proposed as a constraint on the magnitude of the aerosol effective radiative forcing (Ekman, 2014; Rotstayn et al., 2015; Stevens, 2015; Kretzschmar et al., 2017; Aas et al., 2019; Albright et al., 2021; Smith and Forster, 2021). Increasingly it is now recognized that the ocean heat uptake is of overwhelming interest for monitoring the Earth energy imbalance (von Schuckmann et al., 2016; Palmer, 2017; Allison et al., 2020; Forster et al., 2021), since it is a non-volatile indicator of climate change.

Based on this, Smith et al. (2021a) constrained the aerosol ERF from the CMIP6 models by considering the ocean heat uptake from observations between 1971 and 2018, in addition to observations of surface temperature. This study assessed the ERFaer between 1750 and 2019 at –0.9 $\mathrm{W\,m^{-2}}$ and suggested a slightly positive trend between 1980 and 2014 of 0.0025 $\mathrm{W\,m^{-2}\,yr^{-1}}$. The method of Smith et al. (2021a) is applied to assess the trend in aerosol ERF between 2000 and 2019 (Fig. 5). This yields a constrained trend of 0.0114 (–0.003 to 0.0274) $\mathrm{W\,m^{-1}\,yr^{-1}}$, much stronger than the one considering the longer period. The integral change in the 2000 – 2019 period in ERFaer is thus 0.23 $\mathrm{W\,m^{-2}}$ in the best estimate, very close to the suggestion by the analysed models.

Using a Bayesian method, Albright et al. (2021) made use of the observed temperature changes to constraining simulated ERFaer. In Fig. 5 their method is applied to the period investigated here, from 2000 to 2019. This yields a mean trend of 0.0047 $\mathrm{W\,m^{-2}\,yr^{-1}}$ (5 to 95% confidence interval of -0.000912 to 0.0106 $\mathrm{W\,m^{-2}\,yr^{-1}}$). The best estimate of the change in ERFaer for the 20-year-period thus is 0.094 $\mathrm{W\,m^{-2}}$, but the confidence interval includes the 0.2 $\mathrm{W\,m^{-2}}$ from the two other estimates, i.e. this estimate is lower but consistent with the other estimates to within the uncertainty range. They, however, caution that using ocean heat content observations to constrain aerosol forcing requires a better understanding of the relationship between time-variable radiative feedbacks and radiative forcing. A weaker top-of-atmosphere radiative imbalance can be explained by more negative aerosol forcing, or alternatively by stronger radiative damping.

Jenkins et al. (2022) also suggests that 0.2 $\mathrm{W\,m^{-2}}$ is a plausible best-estimate ERF change for the considered period, based on analysis of satellite observations and energy balance of global temperatures. However, they also conclude that large variability signals should mean a very weak trend change cannot be ruled out either.

7 Discussion and conclusions

The trends of aerosols, clouds and radiation in the observations are subject not only to changes in anthropogenic aerosol emissions, but also to other influencing factors. These include changes in natural aerosol emissions, which remain poorly constrained and contribute a substantial fraction of total observed AOD, but also interannual variability, and responses to greenhouse-gas-induced global warming; aerosol-cloud interactions may also be altered in a changing climate (Murray-Watson and Gryspeerdt, 2022).

Natural aerosol emissions, especially of dust, are highly variable and impact the distribution of AOD in specific regions (Chin et al., 2014). Natural aerosol emissions may respond to increasing temperatures (Yli-Juuti et al., 2021). Also volcanic aerosol emissions, both from eruptions and from degassing, are an important contribution in particular to atmospheric sulfate aerosol.

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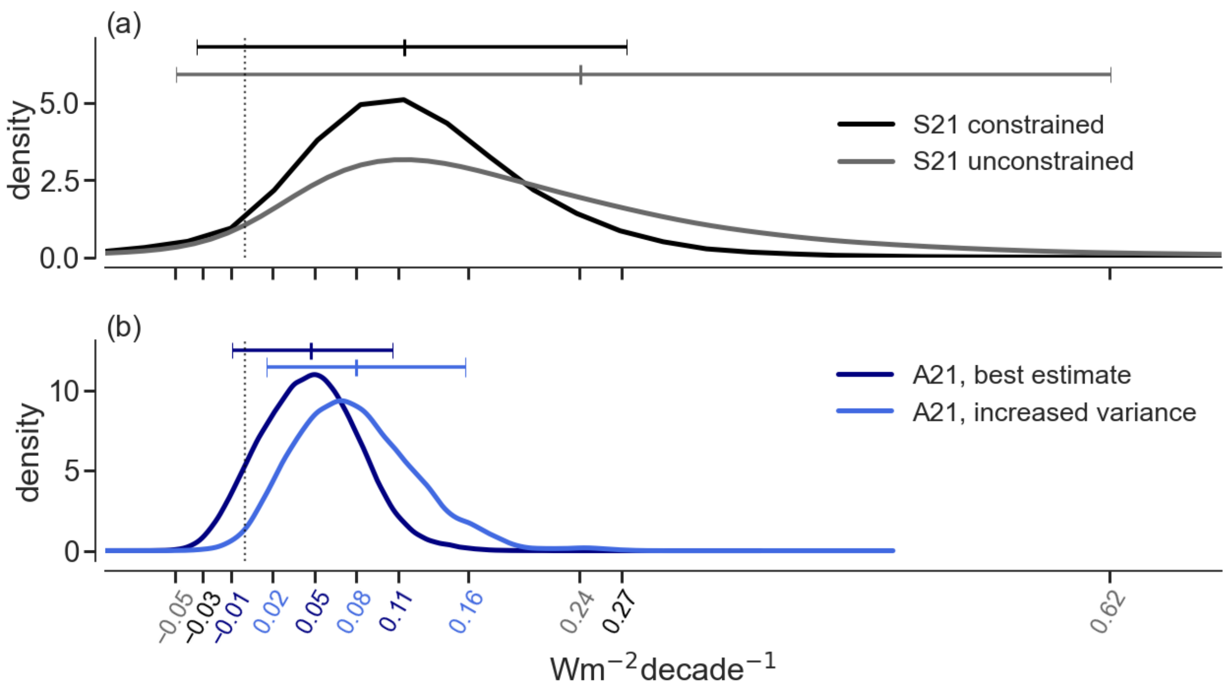

Figure 5. Assessment of the linear trend in ERFaer between 2000 and 2019. (a) as in Smith et al. (2021a, their Fig. 7; abbreviated as S21 in the labels), and (b) as in Albright et al. (2021, labelled A21). The constraint is as in the cited studies. Increased variance in Albright et al. (2021) corresponds to a scenario prescribing internal climate variability that is a factor of five larger than the CMIP6 mean and assuming large, correlated errors in global temperature observations, yielding a fifth-percentile ERFaer lower bound around -2.0 W m^{-2}.

However, in the past 20 years, no strong trends have been observed (Carn et al., 2017). Wild fires may emit large amounts of aerosols. This was in particular the case for the Australian bush fires in 2020 (Boer et al., 2020; Heinold et al., 2021). Whether there were substantial trends in wild fire aerosol emissions in recent decades is not quite clear (Doerr et al., 2016), even if global warming increases the risk of fire (van Oldenborgh et al., 2021). Sea salt aerosols, that are a function of near-ocean-surface wind speed, are subject to variability, both forced and unforced, albeit to a lesser extent than dust (Stier et al., 2006). To the extent that the MODIS, rather than MISR, AOD and AODFM trends above the southern hemisphere oceans are right, such variability in sea salt aerosol may cause the increasing trends (Struthers et al., 2013). Trends in long-range transport of aerosols could be another reason for such increases. However, the satellite retrievals are particularly uncertain in this region, due to the large zenith angles and large cloud cover that both hamper aerosol retrievals.

Towards the end of the time series investigated here, there were specific effects due to the COVID-19 pandemic in 2020 (Forster et al., 2020; Gettelman et al., 2021; Fiedler et al., 2021). For this reason, and due to the particularly large fire activity, the end year of the data analysed here was chosen as 2019.

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(a) SO_2 emissions (% yr^{-1})		(b) OC emissions (% yr^{-1})		(c) BC emissions (% yr^{-1})	
-4.71	+3.22	-0.67	+0.55	-1.44	+1.09
(d) MISR AOD (% yr^{-1})		(e) MISR AODFM (% yr^{-1})			
-1.15	+1.35	-1.66	+1.34		
(f) MODIS AOD (% yr^{-1})		(g) MODIS AODFM (% yr^{-1})		(h) PMAp AOD (% yr^{-1})	
-0.85	+1.74	-0.88	+2.36	-0.44	-0.04
(i) N_d (% yr^{-1})		(j) LWP (% yr^{-1})		(k) Cloud fraction (% yr^{-1})	
-0.43	+0.07	-0.16	-0.11	-0.16	+0.11
(l) CERES rsutcs ($W\,m^{-2}\ yr^{-1}$)		(m) CERES rsut ($W\,m^{-2}\ yr^{-1}$)		(n) CERES net ($W\,m^{-2}\ yr^{-1}$)	
-0.10	+0.04	-0.19	+0.04	-0.23	+0.03
(o) ERF SW clr ($W\,m^{-2}\ yr^{-1}$)		(p) ERF SW ($W\,m^{-2}\ yr^{-1}$)		(q) ERF net ($W\,m^{-2}\ yr^{-1}$)	
-0.09	+0.10	-0.10	+0.09	-0.08	+0.07

Table 1. Mean values derived from the maps in Fig. 1 to 4, averaged over (first value) the regions with substantial negative trends (defined as larger than 0.05 $W\,m^{-2}\,yr^{-1}$ in absolute terms; isolines in Fig. 4) in ERF clear sky, solar from CMIP6 (Fig. 4d) and (second value) substantial positive trends. The regions with negative trends cover 7.3% of the Earth surface, the ones with positive trends, 1.1%. AODFM is retrieved only over oceans, and the PMAp time series only spans ten years from 2008 to 2017.

IPCC AR6	+0.29 (+0.14 to +0.51) $W\,m^{-2}$
Method of Smith et al. (2021), constraint from ocean heat uptake	+0.23 (-0.05 to 0.55) $W\,m^{-2}$
Method of Albright et al. (2021), constraint from surface temperature changes	+0.094 (-0.02 to 0.21) $W\,m^{-2}$
RFMIP models (Fig. 4)	+0.32 $W\,m^{-2}$
Kramer et al. (2021)	+0.12 $W\,m^{-2}$
Raghuraman et al. (2021)	+0.24±0.20 $W\,m^{-2}$

Table 2. Estimates of ERFaer change between 2000 and 2019. The 5 to 95% uncertainty ranges are provided. Kramer et al. (2021) assess RF due to aerosols and use the period 2003 to 2018; Raghuraman et al. (2021) the period 2001 to 2019.

The different quantities investigated here are not independent. The climate models are driven by the emissions; and the emissions inventories, in turn, consider aerosol satellite retrievals. The satellite retrievals of cloud properties and radiation are not linked to the other quantities but are more noisy in their results. Cloud properties respond to variability in climate dynamics, both forced and unforced, beyond the impact of anthropogenic aerosols. Norris et al. (2016) document global patterns of changes in cloud coverage and cloud albedo that show a reduction in cloud cover and albedo in the mid-latitudes and an increase in the Tropics from the 1980s to the first decade of the 21st century, and that are consistent with the expectations due to cloud responses to global warming.

We now turn to discussing quantifying the changes in aerosols, clouds, and radiation. For this, the regions with clear trends in aerosols are identified by subjectively choosing the regions in which the ERF simulated by the CMIP6 models (Fig. 4) exceeds $\pm 0.05\,\mathrm{W\,m^{-2}\,yr^{-1}}$ for the solar, clear sky component. Regions with increasing and decreasing ERF are distinguished. Table 1 summarizes all quantities analysed in Fig. 1 to 4. In the regions with declining clear-sky solar ERFaer, in particular SO_2 emissions decreased strongly, but also OC and BC emissions decrease according to the inventory of Hoesly et al. (2018). In regions with increasing clear-sky solar ERFaer, emissions of all three aerosol species increased. Both MODIS and MISR show corresponding declining trends in column-aerosol metrics for the regions with aerosol reductions, and increasing trends where aerosols increased. The numbers are much larger for MISR than for MODIS for the declining-trend regions (almost a factor of 2 larger in case of AODFM). Cloud responses in these regions are consistent with the expectations. Droplet number concentrations decrease, although by a rate that is a factor of 2 (compared to MODIS AOD) to 4 (compared to MISR AODFM) less than for aerosol optical depth, highlighting that there is not a 1:1 relationship of droplet number and aerosol measured as AOD (e.g., Quaas et al., 2020; Jia et al., 2022). There is only a small LWP response that is inconsistent in sign between regions with increasing and decreasing aerosol emissions. LWP does not just respond to N_d perturbations, but also to global warming (with an expected increase in LWP on average; e.g., Norris et al., 2016). However, the fact that there is little LWP trend where N_d trends are substantial is consistent with other observations-based assessments (Malavelle et al., 2017; Toll et al., 2019). Cloud fraction, in turn, does show decreasing trends in the regions with anthropogenic aerosol decreases, and increasing trends where aerosol increases. Although it is also a function of other drivers, this could be a hint at a systematic (positive, i.e. negative in terms of forcing) aerosol effect on cloud fraction as also documented in earlier statistical studies (Gryspeerdt et al., 2016; Rosenfeld et al., 2019). Consistent with these results, CERES shows decreasing trends in top-of-atmosphere radiation budget. The numbers are stronger for all-sky than for clear-sky, indicating a comparatively strong contribution by aerosol-cloud interactions (Forster et al., 2021; Loeb et al., 2021a). The numbers are consistent in sign with what the CMIP6 models suggest for changes in ERFaer, although more negative (less positive where aerosols increase) in particular when the cloud effects are included.

In conclusion, there are clear, robust and consistent signals for net declining anthropogenic aerosol influence on climate in the period since 2000, i.e. the period, for which high-quality satellite retrievals of all relevant quantities are available. The regions in which aerosol emissions declined (in particular North America, Europe and East Asia) dominate over regions with increasing trends. The overall climate-relevant signal is a decline in negative aerosol effective radiative forcing by about 0.1 to $0.3\,\mathrm{W\,m^{-2}}$, i.e. between 15 and 50% of the $0.6\,\mathrm{W\,m^{-2}}$ increase in CO_2 ERF (Forster et al., 2021) in the same time period. This signal will very likely continue in the future, increasing the urgency for strong measures on reducing greenhouse gas emissions (McKenna et al., 2021).

Data availability. The MODIS cloud products MYD08_D3 from Aqua and MOD08_D3 from Terra were used in this study from the Atmosphere Archive and Distribu- tion System (LAADS) Distributed Active Archive Center (DAAC), https://ladsweb.nascom.nasa.gov/. MISR data were obtained from the NASA Langley Research Center Atmospheric Science Data Center (https://opendap.larc.nasa.gov/opendap/

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MISR/MIL3YAEN.004). CERES data were obtained from https://ceres.larc.nasa.gov/data/. The MetopB data are available as PMAp Climate Data Record (CDR) at https://doi.org/10.15770/EUM_SEC_CLM_0053. AERONET data were used from https://aeronet.gsfc.nasa.gov/data_push/AOT_Level2_Monthly.tar.gz. RFMIP model output is available from the Earth System Grid Federation (ESGF).

Author contributions. The fundamentals stem from active discussions with all authors. J.Q. coordinated the study and led the writing of the manuscript with significant contributions from all authors. H.J. created Fig. 1 to 4, including processing the data, and Table 1. with substantial help from C.S. for the RFMIP models and M.D.-B. for the MetopB data. A.-L.A. and C.S. prepared Fig. 5 and the data processed for it.

Competing interests. The authors declare there are no competing interests.

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