

Aerosol radiative effects with MACv2

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Abstract

Monthly global maps for aerosol properties of the MACv2 climatology are applied in an off-line radiative transfer model to determine aerosol radiative effects. This model-setup cannot address rapid adjustments by clouds, but current evidence suggests their contribution to be small when compared to the instantaneous radiative forcing. Global maps are presented to detail the regional and seasonal variability associated with (annual) global averages. Radiative effects caused by the aerosol presence (direct effects) and by aerosol modified clouds (indirect effects) are examined. Direct effects are determined for total aerosol, anthropogenic aerosol and extracted individual aerosol components. Indirect effects cover the impact of reduced cloud drop sizes by anthropogenic aerosol.

Present-day global annual radiative effects for anthropogenic aerosol yield (1) a climate cooling of -1.0 W/m^2 at the top of the atmosphere (TOA). (2) a surface net-flux reduction of -2.1 W/m^2 and (by difference, 3) an atmospheric effect of a $+1.1 \text{ W/m}^2$. This atmospheric solar heating is almost entirely a direct effect. On a global basis, indirect effects (-0.65 W/m^2) dominate direct effects (-0.35 W/m^2) for the present-day climate response at the TOA, whereas the present-day surface radiative budget is stronger reduced by direct effects (-1.45 W/m^2) than by indirect effects (-0.65 W/m^2).

Natural aerosols are on average less absorbing and larger in size. However, their stronger solar TOA cooling efficiency is offset by a non-negligible infrared (IR) greenhouse warming efficiency. In the sum the global average annual direct forcing efficiencies (per unit AOD) for natural and anthropogenic aerosol are similar: $-12 \text{ W/m}^2/\text{AOD}$ at all-sky conditions and $-24 \text{ W/m}^2/\text{AOD}$ at clear-sky conditions.

The present-day direct TOA impact by all soot (BC) is globally averaged $+0.55 \text{ W/m}^2$. Between $+0.25$ to $+0.45 \text{ W/m}^2$ of that can be attributed to anthropogenic sources, depending on assumptions for the pre-industrial BC reference state. Similarly, the pre-industrial fine-mode reference uncertainty has a strong influence not just on the direct but even more on the indirect effect. Present-day aerosol TOA forcing is estimated to stay within the -0.7 to -1.6 W/m^2 range (with the best estimate at -1.0 W/m^2).

Calculations with scaled temporal changes to anthropogenic AOD from global modeling indicate that the global annual aerosol forcing has not changed much over the last decades, despite strong shifts in regional maxima for anthropogenic AOD. These regional shifts explain most solar insolation (brightening or dimming) trends that have been observed by ground-based radiation data.

1. Introduction

Atmospheric aerosol modulates the radiative energy budget directly (by the aerosol presence) and indirectly (by modifying the properties of clouds). Such impacts are of interest for climate change predictions, because part of today's atmospheric aerosol is anthropogenic. A quantification of aerosol impacts on global scales, however, is difficult. Tropospheric aerosol is highly variable in space and time and the needed pre-industrial reference for anthropogenic impacts is poorly defined. The determination of aerosol impacts requires two simulations: one with aerosol and one with less or no aerosol. Usually

1 complex ‘bottom-up’ simulations with global models are applied, in which emissions of different aerosol
 2 sources are chemically and/or cloud processed, mixed, transported and removed. Further assumptions
 3 to size and water uptake are needed to determine associated aerosol optical properties. These in turn
 4 are needed to estimate aerosol radiative impacts in broadband radiative transfer applications. This path
 5 involves many uncertainties. And many repeated simulations are generally needed to constrain natural
 6 variability (mainly by clouds). These ‘bottom-up’ simulations are essential to account for feedbacks and
 7 delayed or spatially detached responses in the climate system. Fortunately, for aerosol radiative impacts
 8 feedbacks are secondary in strength (*Fiedler et al. 2019*). Thus, off-line radiative transfer applications to
 9 determine an instantaneous impact via a dual call mode seem sufficient. Hereby, selected properties of
 10 aerosol (for direct radiative impacts) and/or of (aerosol modified) clouds (for total / indirect radiative
 11 impacts) are modified with respect to a reference simulation. And baseline properties for aerosol and its
 12 environment are prescribed by monthly global maps linked to observational data. Aerosol properties are
 13 prescribed by the MACv2 aerosol climatology (*Kinne, 2019*). MACv2 defines global (monthly, 1x1deg
 14 gridded) maps for aerosol optical and radiative properties. Aerosol component detail is derived in a
 15 (reverse processing) ‘top-down’ approach to define the spectrally resolved aerosol single scattering
 16 properties (needed for broadband radiative transfer). By prescribing optical properties for aerosol and
 17 clouds (with strong links to observations) simulated aerosol radiative impacts are faster, more precise
 18 and more direct than with ‘bottom-up’ approaches. First the assumed MACv2 and environmental
 19 properties are presented. Then the applied radiative transfer scheme is outlined. And finally radiative
 20 impacts are presented for total and anthropogenic aerosol - also as a function of time.

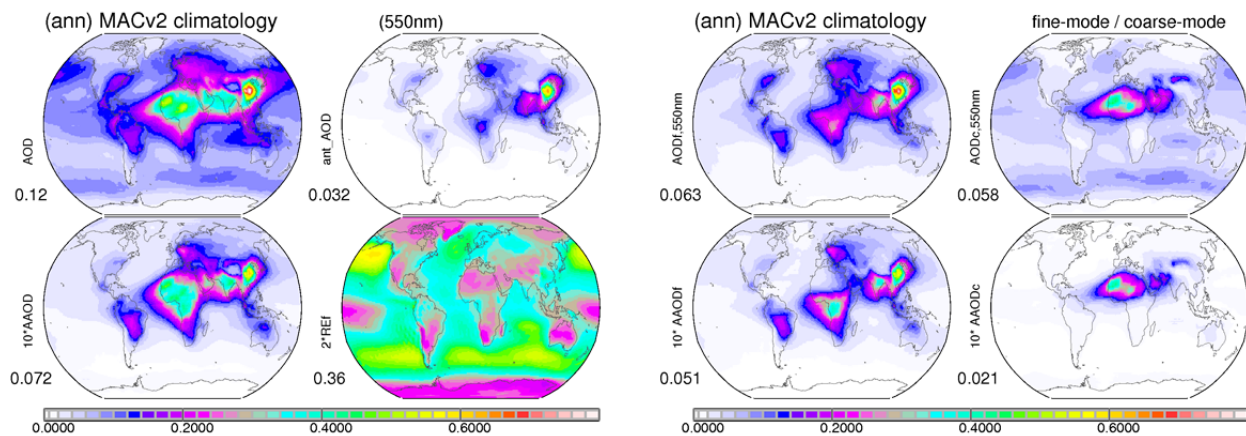
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22 2. MACv2 Aerosol Properties

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24 The Max Planck institute Aerosol Climatology (MAC) in its second version (*Kinne, 2019*) defines
 25 spectrally resolved monthly global fields for aerosol properties with global coverage (at a 1x1 degree
 26 lat/lon spatial resolution). Annual averages of defining MACv2 aerosol properties are shown in Figure 1.

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29 **Figure 1.** annual average maps of the MACv2 aerosol climatology. Global distributions are presented for
 30 present day column properties of aerosol amount (AOD), absorption (AAOD*10), anthropogenic AOD and
 31 fine-mode effective radius (REf*2) in μm . Also presented are the mid-visible AOD and AAOD split into
 32 fine-mode (radii $<0.5\mu\text{m}$) and coarse mode (radii $>0.5\mu\text{m}$) contributions. Labels indicate global averages.

1 These global fields are the result of a data merging process for mid-visible (at 550nm) properties
 2 of aerosol optical depth (AOD, column amount) and absorption aerosol optical depth (AAOD, column
 3 absorption). This merging was done separately for fine-mode (radii <0.5µm) sizes (AODf, AAODf) and
 4 coarse-mode (radii >0.5µm) sizes (AODc, AAODc), as well as for the fine-mode effective radius (REf). In
 5 the merging process regional distributions of monthly background maps are adjusted based on local
 6 matches to trusted monthly statistics by ground based solar photometry observations. Background
 7 maps in MACv2 are median properties of 14 different global models with detailed aerosol modules
 8 (Kinne *et al.*, 2006) in the framework of AeroCom phase 1 simulations. Applied photometry monthly
 9 statistics are sun- and sky-samples by CIMEL instruments of the AERONET network (Holben *et al.* 2001,
 10 Dubovik *et al.*, 2002) and sun-samples of by-hand-operated MICROTOPS instruments of the Marine
 11 Aerosol Network, MAN (Smirnov *et al.*, 2009). The needed spectrally varying single scattering properties
 12 for broadband radiative transfer simulations are set by local mixtures of spectrally predefined (via
 13 refractive index and size) aerosol components. Local component mixture weights are chosen such that
 14 their combined local AODf, AODc, AAODf, AAODc and REf values are consistent with MACv2 data.

15 In MACv2 on a global annual basis about 50% of the total AOD at 550nm (of 0.12) each is
 16 contributed by fine-mode and coarse-mode sizes, whereas the total absorption AAOD at 550nm (of
 17 0.0072) has at stronger fine-mode contributions, at 70%. Still, the other 30% contributions by coarse-
 18 mode sizes to absorption are significant and are associated with larger mineral dust sizes near sources.

19 Figure 1 also presents MACv2 estimates for the distribution of anthropogenic AOD at present-
 20 day conditions (at the mid-visible wavelength of 550nm). Anthropogenic AOD in MACv2 allows only
 21 contributions by smaller fine-mode (radii <0.5µm) aerosol sizes, with major contributions from pollution
 22 and fires. The anthropogenic AOD is determined by applying to the AODf map of MACv2 (local, monthly)
 23 scaling factors $[(AODf, pd - AODf, pi) / AODf, pd]$ based on AeroCom phase 2 simulations with CMIP5
 24 present-day (pd) and pre-industrial (pi) emissions (Lamarque *et al.*, 2010). Anthropogenic coarse-mode
 25 (radii >0.5µm) contributions (e.g. due to land-use change) are ignored, because (1) its AOD is relatively
 26 small (see Figure E1 in Appendix E), (2) its solar and IR forcing effects largely cancel each other (as shown
 27 later) and (3) increases to aerosol concentrations via the coarse-mode (for indirect effects) are minor.

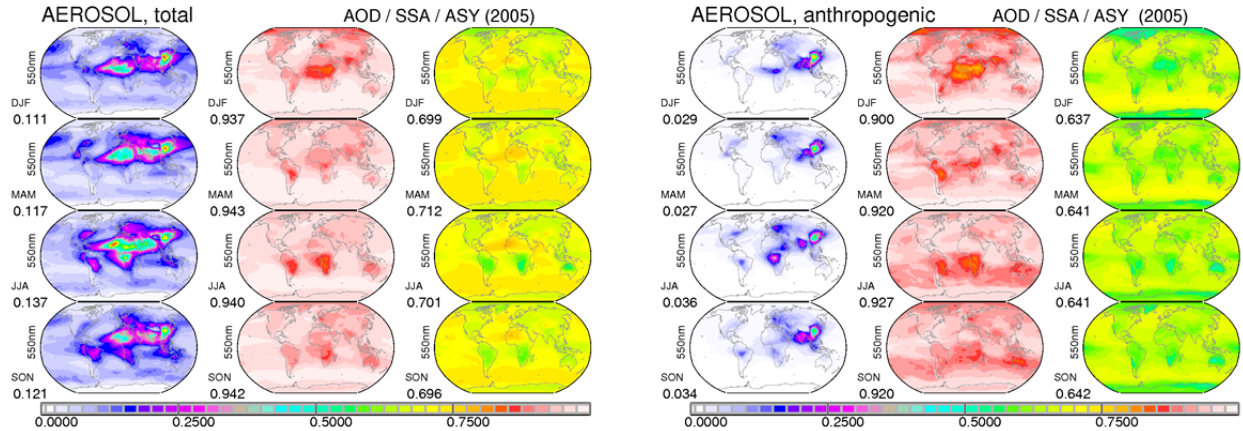
28 For radiative transfer simulations three wavelength dependent aerosol radiative properties are
 29 needed: aerosol optical depth (AOD), single scattering albedo (SSA) and asymmetry-factor (ASY).
 30 Present-day global annual averages of these MACv2 aerosol radiative properties are summarized in
 31 Table 1 for four selected wavelengths. Corresponding seasonal maps in the mid-visible spectral region
 32 (at 550nm) for total aerosol and anthropogenic aerosol radiative properties are presented in Figure 2.

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35 **Table 1** global annual averages of radiative properties for present-day (tropospheric) MACv2 aerosol

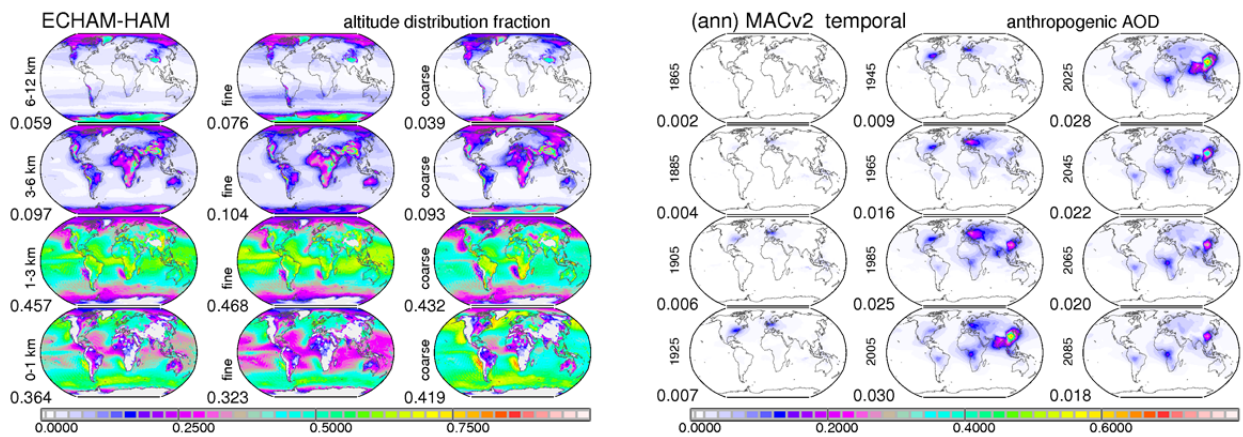
$\lambda(\mu\text{m})$	AOD				SSA			ASY		
	<i>total</i>	<i>coarse</i>	<i>fine</i>	<i>anthr*</i>	<i>total</i>	<i>coarse</i>	<i>fine</i>	<i>total</i>	<i>coarse</i>	<i>Fine</i>
.45	0.144	0.058	0.087	0.043	0.902	0.905	0.900	0.718	0.789	0.670
.55	0.122	0.058	0.063	0.032	0.941	0.964	0.919	0.702	0.767	0.639
1.0	0.081	0.062	0.019	0.009	0.956	0.982	0.870	0.693	0.736	0.533
10	0.049	0.049			0.580	0.560		0.605	0.605	

36 * anthropogenic SSA and ASY in MACv2 are those of the fine-mode



1
2 **Figure 2.** seasonal maps of MACv2 associated present-day mid-visible (550nm) radiative (or single
3 scattering) properties (AOD, SSA, ASY) for total aerosol (left block) and for anthropogenic aerosol (right
4 block). Global averages for each season are indicated below the labels.

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7 In simulations of aerosol radiative effects the relative aerosol altitude distribution with respect
8 to clouds is important. In MACv2 an AOD fractional scaling is applied based on multi-year simulations
9 with the ECHAM-HAM global model (Zhang *et al.*, 2012). This scaling is done independently for fine-
10 mode AOD (AODf) and coarse-mode AOD (AODc). With the AOD scaling by size modes, a vertical scaling
11 for SSA and ASY seems secondary and is ignored. For anthropogenic aerosol only the fine-mode vertical
12 scaling is relevant, because anthropogenic aerosol in MACv2 has only contributions by fine-mode size
13 aerosols. AOD, AODc and AODf distributions with four altitude regions are illustrated in Figure 3.



16
17 **Figure 3** annual maps for fractional contributions of total, fine-mode and coarse mode AOD (left block)
18 for four altitude regimes (0-1, 1-3, 3-6, >6km above sea-level) and for anthropogenic AOD (550nm) for
19 different years from 1865 to 2085 (right block). Values below the label indicate global averages.

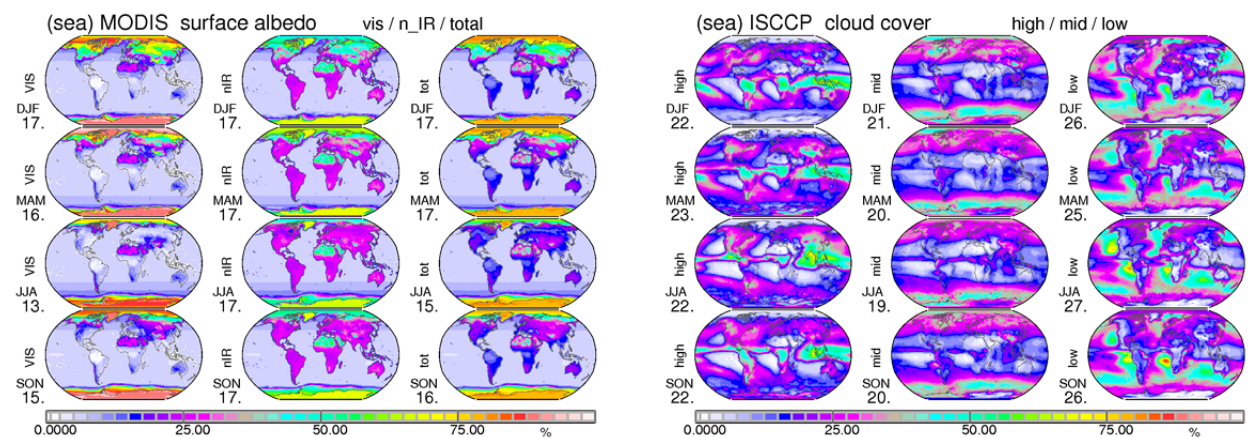
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22 Figure 3 also presents time-slices (1865, 1885, 1905, 1925, 1945, 1965, 1985, 2005, 2025, 2045,
23 2065, 2085) for the anthropogenic AOD (antAOD). Temporal changes in anthropogenic AOD are based

1 on local scaling factors (antAOD,year/antAOD,pd) by ECHAM-HAM (Stier et al., 2006, Zhang et al. 2012)
 2 simulations with temporally changing emissions according to historic data and future projections.

3 The resulting anthropogenic AOD maps in Figure 3 display globally highly uneven distributions.
 4 From the late 19th century into the 20th century anthropogenic AOD steadily increased over Europe and
 5 the eastern US. Only since the middle of the 20th century also other continental regions started to
 6 display anthropogenic AOD. Particularly strong were increases over SE Asia. Over the last three decades
 7 (since 1985) the anthropogenic AOD maxima declined over Europe and the eastern US and maxima over
 8 SE Asia and more recently over S Asia began to dominate. With the shift in regional maxima the present-
 9 day global average anthropogenic AOD appears to have reached a plateau, which is probably a
 10 maximum, as future emission scenarios suggest a decline in global anthropogenic AOD.

12 3. Environmental Properties

14 The environmental properties in radiative transfer simulations are represented by monthly
 15 averages to describe seasonal variations in solar insolation, atmospheric state and properties of clouds
 16 and the surface. Monthly mean of the sun's latitudinal position define variations in TOA solar irradiance.
 17 Surface temperatures (Hansen et al., 2010) via multi-annual monthly local averages help in choices for
 18 standard atmospheric profiles (Andersen et al., 1986; Mc Clatchey et al, 1972). Local combinations of
 19 these standard profiles define local monthly atmospheric state and trace-gas concentrations. For clouds
 20 multi-annual monthly average ISCCP data (Rossow et al., 1993) define scene optical depth and the cloud
 21 cover at low (>680hPa), mid and high (<440hPa) altitudes. For surface properties an IR emittance of 0.96
 22 is assumed. Surface solar albedo data over land are based on MODIS sensor data for the visible (UV/VIS)
 23 and near-infrared (n-IR) spectral regions (Schaaf et al., 2002). Over oceans, solar albedo data account for
 24 a solar elevation dependence (Taylor et al., 1996). Seasonal means of the applied VIS and nIR solar
 25 surface albedo and the ISCCP cloud cover by altitude are presented in Figure 4.



28 **Figure 4** Seasonal averages for applied environmental properties to (1) solar surface albedo (left block)
 29 for the UV/VIS, the near-IR and the total spectral range based on MODIS satellite sensor data and (2)
 30 ISCCP based cloud cover (right block) for high (<440hPa), mid (440-680hPa) and low (>680hPa) altitudes.
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4. Radiative Transfer Method

The atmospheric radiative transfer simulations apply a two stream radiative transfer scheme (Meador and Weaver, 1980). Spectral variability is captured by eight solar and twelve infrared bands with a total of 120 exponential terms to represent atmospheric trace-gas absorption. Vertical variability of the atmosphere is represented by 20 plane parallel homogenous layers and atmospheric state and trace-gas properties are defined via standard atmospheres (Mc Clatchey et al, 1972). Independent simulations at each (of the 64800) grid locations are performed for each month with monthly averages. Hereby, daily average solar radiative effects are based on weighted individual simulations at nine different solar zenith angles. Simulations with (ISCCP) clouds always involve simulations at all eight possible permutations for high-, mid- and low- altitude cloud combinations, assuming random overlap and for satellite view corrected local cloud cover data in the three altitude regimes.

Radiative impacts in the atmosphere are defined by differences of two simulations between a modified and a standard configuration. These ‘dual-call’ radiative transfer applications investigate

- **extra aerosol presence** (*direct effect - with a focus on individual aerosol components*),
- **reduced water droplet radii** due to extra aerosol (*first indirect effect*) and
- **combination of both effects**.

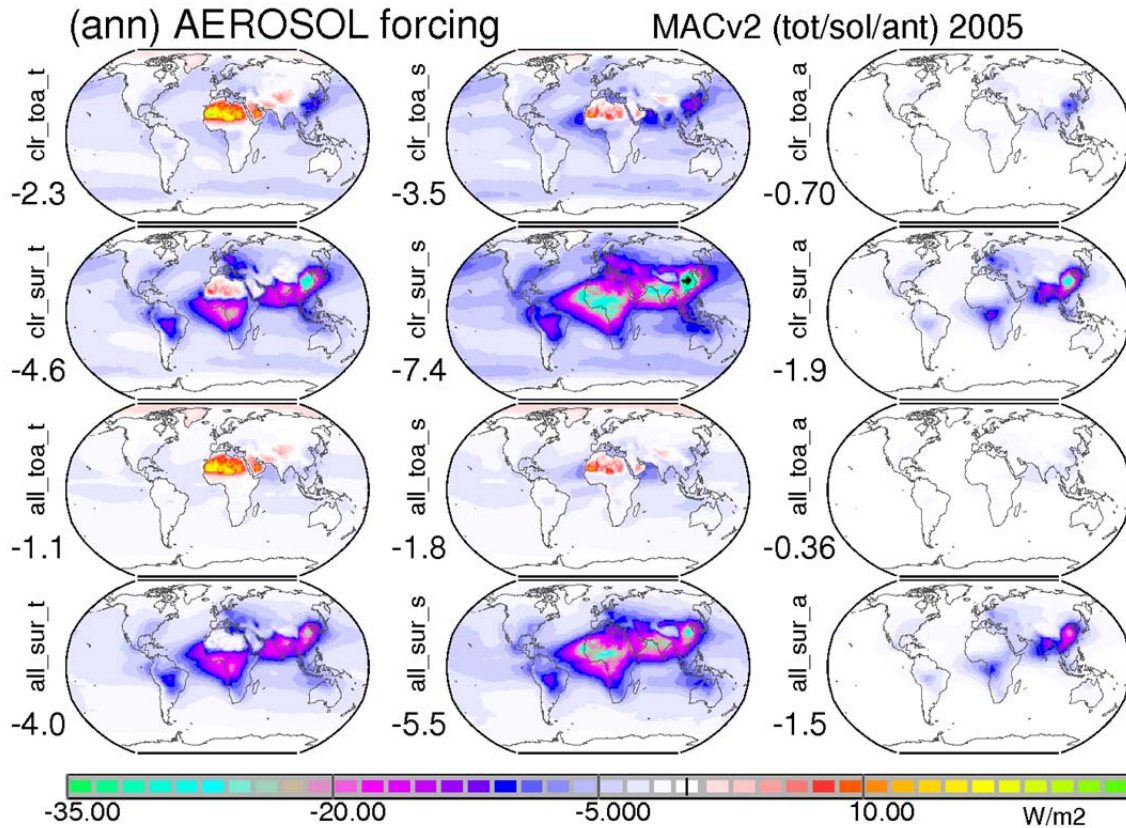
The resulting changes to broadband solar and infrared radiative net-fluxes are particularly relevant

- **at the top of the atmosphere (TOA)** - for the overall climate impact,
- **at the surface** - for impacts on surface processes and
- **in the atmosphere** (by TOA minus surface impact differences) - for impact on dynamics.

Dual-call radiative transfer cannot consider climate feedbacks. Long-term Earth System Model (ESM) simulations with a fixed sea-surface temperature (SST), however, indicated that atmospheric feedbacks (Fiedler et al, 2017, Fiedler et al, 2019 - there referred to as ‘rapid adjustments’) modulate the aerosol radiative impacts at most on the order of 10%. Thus by ignoring radiative forcing feedbacks, no major extra errors are introduced. It also should be pointed out that a dual-call scheme offers more precise answers, as internal (cloud) variability of independent ESM simulations is avoided (Fiedler et al, 2019).

5. Direct Aerosol Effects

Aerosol direct radiative effects are changes to the atmospheric energy distribution from the aerosol (or the extra aerosol) presence. These effects are quantified by the difference of one simulation with all aerosols and one simulation with no or less aerosol. The anthropogenic aerosol impact, for instance is defined by the difference of radiative effects of a simulation with present day (pd) and pre-industrial (pi) conditions. Only coarse mode aerosol sizes are large enough for infrared radiative effects. As only smaller fine-mode aerosols contribute to the anthropogenic AOD in MACv2, only solar impacts for anthropogenic aerosol need to be investigated. Annual average maps of present day direct aerosol radiative effects are summarized in Figure 5 for cloud-free conditions (clear-sky, ‘clr’) and with tropospheric clouds (all-sky, ‘all’) - for both total and anthropogenic aerosol at TOA and surface.



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2 **Figure 5** Annual maps for direct radiative effects of present-day total aerosol (left column), its solar
3 effect (center column) and of present-day anthropogenic aerosol. Clear-sky effects are presented in the
4 upper two rows: at the TOA (row 1) and at the surface (row 2). All-sky effects (with ISCCP clouds) are
5 presented in the lower two rows: at the TOA (row 3) and at the surface (row 4). Note, values for the
6 anthropogenic direct forcing (column 3, row3) are relatively small. More detail is given in Figure 6. Blue
7 colors indicate radiative net-flux losses (or a 'cooling'), and red colors indicate radiative net-flux gains (or
8 a 'warming'). Values below the labels indicate global averages.

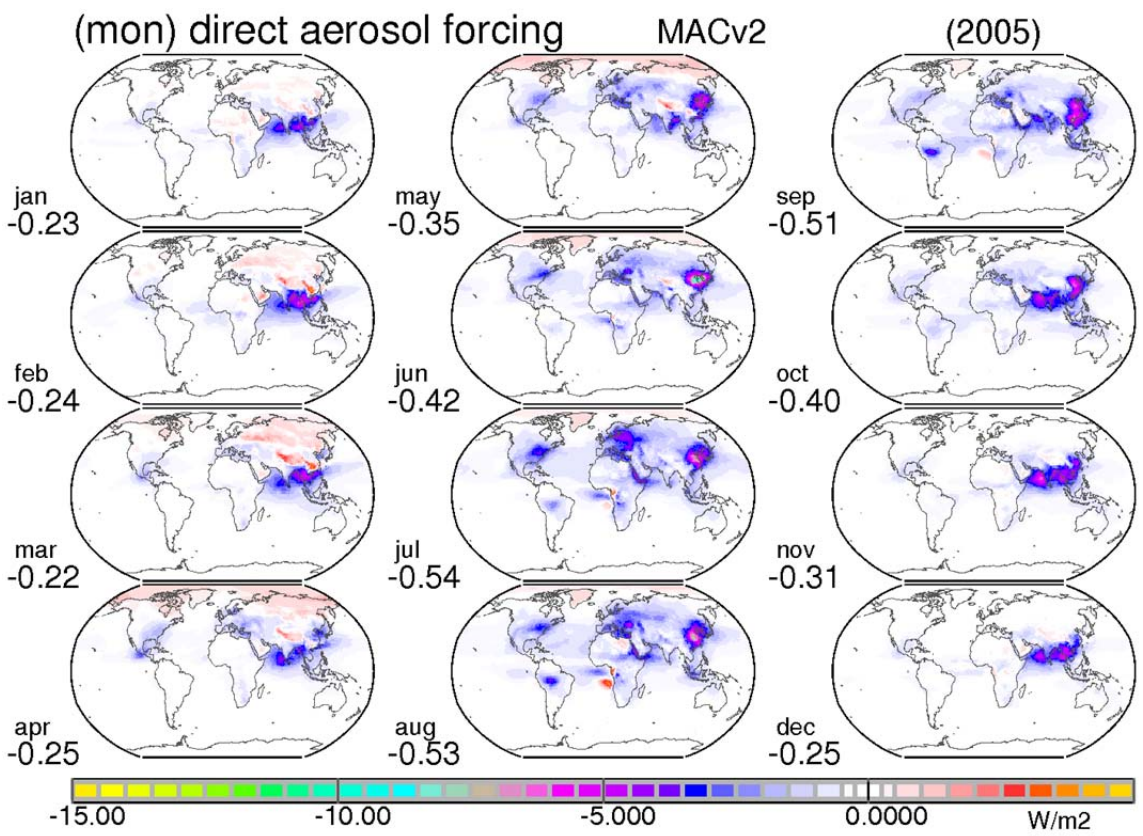
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11 For the aerosol associated net ('down' minus 'up')-flux changes in Figure 5, negative values
12 (blue to purple to light-blue) indicate regions with tendency to 'cool', whereas positive values (red to
13 yellow) display regions that experience a tendency to 'warm'. Global averages for all maps of Figure 5
14 are negative. Thus present day total (left column in Figure 5) and anthropogenic aerosol (right column in
15 Figure 5) on average cool. Hereby global averages of aerosol radiative effects are more negative at the
16 surface than at the TOA, because some aerosol types also absorb in the atmosphere. Radiative effects
17 are usually more negative at clear-sky than at all-sky conditions, because reflecting clouds above aerosol
18 prevent aerosol interactions with solar radiation. This 'clear-sky' minus 'all-sky' difference is much larger
19 at TOA than at the surface, because elevated absorbing aerosols above clouds dim the reflection of
20 clouds to space for a relative warming. All, direct radiative effects spatially unevenly distributed.

21 For total aerosol (in the left column of Figure 5) the negative forcing in most regions is partially
22 offset by regional warming over the N. Africa and Arabia, especially for TOA effects. To understand this
23 response, it should be recalled that the dominating mineral dust aerosol particles in those regions are

1 relatively large, elevated (off the ground) and absorbing. For solar only effects (in the center column of
 2 Figure 5) absorbing mineral dust aerosol over the bright desert surface causes a relative TOA warming
 3 by dimming the surface solar reflection to space. And there are also infrared effect, because mineral
 4 dust sizes are relative large. As mineral dust (unlike sea-salt) is often elevated and strongly absorbing,
 5 mineral dust also contributes with a significant greenhouse effect. This greenhouse effect adds to the
 6 solar warming at TOA and offsets with infrared re-radiation the surface the solar cooling at the surface.
 7 On a global average basis ca 35% of the solar energy losses by present-day aerosol are compensated by
 8 IR energy gains. In other regions, where smaller fine-mode sizes dominate, aerosol cools at TOA (unless
 9 surfaces are bright, as with snow or lower clouds) and always cools at the surface. Hereby the local
 10 surface cooling is much more negative than the local TOA response, when fine-mode aerosol is strongly
 11 absorbing, as over biomass burning regions over Africa and S. America and pollution of SE Asia.

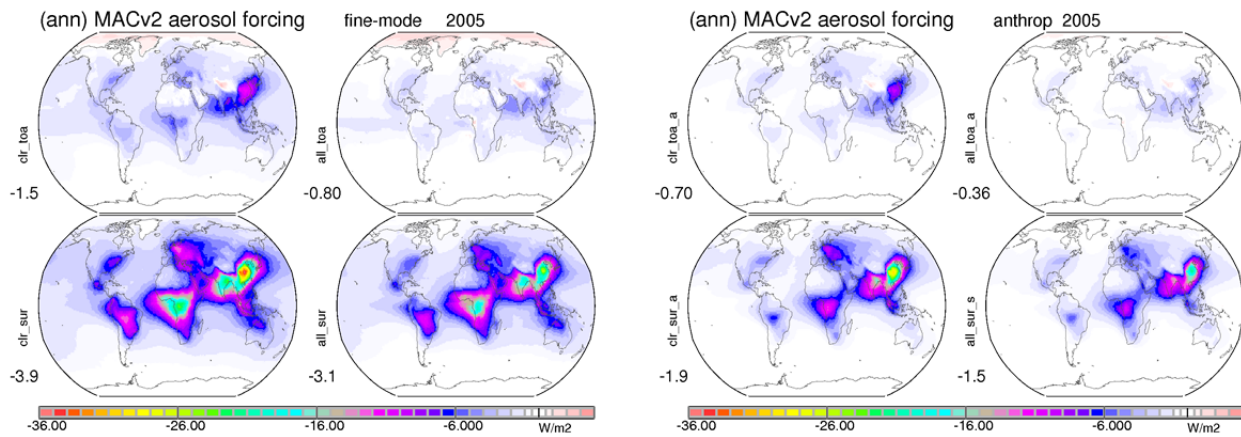
12 For anthropogenic aerosol MACv2 only allows sub-micrometer size contributions. Thus only
 13 solar radiative impacts matter. For present-day anthropogenic aerosol (for climate change relevant)
 14 aerosol direct forcing (the direct radiative effects by anthropogenic aerosol at all-sky conditions at TOA)
 15 yields a global cooling at -0.36W/m^2 . This direct forcing signal is on average more than a magnitude
 16 smaller than detectable solar radiation reductions at the surface or satellite detectable planetary albedo
 17 increases over oceans for total aerosol. More details on seasonal variability for the present-day direct
 18 forcing for anthropogenic aerosol (at an adjusted finer scale) are presented in Figure 6.

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 22 **Figure 6** Monthly maps for present-day aerosol direct forcing with MACv2 aerosol properties

1 On a global average basis the present-day aerosol direct forcing causes a cooling during the
 2 entire year. The TOA cooling is strongest from Jul to Sep mainly due to more sun-hours and higher AOD
 3 values at northern mid-latitudes. Higher AOD are in part caused by increased water uptake. The TOA
 4 cooling is weakest from Dec to April also due to regional warming over snow cover in the northern
 5 hemisphere (e.g. Asia, Arctic). The regional warming over the SE Atlantic in Aug-Sep is due to lower
 6 cloud reflection dimming by elevated biomass burning aerosol. The spatial distribution also illustrates
 7 the location of forcing maxima: May-Oct over E. Asia, Jun-Aug over EU and E. US, Sep to Apr over S. Asia.
 8 More details on present-day direct radiative effects are provided by investigating contributions by the
 9 (sub-micrometer) fine-mode sizes in Figure 7 and by (super-micrometer) coarse-mode size in Figure 8.



12 **Figure 7** Annual maps for present-day fine-mode (left block) and anthropogenic effects (right block) in
 13 MACv2. Aerosol radiative effects at TOA (top row) and at surface (bottom row) are presented for clear-
 14 sky conditions (left column in each block) and all-sky conditions (right column in each block). Blue to red
 15 colors indicate a cooling and pink colors weak warming. Values below the labels show global averages.

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 19 Fine-mode and coarse mode radiative effects differ in their regional contributions. On average
 20 fine-mode contributions to TOA cooling are stronger than coarse-mode contributions, despite a stronger
 21 solar fine-mode absorption (in the atmosphere). This is explained by regional greenhouse contributions
 22 of coarse-mode aerosols (over northern Africa, Arabia and Asia). The partially offsetting local solar and
 23 infrared contributions to coarse mode direct radiative effects at TOA and surface are shown in Figure 8.

24 Also compared in Figure 7 and 8 are size-mode contributions by anthropogenic aerosol to
 25 present-day direct radiative effects. For the fine-mode on average 50% of present-day fine-mode
 26 radiative effects can be considered anthropogenic (with the other 50% already contributing at pre-
 27 industrial times). Anthropogenic coarse-mode mineral dust (due to land use change) contribute at about
 28 10% to the dust AOD and to radiative effects at the surface. At the TOA the coarse-mode radiative effect
 29 contributions are only at 5% at TOA due to partially offsetting solar and infrared effects.

30 Based on a comparison of present-day direct aerosol forcing contributions of -0.36 W/m^2 for
 31 the fine-mode and $+0.02 \text{ W/m}^2$ for the coarse mode, the more than a magnitude smaller coarse mode
 32 contributions to the anthropogenic AOD are ignored in MACv2.

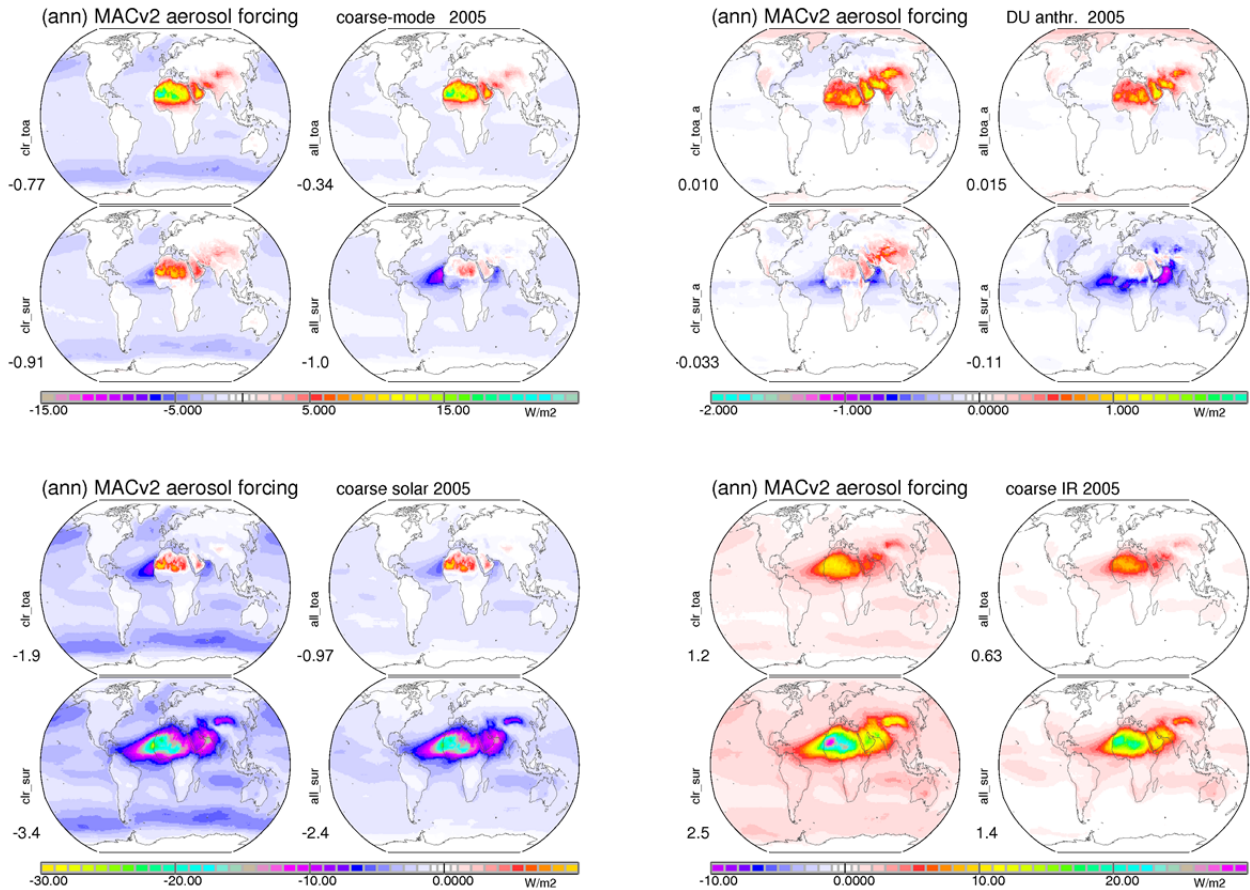
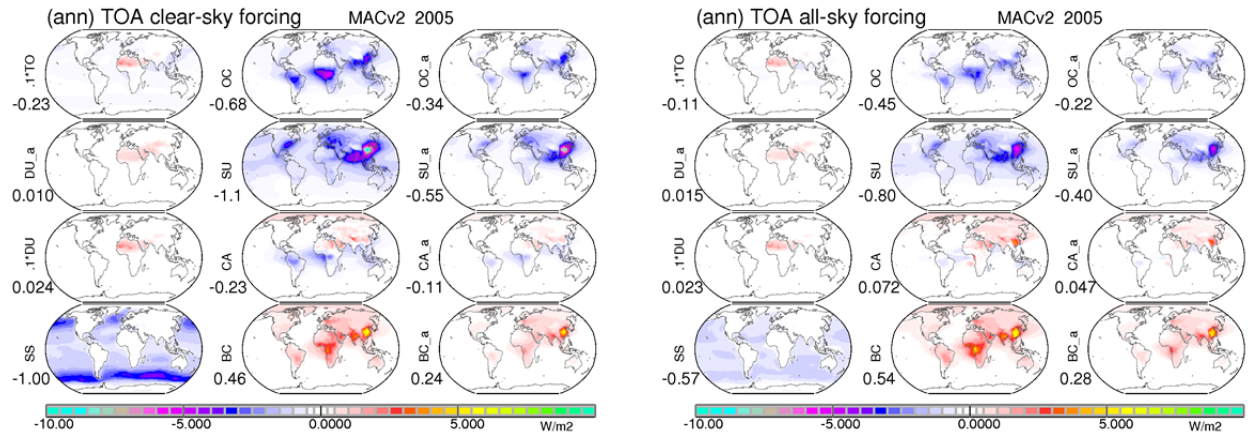


Figure 8 Annual maps for the coarse-mode aerosol direct radiative effects at TOA (top) and surface (bottom) for clear-sky (left) and all-sky (right) conditions. Coarse-mode effects (top, left block) are compared to potential anthropogenic contributions (top, right block, note its 10 times smaller color scale range). Blue to purple colors indicate a cooling and red to yellow colors a warming. In addition, solar (bottom, left block) and IR contributions (bottom, right block) to the coarse-mode aerosol direct radiative effects are presented. Note the different scales. Values below the labels indicate global averages.

With the AOD attributions in MACv2 into radiatively defined components via their mid-visible absorption properties (Kinne, 2019) and via the anthropogenic definition in MACv2, present-day direct aerosol radiative effects of components and of their anthropogenic contributions could be determined. For smaller fine-mode aerosol, direct radiative effects are contributed by non-absorbing aerosol represented by sulfate (SU), by weakly absorbing organic matter (OC) and by strongly absorbing soot (BC). Also the combined carbon (CA = OC+BC) effect is examined to illustrate the BC impact, when co-emitters of soot are included. For coarse-mode aerosol, direct radiative effects are contributed by non-absorbing sea-salt (SS) and weakly absorbing mineral dust (DU). Hereby stronger coarse-mode absorption translates into larger mineral dust sizes. Annual maps for present-day component TOA radiative effects at clear-sky and all-sky conditions (forcing) are shown in Figure 9. More details on component direct radiative effects are presented in Appendix E.



1 **Figure 9** Annual maps for today's direct forcing at TOA at clear-sky (left block) and at all-sky conditions
 2 (right block) by total aerosol (TO, divided by 10 to fit common scale) and by coarse-mode components of
 3 anthropogenic dust (DU_a), dust (DU, divided by 10 to fit common scale) and sea-salt (SS) in the left
 4 column, by fine-mode components of non-absorbing sulfate (SU), of weakly absorbing organic matter
 5 (OC), of strong absorbing soot (BC) and of the combined carbon (CA=OC+BC) component in the center
 6 column and by the anthropogenic contributions of all fine-mode components in the right column. Global
 7 averages for the annual direct aerosol component TOA effects are presented below the labels.
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 11 The radiative TOA direct radiative effects of individual components are quite diverse. Climate
 12 cooling by sulfate (SU), sea-salt (SS), organic matter (OC) and dust (DU-o) over oceans is on average
 13 stronger than climate warming by soot (BC) and dust (DU-c) over continents. As MACv2 only considers
 14 anthropogenic contributions to the fine-mode, investigations of anthropogenic impacts can be reduced
 15 to cooling by non-absorbing SU and weakly absorbing OC and warming by strongly absorbing BC. The
 16 combined carbon (CA) approximates the impacts of co-emitted gases that quickly condensate on
 17 existing particles (including soot) to increase scattering on existing particles (also in the context of an
 18 upper limit near 2 for a soot absorption increase via a scattering shell). The combined CA forcing is near
 19 neutral (+0.05 W/m²). This suggests that for approximate estimates for the global average direct forcing
 20 only changes in fine-mode non-absorbing AOD since pre-industrial times should be considered.

21 The CA near neutral response, however, only applies, if the BC anthropogenic fraction is that of
 22 the fine-mode AOD fraction. With a higher soot (BC) anthropogenic fraction (especially near pollution
 23 regions), as assumed in the BC assessment (*Bond et al., 2013*), the present-day BC forcing increases to
 24 +0.44 W/m² as shown in Appendix E. This added BC warming (with an alternate present-day CA
 25 warming near +0.20 W/m²) would reduce the present-day total direct aerosol forcing to -0.20 W/m²
 26 climate cooling. However, as this higher alternate fine-mode anthropogenic fraction for BC is also linked
 27 to a year 1750 reference (*Dentener et al., 2006*), such a large BC anthropogenic fraction seems unlikely
 28 for the year 1850. Still, given the uncertainty to the BC pre-industrial state an overall present-day
 29 aerosol direct forcing of -0.25 W/m² (down from -0.36 W/m²) cannot be easily ruled out.

30 The consideration of co-emitted gases soot (BC) removal processes may not have the often
 31 attributed potential for short term climate warming mitigations. Thus, the singled out present-day
 32 warming +0.55 W/m² for all soot (BC) with estimated anthropogenic contributions between +0.25 to
 33 +0.40 W/m² is deceiving as co-emitters also have to be considered in removal processes.

1 Annual averages of MACv2 aerosol associated direct radiative effects for present-day
 2 atmospheric conditions are summarized in Tables 2 and 3. Table 2 compares radiative effects at TOA,
 3 atmosphere and surface for all aerosol, for fine-mode aerosol, for coarse-mode aerosol and for
 4 individual aerosol components. Table 3 compares solar and infrared contributions of components with
 5 non-negligible infrared impacts. Global maps for component radiative effects of Table 2 are presented in
 6 Appendix D.

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 9 **Table 2** annual average MACv2 climatology associated aerosol radiative effects for today's tropospheric
 10 aerosol at the top of the atmosphere (TOA), at the surface and (by difference) for the atmosphere. Aside
 11 for total aerosol (in row 1) also effects of components and if applicable their anthropogenic contributions
 12 are indicated. Considered fine-mode components are sulfate (SU), organic matter (OC), soot (BC) and the
 13 combined carbon (OC+BC). Considered coarse-mode components are sea-salt (SS) and dust (DU).

direct effect (W/m ²)	TOA				ATMOSPHERE				SURFACE			
	total		anthr		total		anthr		total		anthr	
	all	clear	all	clear	all	clear	all	clear	all	clear	all	clear
total	-1.1	-2.3			+2.9	+2.3			-4.0	-4.6		
fine	-0.80	-1.5	-0.36	-0.70	+2.3	+2.4	+1.1	+1.2	-3.1	-3.9	-1.5	-1.9
- SU	-0.83	-1.2	-0.41	-0.58	+0.01	+0.02	+0.00	+0.01	-0.84	-1.2	-0.41	-0.59
- CA	+0.08	-0.23	+0.05	-0.10	+2.0	+2.2	+1.0	+1.2	-2.1	-2.5	-1.0	-1.3
- OC	-0.45	-0.68	-0.22	-0.34	+0.49	+0.52	+0.23	+0.24	-0.94	-1.2	-0.45	-0.58
- BC	+0.55	+0.46	+0.28	+0.24	+1.7	+1.8	+0.89	+0.94	-1.2	-1.4	-0.61	-0.70
- BC*			+0.44	+0.37			+1.4	+1.5			-0.97	-1.1
coarse	-0.34	-0.77			+0.66	+0.14			-1.00	-0.91		
- SS	-0.57	-1.00			+0.01	-0.10			-0.58	-0.90		
- DU	+0.23	+0.24	+0.015	+0.010	+0.68	+0.25	+0.12	+0.13	-0.45	-0.01	-0.11	-0.33

15 *based on AeroCom 1 - ref year 1750 (and not AeroCom 2 - ref year 1850) anthropogenic BC fine-mode fractions

16
 17
 18
 19 **Table 3** annual average MACv2 climatology associated aerosol radiative effects for today's tropospheric
 20 aerosol at the top of the atmosphere (TOA) and at the surface, by separating solar and IR contributions.
 21 Aside for the total aerosol (in row1) also effects are presented for components with an IR impact, such as
 22 seasalt (SS), dust (DU) and anthropogenic dust (aDU).

direct effect (W/m ²)	TOA						SURFACE					
	all-sky			clear-sky			all-sky			clear-sky		
		solar	IR		solar	IR		solar	IR		solar	IR
total	-1.1	-1.8	+0.66	-2.3	-3.5	+1.2	-4.0	-5.5	+1.5	-4.6	-7.4	+2.8
coarse	-0.34	-0.97	+0.63	-0.77	-1.9	+1.2	-1.00	-2.4	+1.4	-0.91	-3.4	+2.5
- SS	-0.57	-0.72	+0.16	-1.00	-1.4	+0.39	-0.58	-0.78	+0.20	-0.90	-1.4	+0.55
- DU	+0.23	-0.24	+0.47	+0.24	-0.53	+0.77	-0.45	-1.6	+1.2	-0.01	-1.9	+1.9
- aDU	+0.015	-0.07	+0.08	+0.01	-0.12	+0.13	-0.11	-0.29	+0.18	-0.03	-0.37	+0.34

24
 25

6. Direct Forcing Efficiencies

Forcing efficiencies offer a shortcut to radiative effects without actually performing radiative transfer simulations. Radiative forcing modulations by local monthly environmental properties (such as surface albedo, solar insolation and even clouds) are already included. For instance, a satellite retrieved AOD value or an anthropogenic AOD enhancement is then quickly associated with a radiative effect simply by multiplying the AOD value with the appropriate forcing efficiency. In such applications even information on the likely aerosol composition is included. Applying environmental assumptions and MACv2 aerosol properties in (radiative transfer) off-line simulations, aerosol direct forcing efficiencies are presented with respect to the mid-visible AOD at 550nm (as this particular AOD value is frequently used in conjunction with satellite retrievals and global modeling). Annual maps for forcing efficiencies per unit AOD for total and anthropogenic aerosol (corresponding to the radiative effects of Figure 5) are presented in Figure 10. Seasonal variations of global averages are small, as shown in Appendix D.

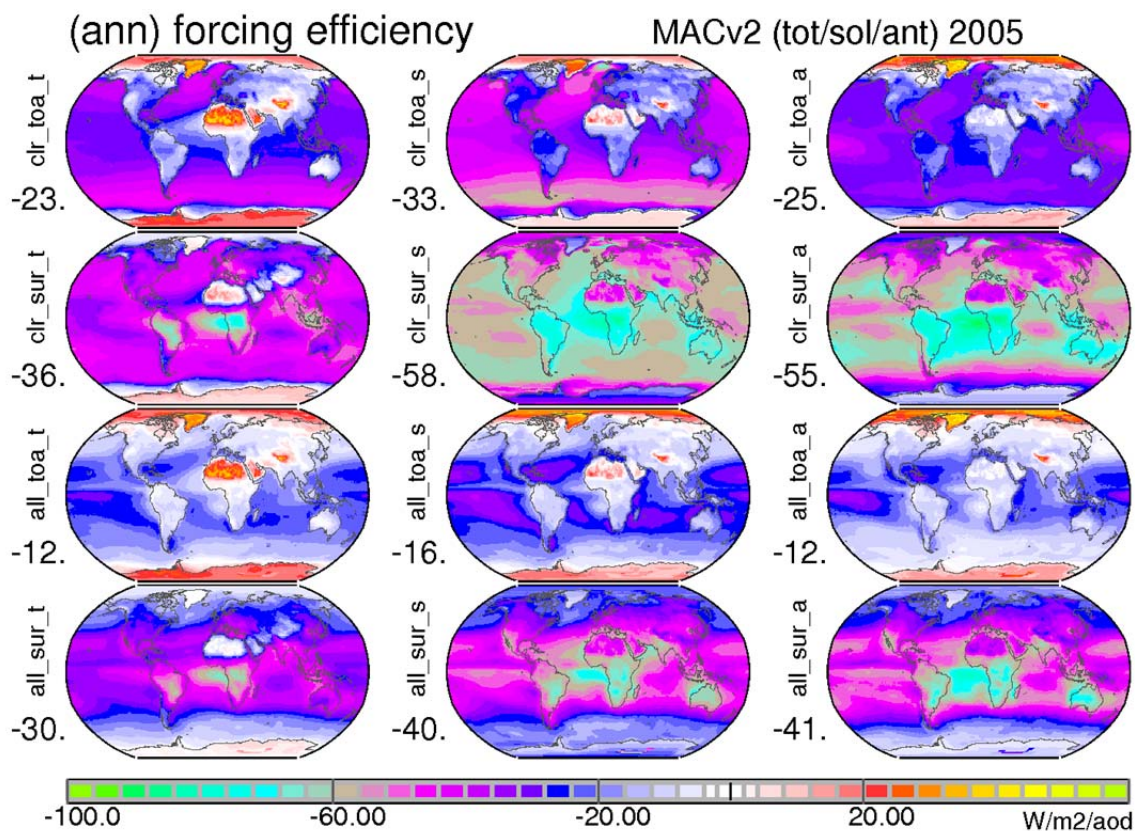
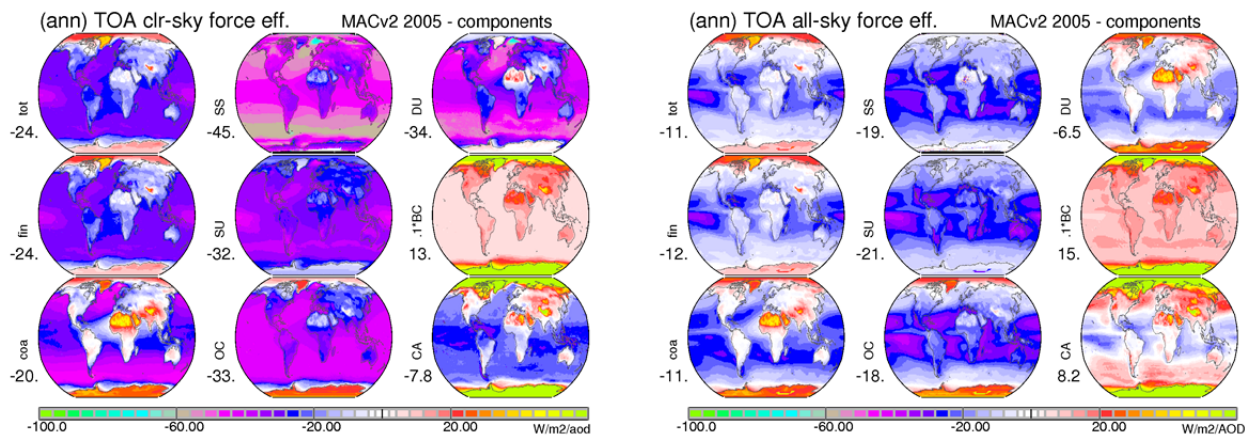


Figure 10 Annual maps for aerosol direct radiative effect efficiencies (per unit AOD) of present-day all aerosol (left column), its solar effect only (center column) and of present day anthropogenic aerosol (right column). All maps correspond to direct radiative effects of Figure 5: Clear-sky efficiencies are presented in the upper rows: at TOA (row 1) and at surface (row2). All-sky efficiencies (with ISCCP clouds) are presented in the lower rows: at TOA (row 3) and at surface (row 4). Values indicate global averages.

1 At TOA, global average direct aerosol radiative efficiencies (per unit AOD) are at -24 W/m²/AOD
 2 for cloud-free ('clear-sky') conditions and at -12 W/m²/AOD for more realistic conditions with clouds
 3 ('all-sky'). These global forcing efficiencies are almost identical for total and anthropogenic aerosol. This
 4 is a coincidence. The on average 30% stronger solar TOA cooling efficiencies for total aerosol are offset
 5 by a coarse-mode associated IR warming efficiencies - mainly by elevated mineral dust. Spatially,
 6 however, there are large differences associated with aerosol properties (e.g. absorption and size) and
 7 background reflectance data (e.g. surface albedo, lower altitude cloud cover).

8 At the surface global average only solar direct aerosol radiative efficiencies (per unit AOD) agree
 9 for total and anthropogenic aerosol at -56 W/m²/AOD for clear-sky and at -41 W/m²/AOD for all-sky
 10 conditions. For combined (solar and IR) direct forcing efficiencies those for total aerosol are ca 30% less
 11 negative, due to positive infrared re-radiation ('greenhouse effect') contributions.

12 With the AOD attributions in MACv2 into radiatively pre-defined aerosol types (*Kinne, 2019*) also
 13 TOA efficiencies for individual aerosol components are determined. TOA forcing efficiencies for the SU,
 14 OC, BC, DU and SS components are presented for clear-sky and all-sky conditions in Figure 11.
 15
 16



17 **Figure 11** Annual maps for present-day direct component forcing efficiencies (per unit AOD) at TOA at
 18 clear-sky (left block) and at all-sky conditions (right block) for total, coarse mode and fine-mode aerosols
 19 (left column), for mainly scattering components (center column) of sea-salt (SS), fine-mode (SU), organic
 20 matter (OC) (center column) and components with absorption (right column) of dust (DU), soot (BC) and
 21 for the combined carbon (CA=OC+BC). The large positive forcing efficiencies for BC (130 W/m²/AOD for
 22 clear-sky and 150 W/m²/AOD for all-sky) are divided by 10 to fit the common color scale. Global
 23 averages are presented below the labels.
 24
 25
 26

27 Direct radiative TOA forcing efficiencies for scattering fine-mode (SU) and coarse-mode (SS)
 28 components are strongly negative and usually more negative over the darker background of oceans.
 29 This also applies for weakly absorbing organic matter (OC) - except over (brighter) polar regions. The
 30 efficiencies are at least -30 W/m²/AOD for clear-sky and near -20 W/m²/AOD for all-sky conditions.

31 Direct radiative TOA forcing efficiencies for coarse-mode absorbing dust (DU) are also much
 32 more negative at clear-sky conditions compared to all-sky conditions. The clear-sky forcing efficiencies

1 over oceanic regions are almost as negative as for seasalt, but less negative over continents and
2 continental outflow regions and even slightly positive near source regions. The all-sky forcing efficiency
3 for dust displays only weak negative forcing efficiencies over oceans and usually positive forcing
4 efficiencies over continents. Over major dust source regions (e.g. N. Africa and Arabia) all-sky forcing
5 efficiencies for mineral dust (DU) are particularly strong at +40 W/m²/AOD.

6 Direct radiative TOA forcing efficiencies for strongly absorbing soot (BC) are even more positive
7 (than the all-sky forcing for dust near sources) and high everywhere over the globe, with maxima over
8 (bright) polar surfaces. Unlike for other components BC forcing efficiencies near +150 W/m²/AOD are
9 larger at all-sky conditions than at clear-sky conditions (with +120 W/m²/AOD). This is explained by BC
10 aerosol dimming of solar reflection from lower altitude clouds.

11 Direct radiative TOA forcing efficiencies for combined carbon (BC+OC) represents the presence
12 of mainly scattering BC co-emitters. Now, the high BC efficiencies are reduced to -10W/m²/AOD at
13 clear-sky and to +10W/m²/AOD at all-sky conditions. Thus, by ignoring scattering co-emitters, climate
14 warming potential of BC is overrated.

16 7. Indirect Aerosol Effects

17
18 Extra atmospheric aerosol loads (as from anthropogenic sources) modulate the atmospheric
19 energy distribution not only directly and but also indirectly. Indirect effects are contributed through
20 aerosol imposed changes to other atmospheric properties, most importantly to properties of clouds. An
21 important aspect is that added aerosols (relatively numerous from anthropogenic fine-mode sources)
22 increase the concentrations of those aerosols that can serve as cloud condensation nuclei (CCN). With
23 more available CCN at a condensation event, the available supersaturated water vapor is distributed
24 onto more aerosol nuclei, so that the resulting water cloud droplets are more numerous and smaller in
25 size - assuming that no changes to the cloud liquid water content (LWC) occur. With smaller drop sizes
26 the solar reflection of a water cloud and along with it the planetary albedo increases (*Twomey, 1974*) for
27 an added climate cooling. Examples are so-called 'ship tracks', when satellite sensors detect increases in
28 planetary albedo of low altitude clouds above the path of polluting ships. But there are further impacts
29 associated with smaller droplets affecting both cloud cover and cloud water content. The mixing with
30 dryer air at cloud boundaries reduces the cloud lifetime (especially if the cloud cover is low), whereas
31 the delay in the onset of precipitation extends the cloud lifetime (especially if cloud cover is high). In
32 addition, there are potential but less investigated aerosol impacts involving mixed-phase and ice clouds.

33 The strongest observational evidences for aerosol indirect effects involve low altitude water
34 clouds. In contrast, likely aerosol impacts are small for mixed phase clouds (*Christensen et al. 2016*) and
35 for ice-clouds model simulation cannot even agree on the sign of overall impacts (*Penner et al., 2018*).
36 With a focus on aerosol modifications to lower altitude water clouds, there remains the question, if
37 changes to cloud lifetime and/or cloud cover matter in comparison reductions in cloud droplet sizes (the
38 Twomey-effect). A recent satellite retrieval analysis involving a large sulfate aerosol anomaly over the
39 northern Atlantic (*Mallavele et al., 2017*) confirms the dominance of the Twomey-effect. It was found
40 that sharply increased sulfate aerosol concentrations strongly reduced the cloud droplet sizes. However
41 no significant changes to the cloud liquid water content were retrieved.

1 It seems straightforward to convert MACv2 suggested CCN increases due to anthropogenic AOD
2 in cloud droplet number concentration (CDNC) increases (and subsequently in associated cloud droplet
3 size reductions, assuming that the cloud water content did not change). Unfortunately, MACv2 based
4 CCN estimates (derived from fine-mode composition and fine-mode extinction, *Kinne 2019*) depend
5 strongly on assumptions for the supersaturation (or vertical winds at cloud base), which are neither
6 known nor can be appropriately represented by averages. To make matters worse, uncertainties affect
7 not only current CCN but also background CCN estimates, as both are needed to derive CDNC changes -
8 due to CCN saturation effects.

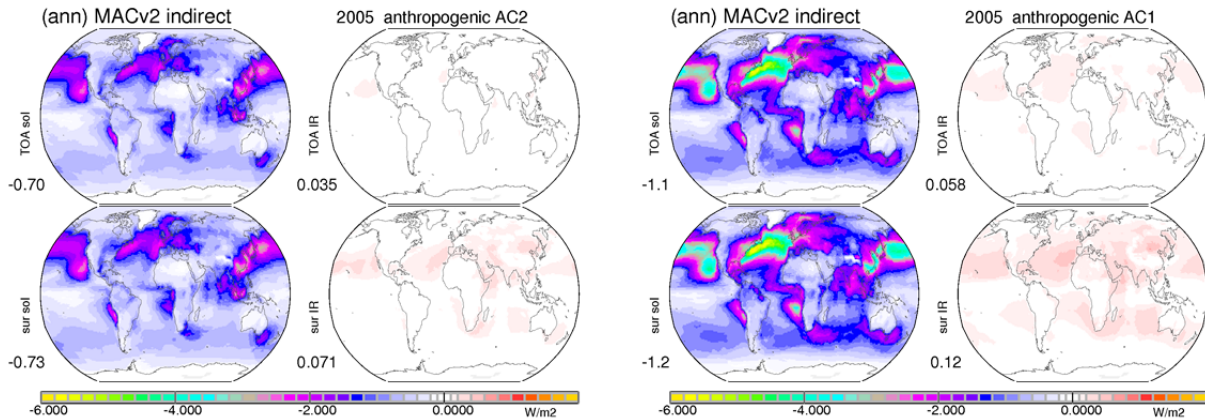
9 Thus, a simpler approach based on satellite retrievals was selected which directly links aerosol
10 number with cloud droplet number, as explained in more detail in Appendix A. It involves retrievals of
11 the same satellite sensor for fine-mode AOD (AODf) as a proxy for aerosol number and for CDNC at the
12 cloud-tops of low altitude clouds as proxy for cloud droplet number. Based on multi-spectral sensor data
13 both properties can be retrieved but not simultaneously at the same time and location. Here it is now
14 stipulated, that regional associations of monthly averages between AODf and CDNC offer meaningful
15 statistical constraints on aerosol-cloud interactions. Combining all monthly local matches over oceans,
16 where both, quality (no side-viewing retrieval, no broken cloud scenes) CDNC retrievals and sufficiently
17 large AODf retrievals (greater 0.05) were available, the AOD-size bin median associations are best
18 captured by a logarithmic relationship (more details are provided in Appendix A).
19

$$20 \quad \text{CDNC, factor} = \ln(1000 * \text{AODf}_{[\text{natural} + \text{anthropogenic}] + 3}) / \ln(1000 * \text{AODf}_{[\text{natural}] + 3})$$

21
22 The relationship predicts a CDNC increase factor due to extra AODf. Hereby the CDNC increase
23 depends not only on extra AODf from anthropogenic sources but also on natural AODf background. The
24 factor 3 in the formula accounts for CDNC associated with coarse-mode aerosols, which are important to
25 be included at very clean background conditions.

26 The CDNC-AODf relationship associated with observational statistics did not change significantly
27 with the use of different satellite sensor data (as illustrated in Appendix A). In contrast, the CDNC factor
28 increases in AeroCom global modeling were found more variable and on average much stronger (as
29 shown in Appendix A). In other words, if observational associations can be trusted, then in most
30 AeroCom models (with detailed aerosol schemes) the Twomey effect (associated with extra
31 anthropogenic aerosol) is over-parameterized. Note, that for better comparison the modeling data were
32 subsampled at locations of contributing CDNC vs AODf retrieval pairs.

33 For estimates of the aerosol first indirect effect in MACv2 the satellite retrieval based CDNC
34 increase factors were applied for every month and grid point, thus also over continents where no and
35 not sufficiently accurate satellite retrievals were available. After CDNC increases are converted into
36 cloud droplet radius reductions ($dR = 1/d(\text{CDNC})^{**[1/3]}$), assuming no changes to the cloud liquid water
37 content) two scenarios were simulated in an off-line radiative transfer code: The one scenario applied
38 reduced cloud droplet sizes according to the CDNC increases and the other scenario used the base-line
39 droplet size. The differences of these two simulations define aerosol indirect effects in MACv2. Present-
40 day annual average indirect effects for the solar and the infrared spectral region at TOA and surface are
41 presented for local anthropogenic AOD in the context of pre-industrial fine mode AOD in Figure 12.



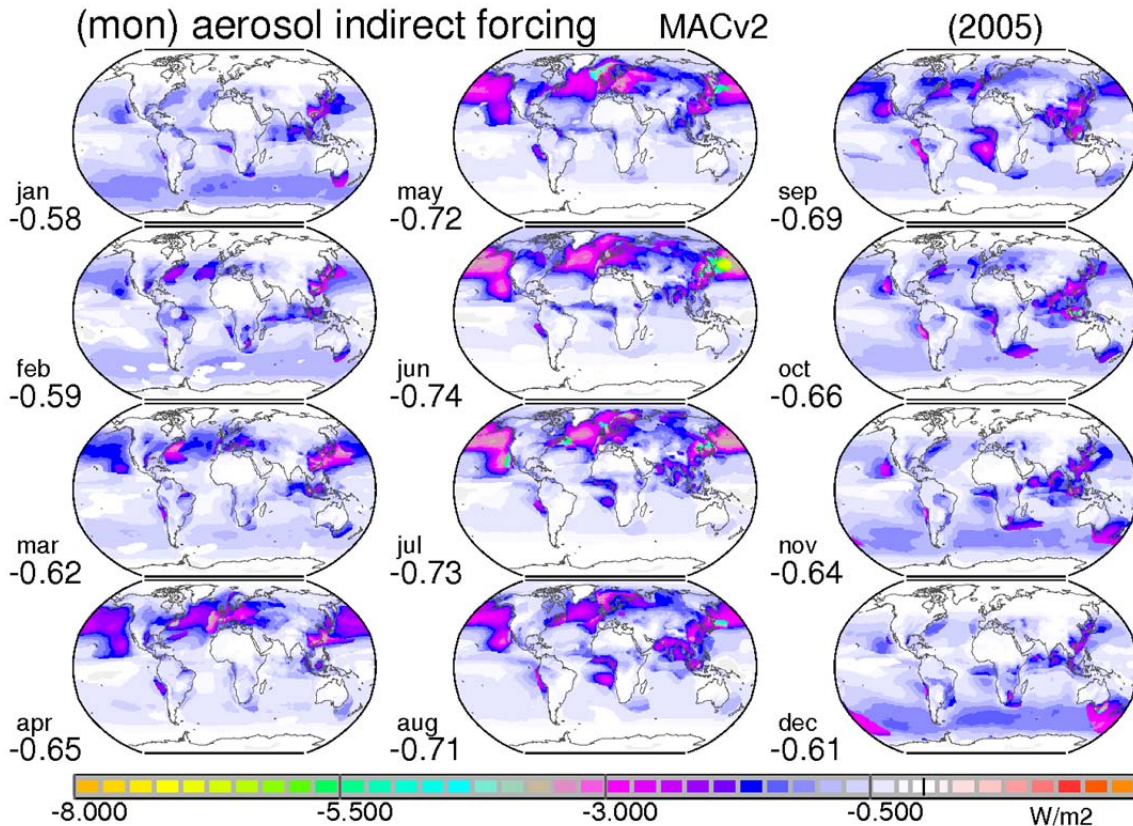
1
2 **Figure 12** Annual maps of indirect (Twomey) effects for present-day anthropogenic aerosol based on
3 year 1850 anthropogenic fine-mode fractions of AeroCom 2 (left block) and on 1750 anthropogenic fine-
4 mode fractions of AeroCom 1 (right block). Compared are in each block solar (left column) and infrared
5 (right column) radiative effects at TOA (top row) and at the surface (bottom row). Blue colors indicate a
6 'cooling' and red colors a 'warming'. Values below the labels are global averages.

7
8
9 Figure 12 compares the impacts for two different estimates for the present-day anthropogenic
10 AOD. One definition is based on the year 1850 and CMIP5 emissions (Lamarque et al., 2010), the MACv2
11 standard. The other definition applies 1750 as reference year and AeroCom phase 1 emissions (Dentener
12 et al., 2006). Global annual associated anthropogenic AOD maps are given in Appendix E in Figure E1.

13 The aerosol first indirect radiative effect is mainly a solar response with increases to the
14 planetary albedo and complementary decreases to the solar surface net-fluxes. There is a lot of spatial
15 variability with the largest contributions over (dark) oceans and the Northern Hemisphere. On a global
16 annual average basis (with the pre-industrial year 1850 reference and CMIP 5 emissions) the low cloud
17 solar cooling at -0.70 W/m² dominates its infrared (greenhouse) warming at $+0.04$ W/m². With the
18 alternate (ca 30% larger) anthropogenic AOD the low cloud solar cooling increases by 50% to -1.1 W/m².
19 The strong response is explained for two reasons: Aside from the increase to the anthropogenic AOD
20 also the (pre-industrial) background is reduced. And with the reduced background (via the logarithmic
21 relationship) the CDNC response is stronger with respect to the same AOD_f perturbation. Thus, the
22 indirect aerosol forcing is very sensitive to anthropogenic assumptions. Atmospheric radiative effects
23 (e.g. solar heating) are small, as cloud impacts at TOA and surface are almost identical.

24 Spatial variations on a monthly basis for simulated present-day aerosol TOA indirect forcing of
25 MACv2 (with the year 1850 reference and CMIP5 data) are presented in Figure 13. Figure 13 illustrates
26 that the aerosol indirect forcing (via the Twomey-effect) is not just influenced by anthropogenic aerosol
27 and background aerosol conditions but also by environmental properties. Important elements are solar
28 energy, sun elevation, solar surface albedo, likelihood of single layer low altitude clouds and a moderate
29 low cloud optical depth for highest susceptibility. The indirect forcing via smaller cloud drops is
30 strongest over mid-latitude oceans (with relative dark surfaces), during spring and summer (with longer
31 sun-shine hours) and mainly the Northern Hemisphere (where most anthropogenic aerosol is found).
32 Indirect effects over stratocumulus regions are relative moderate.

33



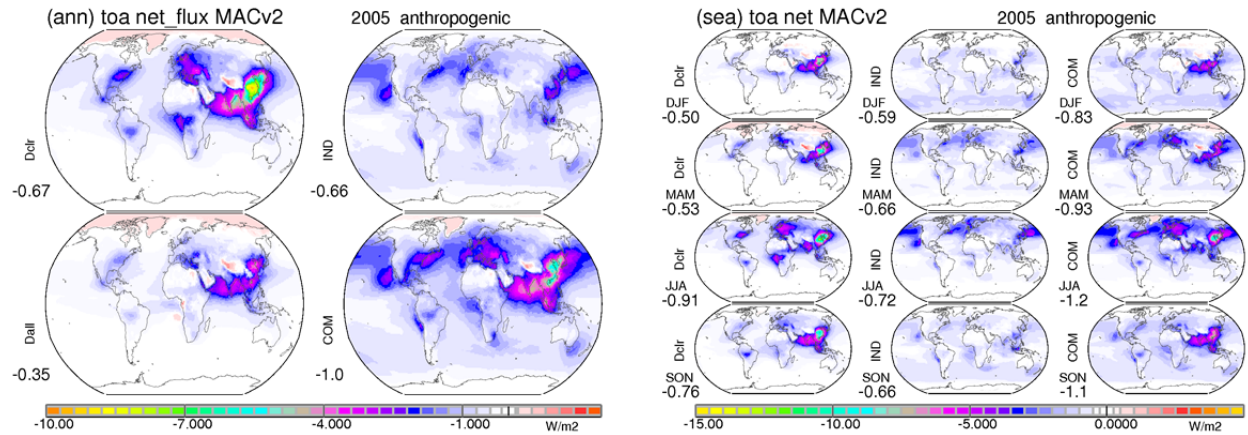
1
2 **Figure 13** Monthly maps for today's indirect (Twomey) forcing by present-day anthropogenic aerosol
3
4

5 Extra analysis of local environmental potential for a Twomey effect and maps for indirect forcing
6 efficiencies is provided in Appendix F. Due to very low aerosol natural background conditions, indirect
7 forcing efficiencies over Southern Oceans can be very high. Overall, however, contributions from those
8 regions are minor. Nonetheless, possible indirect effect overestimates in these regions cannot be ruled
9 out, also because the AODf vs CDNC relationship is not observationally constrained at very low AODf.
10

11 **8. Direct vs Indirect**

12
13 MACv2 associated aerosol present-day direct and indirect radiative effects are now compared.
14 Annual maps for TOA direct radiative effects at clear-sky conditions, for direct forcing (with clouds),
15 indirect forcing and the combined forcing (direct and indirect) are presented in Figure 14.

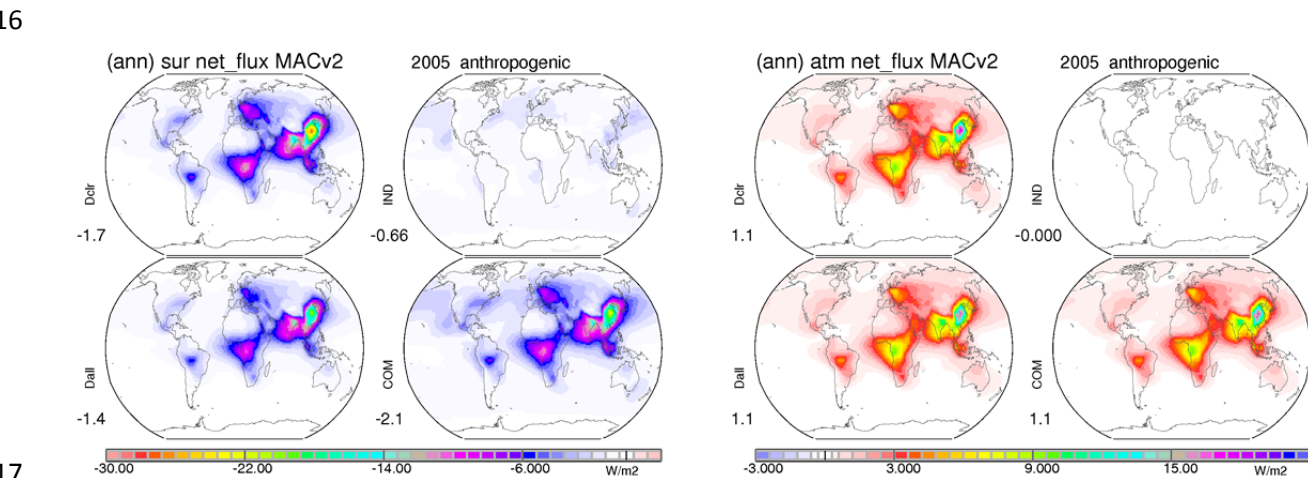
16 At the TOA climate cooling by the present-day indirect effect (globally averaged at -0.66 W/m^2)
17 is about twice as large as the present-day direct effect (globally averaged at -0.35 W/m^2). However, the
18 spatial variability of the direct forcing is much more diverse, including regions with local warming (via
19 dimming over snow, over lower clouds or during polar nights). Direct effect dominates near continental
20 sources while the indirect effect has stronger impacts over oceanic regions off sources. The combined
21 climate impact is a cooling (globally averaged at -1.0 W/m^2) with cooling everywhere except over
22 Greenland. The present-day clear-sky cooling is about 60% of the combined (direct and indirect) cooling.



1
 2 **Figure 14** TOA radiative effects by present-day anthropogenic aerosol. Annual (left block) and seasonal
 3 maps (right block) compare direct radiative effects at clear-sky conditions (Dclr) and all-sky conditions
 4 (Dall), aerosol indirect (Twomey) effects through modified clouds (IND) and the combined (direct and
 5 indirect) effect (COM). Blue colors indicate 'cooling' net-flux losses and (rare) red colors indicate
 6 'warming' net-flux gains. Values below the labels indicate global averages.

7
 8
 9 Figure 14 also compares seasonal variations. With larger AOD, less snow cover and more
 10 sunshine both indirect and direct effects have maximum impacts during the northern hemispheric
 11 summer season.

12 Annual maps for radiative effects at the surface and for the atmosphere are shown in Figure 15.
 13 Compared are (as in Figure 14) direct radiative effects without and with clouds, indirect radiative effects
 14 and the combined effects (direct with clouds and indirect).



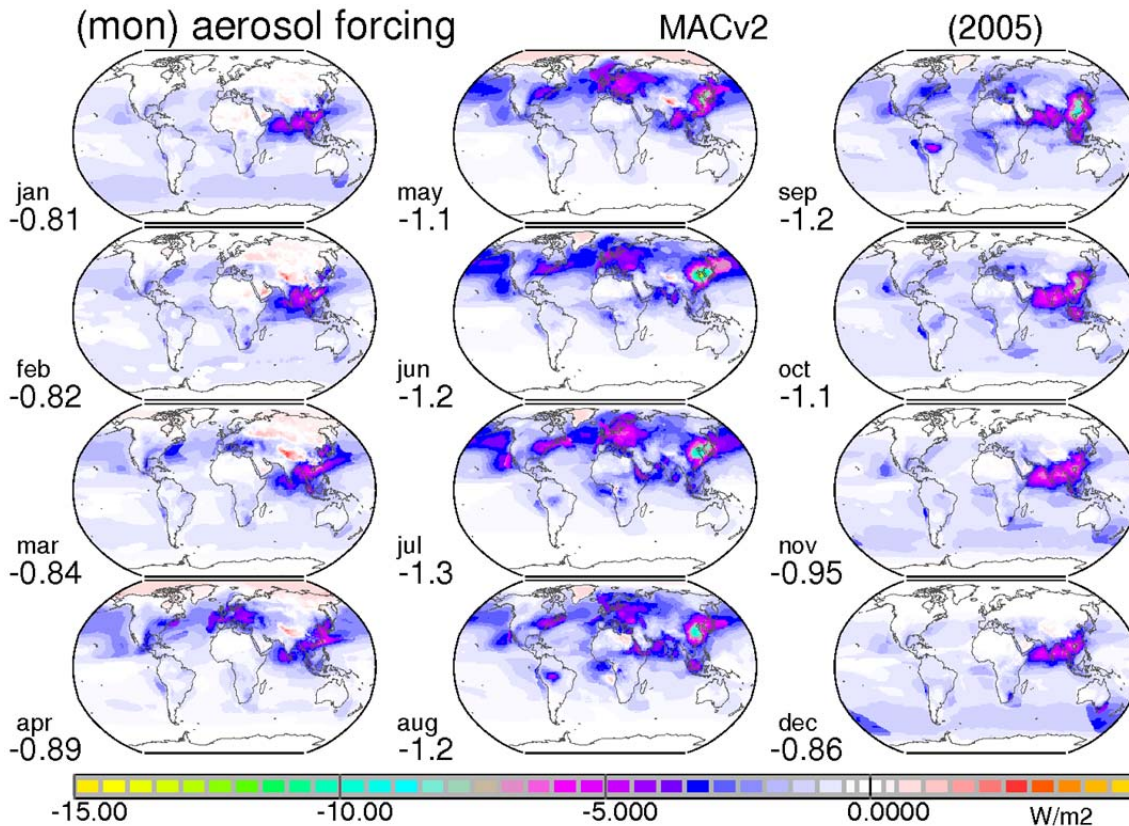
17
 18 **Figure 15** Annual radiative effects by present-day anthropogenic aerosol at the surface (left block) and
 19 for the atmosphere (right block). Maps compare direct effects at clear-sky conditions (Dclr) and all-sky
 20 conditions (Dall), indirect (Twomey) effects through modified clouds (IND) and the combined (direct and
 21 indirect) effect (COM). Blue colors indicate 'cooling' net-flux losses and red colors indicate 'warming' net-
 22 flux gains. Values below the labels indicate global averages.

23

1 In the atmosphere, present-day anthropogenic aerosol warms the atmosphere – on average at
 2 +1.1 W/m². The atmospheric warming is highly uneven with warming in excess of 10W/m² near sources
 3 of pollution (e.g. S.Asia, E.Asia) and wildfire regions (e.g. central Africa). The associated solar heating is
 4 almost entirely a the direct effect. Thus, aerosol direct effects control the atmospheric response.

5 At the surface the present-day direct effect yields, due to atmospheric losses, a more negative
 6 radiative effect than at TOA. The present-day direct effect reduces the (solar) flux is on average by about
 7 -1.45 W/m² . This is stronger than the flux reductions of about -0.66 W/m² for the indirect effect. Thus
 8 on a global annual average basis, at surface net-flux losses by aerosol are dominated by direct effects.

9 Monthly maps for the present-day combined (direct and indirect) TOA forcing are shown in
 10 Figure 16. More maps on TOA forcing including fractional contributions of clear-sky regions and cloudy-
 11 sky regions are presented in Appendix C.



14 **Figure 16** Monthly maps for today's total forcing by present-day anthropogenic aerosol

15
 16
 17
 18 Interestingly global averages of the clear-sky direct impact at TOA (in Figure 14) and at the
 19 surface (in Figure 15) resembles in magnitude and regional distribution that of the combined (direct and
 20 indirect) impact. Thus, for rough estimates on the aerosol impacts, already clear-sky radiative
 21 simulations could provide rough estimates for global averages for aerosol climate impacts.

9. Forcing over Time

The MACv2 aerosol climatology offers global maps for changing anthropogenic AOD over time (Kinne, 2019, Figure 8). Hereby the historic scaling back to year 1850 is based on transient ‘bottom-up’ ECHAM simulation (Stier et al., 2005) with NIES emissions, while future scaling is based on regionally simulated responses to changing sulfate emissions of the IPCC 5 RCP 8.5 future scenario. Results of radiative transfer simulations that apply these changing anthropogenic AOD data over time are presented in Figure 17. Maps for selected years (from 1865 to 2065) compare in 40-year steps the annual forcing of the direct, the indirect (Twomey) and the combined (direct and indirect) effect.

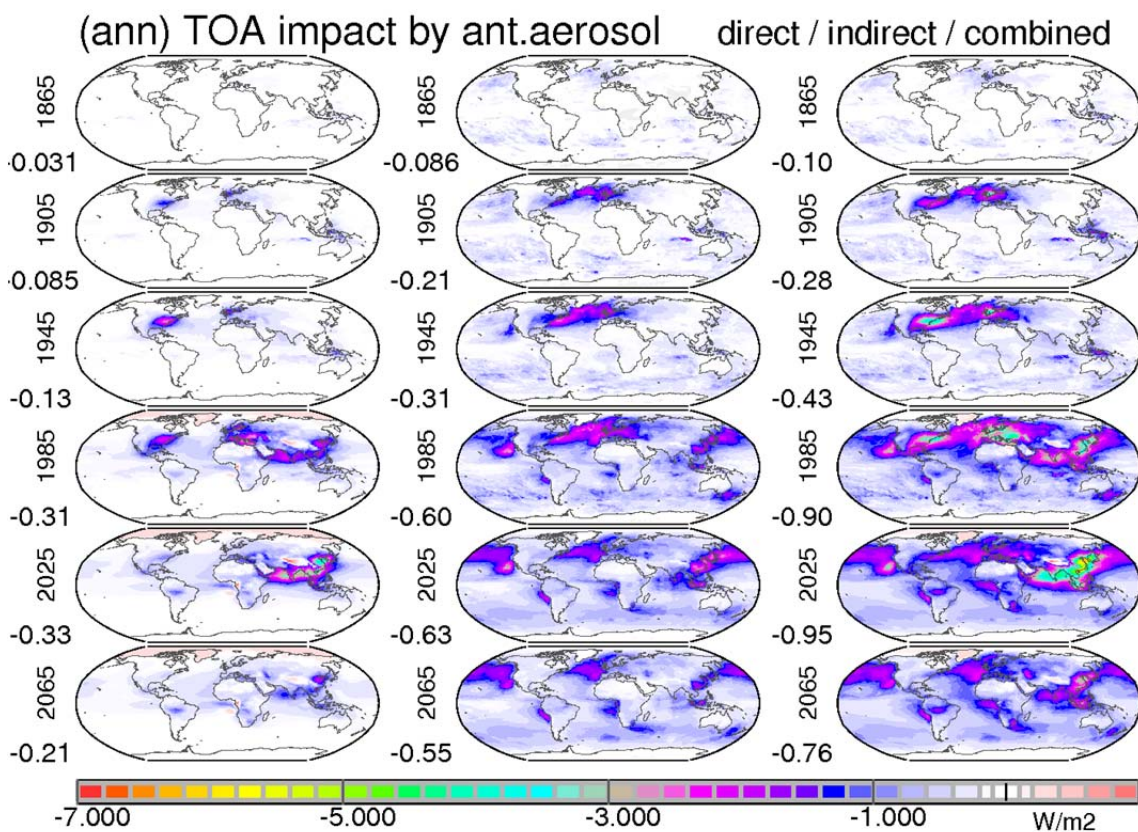


Figure 17. temporal evolution of the climate forcing by anthropogenic aerosol. For selected years (1865, 1905, 1945, 1985, 2025 and 2065) annual maps or the direct (column 1), the first indirect (column 2) and the combined effect (column 3) are compared. Values below the labels indicate global averages.

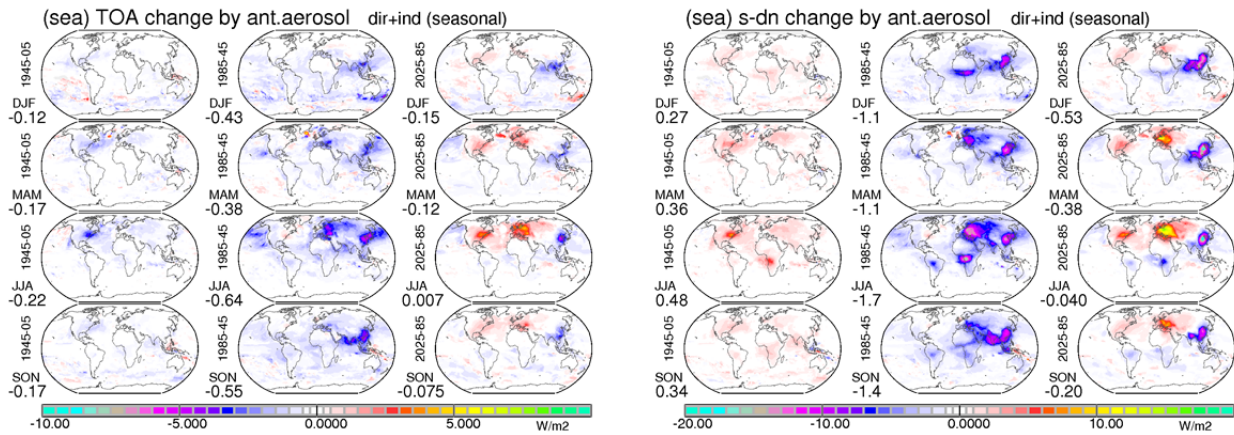
The forcing time-slices show that in the early years of the industrial revolution the fractional forcing contributions by indirect effect were relatively high. This is a consequence of a stronger indirect response at a lower background conditions, which is predicted by the applied logarithmic relationship.

The temporal time-slice maps demonstrate that early into the industrial period mainly the US and Europe were affected by aerosol cooling. By 1985 with SE Asia had emerged as a third affected

1 region. Since then the aerosol cooling over SE Asia kept on growing while aerosol cooling by over Europe
 2 and the US declined, also due to successful mitigations efforts. With these opposing regional shifts
 3 during the last decades the global average aerosol cooling stayed relatively stable at just below -1.0
 4 W/m². As in the meantime (by 2015) anthropogenic aerosol loads had reached their regional maximum
 5 over E Asia (while not yet over S Asia) no further increases for future aerosol cooling are being expected,
 6 even if future emission scenarios project the development of a new maximum over W Africa.

7 A different way to illustrate changes and regional shifts associated with extra anthropogenic
 8 aerosols are forcing difference over selected time-periods. Total TOA forcing differences and changes to
 9 the downward solar flux at the surface presented by season for three 40-year intervals in Figure 18.

10
 11



12

13 **Figure 18.** Seasonal changes over 20 year periods (1945-1905, 1985-1945, 2025-1985) in aerosol forcing
 14 (left block) and to solar downward fluxes by anthropogenic aerosol (right block). Blue colors indicate
 15 time-periods of increased cooling aerosol or a dimming (decreases to the solar surface fluxes), while red
 16 colors indicate time-periods of a reduced cooling by aerosol or a brightening (increases the solar surface
 17 fluxes). Values below the labels indicate global average changes for the selected time periods.

18

19

20 Between 1945 and 1905 TOA cooling occurred mainly over the eastern US during summers.
 21 Between 1985 and 1945 TOA cooling strongly increased during summer over near Europe and E. Asia
 22 and during the (dry) fall season over S. Asia. Between now and 1985, the (mainly summer) TOA cooling
 23 strongly decreased over Europe and the eastern US but kept increasing over SE Asia.

24 Also shown in Figure 18 are MACv2 aerosol simulated changes to the downward solar fluxes.
 25 These temporal regional and seasonal changes are consistent with observations by ground-based
 26 radiation (Wild, 2015). These results strongly suggest that the observed long-term average dimming
 27 (until 1985) following a long-term average brightening (since 1985) can be mainly attributed to
 28 anthropogenic aerosol.

29

30

31

32

10. Uncertainty

The calculations of the aerosol radiative effects and the aerosol radiative forcing include many uncertainties. The focus here is on uncertainties to the aerosol fields of the MACv2 (although also approximations in radiative transfer model, simplifications to environmental data and the application of monthly averages will contribute). An initial test in Figure 19 compares the impact of a different background maps for AODf and AODc in developing the MACv2 aerosol climatology.

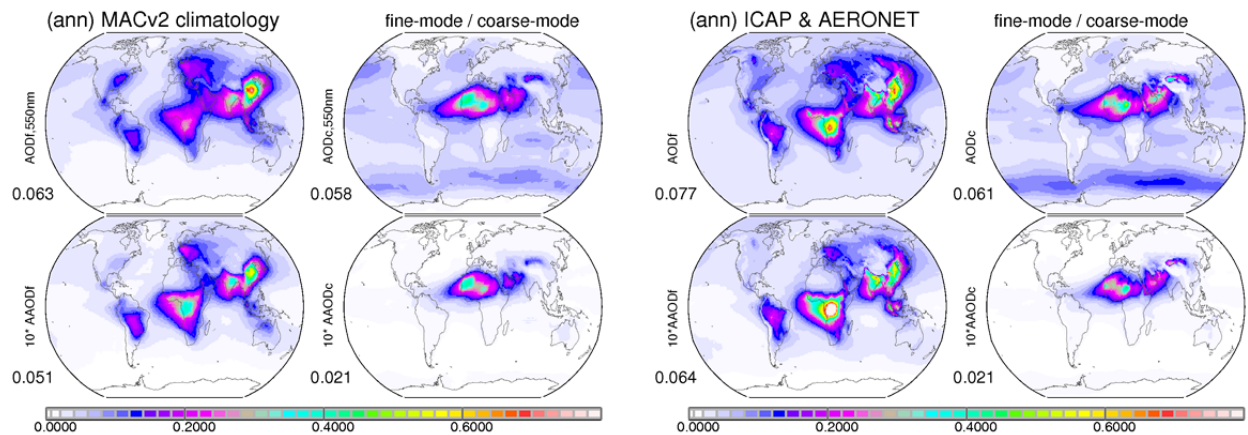


Figure 19 estimates of today's annual maps for tropospheric AODf, AODc, AAODf and AAODc by combining AERONET data with an AeroCom1 median (left block) and with satellite AOD data assimilating ICAP averages (right block). AAOD data are multiplied by 10. Values below labels show global averages.

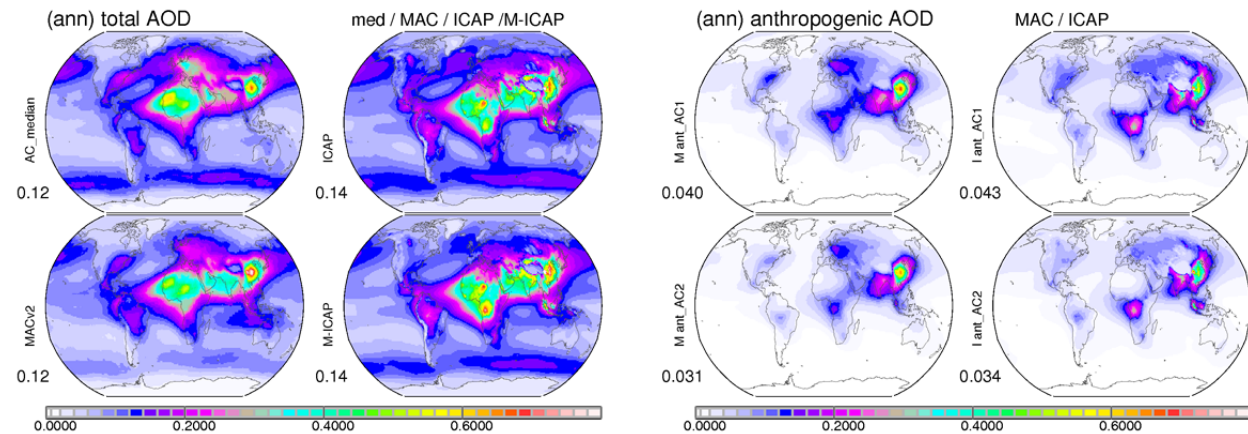
As an alternate modeling background in the data merging of MACv2, ICAP ensemble data (Peng *et al.*, 2018) from the satellite AOD assimilation community are used. Three hourly data of years 2015 and 2016 for averages of up to seven assimilations are combined into local monthly mean AODf and AODc. Global maps of these aerosol properties replace AODf and AODc maps of the AeroCom phase 1 background data in the merging with the sun-photometer statistics. All other properties (e.g. the absorption strength of each size mode) remain identical.

The climatology with the ICAP background yields on average ca 15% larger values for the mid-visible AOD. Patterns are slightly shifted but major differences are much stronger AODf (and AAODf) maxima over central Africa. It is difficult to judge if these differences are the result of (in assimilations applied MODIS) satellite AOD retrieval overestimates in that region or due to overlooked emissions in modeling. Unfortunately, there are no AERONET reference sites in that region. Globally the AODf is ca 25% larger. Fortunately, most of this extra AODf has a strong natural component so that anthropogenic AOD is increased by just 10% as shown in Figure 20.

The assumed pre-industrial reference states to define 'anthropogenic' contributions introduce probably the largest uncertainty. To illustrate this point, the standard anthropogenic AODf fraction based on a reference year 1850 and CMIP5 emissions (Lamarque *et al.*, 2010) was replaced by an alternate approach based on a reference year 1750 and AeroCom emissions (Dentener *et al.*, 2006).

1 Figure 20 shows that with the alternate approach the anthropogenic AOD in MACv2 increases (globally
 2 averaged) by 30% from 0.031 to 0.040. Considering the uncertainty in AOD but limiting the reference to
 3 the year 1850 then the range for anthropogenic AOD (at 550nm) is between 0.030 and 0.038. Another
 4 big uncertainty is the anthropogenic fraction for soot (BC) component, which dominates the absorption.
 5 The present-day anthropogenic BC component warming is uncertain between +0.25 and 0.45 W/m². If
 6 all of the present-day BC is a considered anthropogenic then there is an +0.55 W/m² upper ceiling. Thus,
 7 there is no support for larger present-day BC direct warming contributions (as in *Bond et al, 2013*).

8 The neglect of larger coarse-mode aerosol in the anthropogenic definition in MACv2 seems less
 9 important. Potential coarse-mode anthropogenic contributions via mineral dust (e.g. due to land use
 10 change) seem secondary, especially as anthropogenic dust forcing is near neutral, with solar and
 11 infrared radiative effects largely canceling each other.



14 **Figure 20** uncertainty estimates for present-day total and anthropogenic AOD from different realizations.
 15 Annual maps for tropospheric total AOD (left block) and for anthropogenic AOD (right block) are shown.
 16 Total AOD maps compare model-ensemble data of AeroCom1 experiments (left column) and ICAP
 17 satellite data assimilations (right column), before (top) and after (bottom) the data merging with sun-
 18 photometer data. Anthropogenic AOD maps are results of applying the two different anthropogenic
 19 fractions (based on emissions by Dentener - AC1 and Lamarque - AC2) to the fine-mode AOD of the
 20 AERONET adjusted maps. Values below the label show global averages.

24 Sensitivity studies in Appendix B demonstrate that a 25% larger anthropogenic AOD means a
 25 25% stronger direct forcing and a 40% larger indirect forcing. An alternate ICAP background in the data
 26 merging of MACv2 increases the total AOD for an up to a 15% stronger aerosol direct and aerosol
 27 indirect forcing. And a likely BC anthropogenic fraction that is larger than the fine-mode anthropogenic
 28 fraction will contribute to a less negative direct effect. Also the treatment of the indirect effect makes
 29 many assumptions and is highly simplified. Finally all presented results refer to an instantaneous impact.
 30 Short-term cloud adjustments are not included, but they are much smaller (on the order of 10%) in
 31 comparison (Fiedler et al., 2019) with a tendency to reduce the aerosol radiative effects.

32 Considering all contributing uncertainties, a -0.7 to -1.6 W/m² range is estimated for present-day
 33 global annual average aerosol forcing (assuming a 1850 pre-industrial reference year) and a most

1 likely value near -1.0 W/m^2 . Note, that the less negative lower bound (-0.7 W/m^2) is more certain than
2 the more negative (-1.6 W/m^2) upper bound.

3 The main argument towards the upper bound (-1.6 W/m^2) is that the anthropogenic fine-mode
4 fraction could be larger. The upper bound of the direct effect at -0.45 W/m^2 is much better constrained
5 than the upper bound for the indirect effect. An alternate anthropogenic scaling using AeroCom phase 1
6 emissions raised indirect effects (from -0.65) to -1.1 W/m^2 and direct effects (from -0.35) to -0.50 W/m^2
7 for a combined -1.6 W/m^2 cooling. However, this alternate larger anthropogenic fine-mode fraction
8 applies an earlier pre-industrial reference year (1750). Thus, such a negative aerosol forcing is unlikely
9 for a year 1850 reference. Also the simplicity in describing the aerosol indirect effects in MACv2 is very
10 simple. There are calls for a more processed based modeling treatment or observational approaches
11 which better distinguish specific aerosol and cloud properties (*Grandey et al., 2010*). However, since the
12 eventual global patterns for the indirect radiative forcing are already well matched (as environmental
13 properties play a big role), a more detailed indirect effect treatment may come at the expense of added
14 uncertainty. Another argument for an upper bound of -1.6 W/m^2 is that such strong cooling potential by
15 anthropogenic AOD at the middle of the 20th century would have caused global climate cooling which
16 was not observed (*Kretzschmar et al., 2016*).

17 The main argument for the lower bound (-0.7 W/m^2) is that the lower bound of the direct effect
18 (which is better constrained than the indirect effect) is estimated at -0.2 W/m^2 . This lower value can be
19 explained with a possibly higher anthropogenic fraction for absorbing BC AOD (than simply applying the
20 anthropogenic fine-mode AOD for anthropogenic BC). For the present-day indirect effect there remains
21 uncertainty, however, at least a cooling of -0.5 W/m^2 should be expected for the reduced drop size
22 effect. (In that context it should be noted that the less negative indirect effects of MACv2-SP in
23 Appendix B do not qualify due to their incomplete global coverage for anthropogenic AOD).

24 To reduce the aerosol forcing range (-0.7 to -1.6 W/m^2) progress in quantifying the indirect
25 effect(s) and pre-industrial references for all aerosol components are needed.
26

27 **11. Summary**

28
29 The MACv2 global monthly aerosol climatology, tied to observational monthly statistics on
30 aerosol amount, absorption and size from sun-/sky-photometry, was applied in dual-call off-line
31 radiative transfer to determine aerosol effects on atmospheric radiation. The direct (added presence)
32 effects of present-day aerosol and the climate change relevant effects of anthropogenic contributions
33 (since pre-industrial times) are determined. Hereby results are usually presented via global annual and
34 even monthly maps to visualize regional and temporal detail, as not only aerosol properties but also
35 influential environmental properties have strong regional and seasonal signatures. Still, for simplicity,
36 most discussions below apply resulting global annual averages. Hereby radiative net-flux changes are
37 examined at the climate relevant TOA location, at the surface for exchange processes near the ground
38 and for the atmospheric dynamics (via the TOA minus surface impact). Radiative net-flux losses refer to
39 a cooling and radiative net-flux gains refer to a warming.

40 Present-day total aerosol by its presence (direct effect) reduces net-fluxes by -1.1 W/m^2 at the
41 TOA and by -4.0 W/m^2 at the surface, so that the energy in the atmosphere is increased by $+2.9 \text{ W/m}^2$.

1 Hereby, the net-flux losses are composed of larger solar losses and a partly offsetting smaller infrared
2 gains, at -1.8 and $+0.7$ W/m^2 at the TOA and -5.5 and $+1.5$ W/m^2 at the surface, while atmospheric solar
3 warming of $+3.7$ W/m^2 is reduced by -0.8 W/m^2 in atmospheric IR cooling.

4 Anthropogenic aerosol is only considered to contribute to sub-micrometer aerosol sizes so that
5 only solar direct radiative effects matter (and potential IR radiative effects can be neglected). Present-
6 day anthropogenic aerosols by their added atmospheric presence (direct effect) are estimated to reduce
7 solar TOA net-fluxes by -0.36 W/m^2 and surface net-fluxes by -1.5 W/m^2 . By difference the atmosphere
8 is warmed by $+1.1$ W/m^2 . In addition, the major aerosol induced impacts to water clouds is considered
9 (the first indirect effect). More numerous aerosols reduce the cloud droplet sizes. This Twomey effect is
10 implemented via satellite retrieval based associations between aerosol and drop concentrations. The
11 present-day first indirect effect reduces mainly solar radiative net-fluxes at the TOA and at the surface
12 by -0.65 W/m^2 . Thus, the combined direct and indirect yields a cooling of -1.0 W/m^2 at the TOA and of $-$
13 2.1 W/m^2 at the surface. On average, the indirect effect dominates the (TOA) climate cooling, the direct
14 effect dominates the cooling at the surface and the direct effect determines the atmospheric heating.
15 Spatially both direct and indirect effects are strongest during the NH summer. Direct effects are
16 strongest near continental sources. Indirect effects are stronger away from sources over (dark) oceans.

17 Uncertainties with respect to the aerosol properties should consider that the total AOD could be
18 15% larger than in MACv2 and that a 25% larger fine-mode anthropogenic fraction than MACv2 is
19 possible. Simulations with a 30% larger anthropogenic fraction, yield a 30% larger direct effect and an
20 even 50% larger indirect effect. Thus, the uncertainty for present day forcing (the anthropogenic impact
21 at the TOA) is very likely between -0.7 and -1.6 W/m^2 , with -1.0 W/m^2 as best estimate. Hereby, the
22 direct effect is much better constrained (-0.20 to -0.45 W/m^2) than the indirect effect (-0.5 to -1.1
23 W/m^2). A better estimate for pre-industrial references (for fine-mode, the BC component and even for
24 dust) would help in reducing these uncertainties. And also the rather simple representation of the
25 indirect effect needs to be validated.

26 As MACv2 optical properties also allow to attribute direct radiative effects to components. The
27 'top-down' component estimates of this presentation for present-day forcing agree with those from
28 'bottom-up' modeling, as summarized in Appendix G. Present-day total soot (BC) warms at the TOA with
29 $+0.55$ W/m^2 . At least 50% and at most 85% could be attributed to BC, depending on the definition for
30 the anthropogenic BC fraction but certainly not more than 100%. With the possibility that the
31 anthropogenic fraction for soot (BC) that is larger than for the fine-mode fraction (in other words that
32 there was much less BC in the fine-mode aerosol in pre-industrial aerosol) then the assigned BC
33 warming (from now $+0.28$ W/m^2) would increase (to about $+0.38$ at 70% fraction or even $+0.45$ W/m^2 at
34 an 85% fraction). This in turn would reduce the present-day direct aerosol forcing to a -0.26 W/m^2 (at
35 70% anthropogenic BC) or just -0.19 W/m^2 (at 85% anthropogenic BC). Thus, not only accurate
36 information on the pre-industrial state of the fine-mode AOD but also on its components, mainly that of
37 the BC component, at that time (e.g. soot properties) is needed.

38 The combined carbon (BC and OC) component, which approximates the effect of BC co-emitters,
39 has a near neutral radiative forcing behavior. Thus, short term climate warming mitigation concepts via
40 a soot removal may not be very effective as also mainly scattering aerosol resulting from co-emitted
41 trace-gases would be removed. The climate impact for mineral dust (while showing strong warming over
42 continents, yet strong cooling over oceans) behaves globally almost climate neutral. Thus, potential

1 anthropogenic impacts from dust, as a result of land-use change, as uncertain as they are, seem less
2 importance for climate change considerations. Thus in the end it are the (in the mid-visible) non-
3 absorbing sub-micrometer size aerosol (mainly sulfate and nitrate) that regulate the anthropogenic
4 AOD.

5 Calculations with model predicted temporal changes to the anthropogenic AOD indicate that
6 qualitatively the anthropogenic aerosol forcing has not changed much over the last decades and is not
7 likely to increase over the next decades, despite strong regional shifts. These regional shifts explain most
8 solar insolation (brightening or dimming) trends that have been locally observed by decadal time-series
9 ground-based radiation, especially over Europe and the US.

10

11 Resource

12

13 MACv2 properties are accessible at ftp://ftp-projects.zmaw.de/aerocom/climatology/MACv2_2018/ .

14 The data are placed in several subdirectories and a README file describes data content of file-names

15 /550nm	(mid-visible) aerosol properties at 550nm wavelength
16 /CCN	lower cloud-base condensation nuclei and critical radii at diff. supersaturation
17 /detail	ancillary data for radiative transfer simulations
18 /documents	some documentation and figures
19 /forcing	MACv2 associated radiative effects
20 /program	fortran code and ancillary data to create MACv2 aerosol properties
21 /program_force	fortran code and ancillary data to determine MAC aerosol radiative effects
22 /retrieval	MACv2 fields for under-determined solar reflection based AOD retrievals
23 /spectral	2005 optical data at 3 different spectral resolutions: 20, 30 (RRTM),31 bands
24 /time	same as in /spectral ... but data for different years (from 1850 to 2100)

25

26

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28

29 This study relied on observational data when possible. Central to the effort are data provided by the
30 ground-based sunphotometer network of AERONET lead by B. Holben and the MAN network lead by A.
31 Smirnov. Also satellite data of the MODIS and AATSR sensors were applied to quantify aerosol indirect
32 effects. Hereby in particular CDNC retrievals contributed by D.Grosvenor, J.Rausch and M. Christensen
33 and analysis work by J.Müsse, who created all figures in Appendix A are acknowledged. Another
34 essential element to this study is global model output from simulations with bottom-up processing in
35 aerosol modules as part of the AeroCom initiative lead by M. Schulz and M. Chin. An ensemble median
36 provides data on spatial context, estimates on aerosol anthropogenic fractions (also as a function of
37 time) and aerosol vertical distribution. Thus, all modeling groups contributing to AeroCom experiments
38 are acknowledged. Finally this work was support by EU-projects, in particular the FP7 EU-Bacchus
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40 lead by T. Popp and G. de Leeuw and coordinated by S. Pinnock.

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Appendix A the satellite based AODf vs CDNC relationship

Observational relationships, which employ regional associations between satellite retrieved properties for fine-mode AOD (AODf) and cloud droplet number concentration (CDNC) allow to associate increases in AODf from anthropogenic sources with increases in CDNC. For selected AODf ranges the median CDNC value is determined and then all median CDNC values are combined to result in logarithmic curve fit. By applying the curve twice locally, once for the present-day AODf and once for the pre-industrial AODf an associated CDNC increase is determined. Hereby the CDNC increase depends on the AODf difference and on the AODf (pre-industrial) background. Further assuming that the cloud water of the affected cloud remains constant, CDNC increases are easily converted into drop size reductions, the needed input to simulate associated increases to the planetary albedo, the Twomey effect (Twomey, 1974), in off-line radiative transfer simulations.

AODf and CDNC cannot be retrieved by the same sensor at the same time. However spatial associations within larger regions are expected to offer insights on potential relationships. Retrieval averages are matched for relatively large 1x1 degree in latitude/longitude regions. This is in part done to avoid exaggerations in associations, as local indirect effects (e.g. ship pollution impacts) are usually weaker in the context of larger spatial scales (Coakley and Walsh, 2002). For better statistics also only monthly averages are matched. Their use seems justified, because at these large spatial scales monthly associations are almost identical to those using daily data instead (Christensen, private communication).

Associations between AODf and CDNC were only considered over oceanic regions (where satellite retrievals for both properties are more reliable due to the dark background) and only when both sufficient signal and quality could be assured. Thus, only CDNC retrievals for overcast conditions under near nadir views are considered. The investigated matches include two different MODIS sensor based CDNC retrievals for an entire year (collection 5.1, for year 2007 provided by Dan Grosvenor, Leeds and collection 6.0 for year 2008 provided by John Rausch, Vanderbilt) with matching AODf data from NASA's LAADS site and an ATSR sensor based CDNC retrieval (for year 2008 provided by Matt Christensen) for a single season with matching AODf data by RAL's ORAC retrieval. 1x1 degree regions with valid matches between AODf and CDNC data are illustrated in Figure A1.

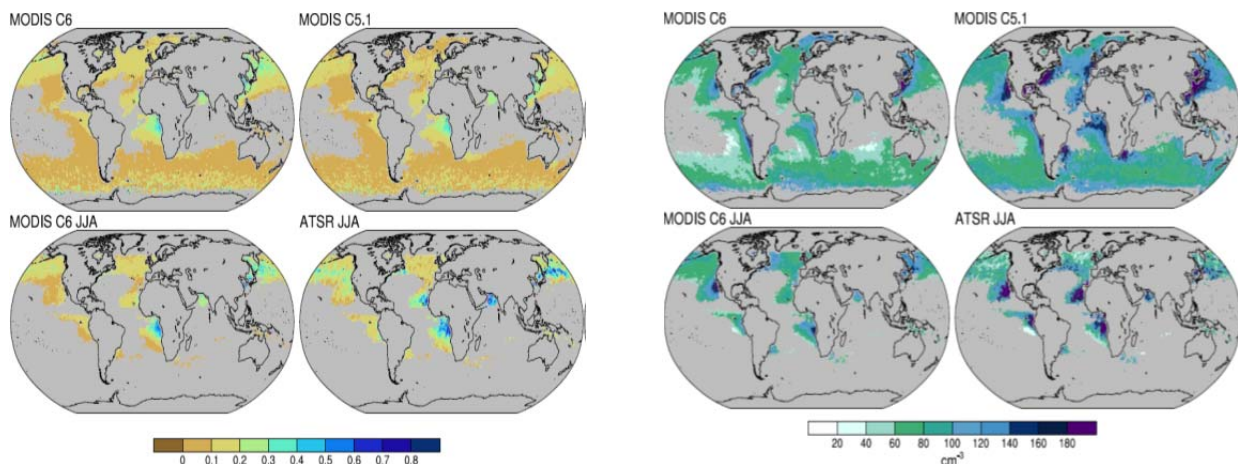
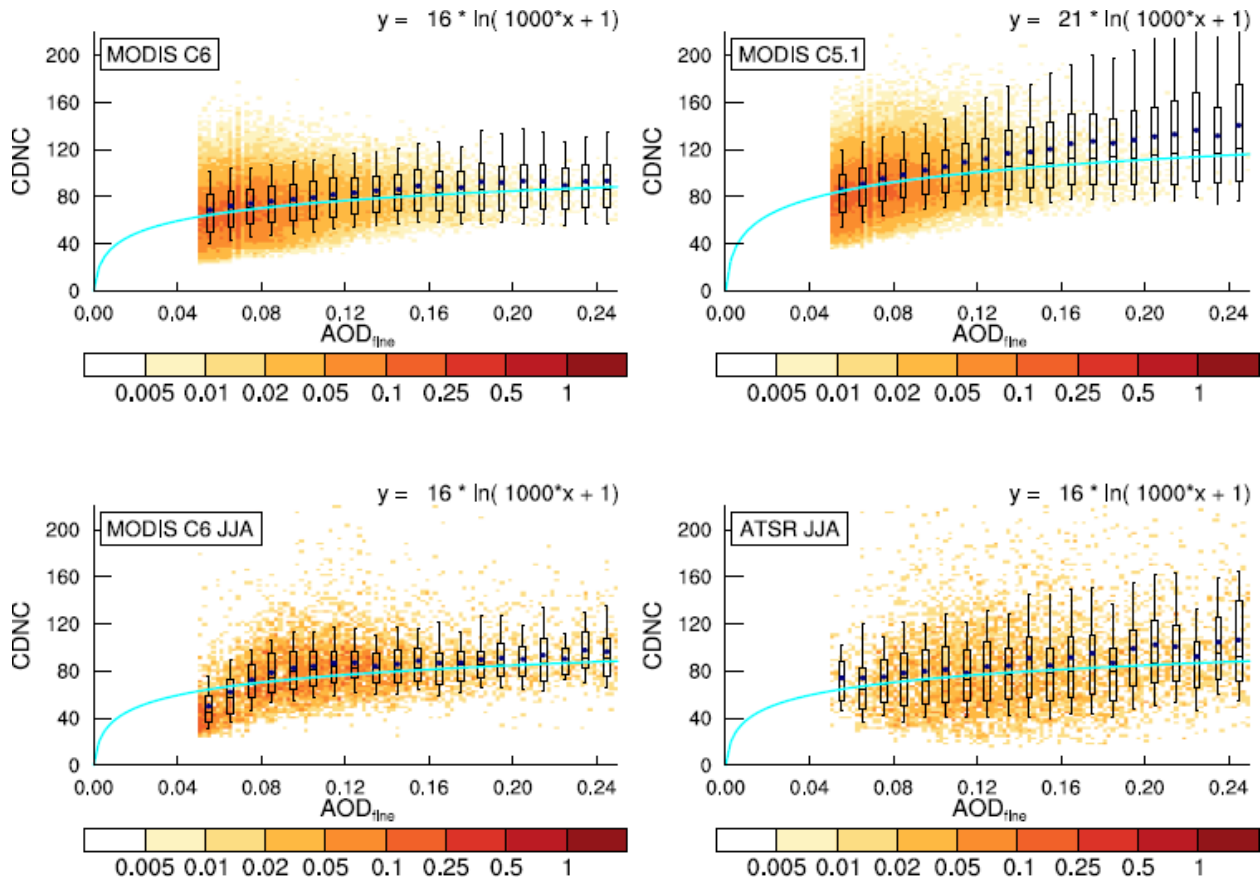


Figure A1 Annual maps (top) and a JJA seasonal maps (bottom) for AODf (left block) and CDNC (right block) for different retrievals with MODIS and ATSR sensor data.

1 Between the three different retrievals for CDNC and AODf there are often large absolute
 2 differences. The more important relative differences (as expressed by the logarithmic shapes,
 3 constructed from connection median CDNC points in AODf sub-bins), however, are smaller. The
 4 resulting logarithmic fits along with the data scatter are presented in Figure A2. In that figure the AODf
 5 to CDNC associations of all available months and locations are combined in a single plot.
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 7

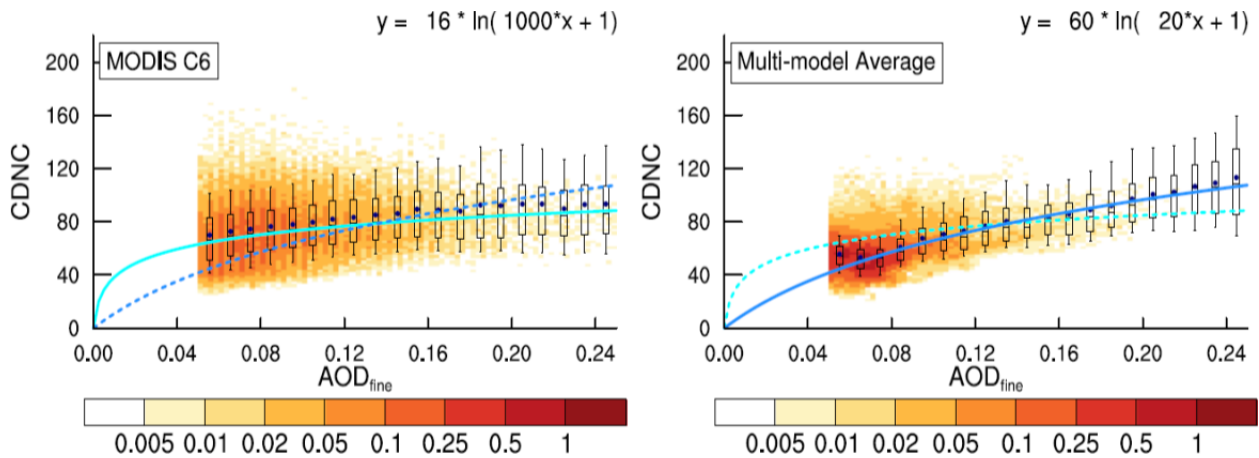


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 10 **Figure A2** AODf vs CDNC relationships for MODIS 6 (year 2008, upper left), for MODIS 5.1 (year 2007,
 11 upper right), for MODIS 6 (JJA of year 2008, lower left) and ATSR (JJA of year 2008, lower right). Matches
 12 were removed for mid-visible AODf values smaller than 0.05 (due to too poor signal to noise ratios) and
 13 larger than 0.25 (due to poor statistics). For individual AOD bins box-boundaries indicate the upper and
 14 lower quartiles, star symbols indicate averages and horizontal lines indicate median values. Logarithmic
 15 functions (displayed on top and illustrated by light blue lines) were fitted to the median bin values.
 16
 17

18 The fits for the four scatterplots follow an expected log-normal fit as for a given AODf increase
 19 the associated CDNC increase will be smaller the higher the background AODf due to nuclei saturation.
 20 While there are differences to the pre-factors of the fitting functions, the multipliers for AODf (x)
 21 are identical (actually the same multiplier of 1000 applies well for all sensor data). The pre-factor
 22 cancels out when determining CDNC increase factors, because these increase factor are based on the ratio of

1 two applications (CDNC [natur+anthrop] / CDNC [anthrop]). Thus, for the different satellite sensor data
 2 the derived CDNC factor increases from the different sensor data or data-subsets are basically identical.

3 The agreement for the (observational) fit functions for different satellite sensor data is quite in
 4 contrast to the diversity and to an (on average) much stronger Twomey effect in global modeling. These
 5 results are based on an analysis of output from nine global models with complex aerosol schemes that
 6 participated in the AeroCom (<http://aerocom.met.no>) indirect experiment. For the comparisons, the
 7 model simulated output for AODf and CDNC was sub-sampled at the same locations of the satellite
 8 matches. Scatter plots and associated fit functions between observational data (here MODIS coll.6) and
 9 the AeroCom model ensemble average are presented in Figure A3.



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 14 **Figure A3** AODf vs CDNC relationships as in figure A2 here based on a satellite retrieval (left, here MODIS
 15 6) and on the AeroCom model ensemble average (right). Note the much steeper slope in global modeling
 16 (dark blue lines) which indicates a much stronger Twomey effect by global modeling than by satellite
 17 retrieval based ‘observations’ (light blue lines).

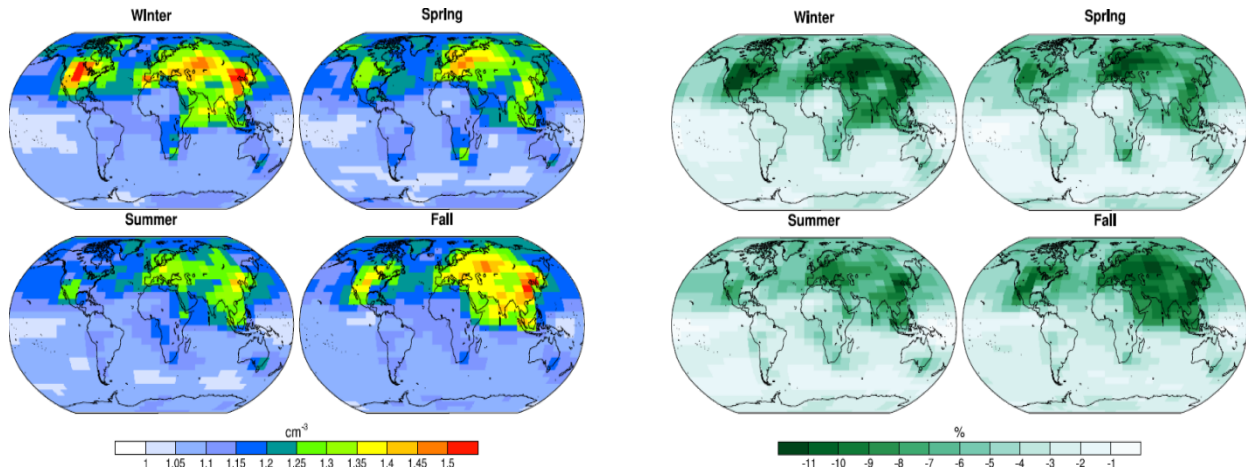
18
 19
 20 The ‘+1’ security value in the logarithmic fit (to avoid negative CDNC values) for the fine-mode
 21 AOD relationship to CDNC is raised ‘+3’ to account for additional nuclei from coarse-mode aerosol.
 22 These contributions are particular important in fine-mode sparse regions over the southern oceans, to
 23 avoid potential overestimate for CDNC-factor increases. Thus:

$$24 \quad \text{CDNC, factor} = \ln(1000 \cdot \text{AODf}_{[\text{natural} + \text{anthropogenic}] + 3}) / \ln(1000 \cdot \text{AODf}_{[\text{natural}] + 3})$$

25
 26
 27 Applying today’s total (natural and anthropogenic) AOD and natural AOD of MACv2, the CDNC
 28 increase factors due to anthropogenic aerosol are determined. And with the assumptions that the water
 29 content remains unchanged these CDNC increases are easily converted into droplet radius reductions.

$$30 \quad \text{radius, factor (\%)} = 100 / [(\text{CDNC, factor})^{**}(1/3)]$$

31
 32
 33 The seasonal averages for CDNC factor increases and water cloud droplet radius reductions (in
 34 %) associated with today’s anthropogenic aerosol in the context of pre-existing aerosol are presented in
 35 Figure A4. There larger cloud droplet radius reductions are on the order of 10 %.



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Figure A4 seasonal CDNC factor increases (left) and associated droplet radius reductions in % (right) due to today's anthropogenic aerosol and the pre-industrial fine-mode background as defined by MACv2.

7 Appendix B MACv2 vs MACv1 and MACv2 vs MACv2-SP

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There are different MAC climatology flavors for aerosol optical properties in circulation. As impact differences are of interest, radiative effects at the TOA by today's anthropogenic AOD of the MACv2 climatology are compared to those when applying in off-line simulations (1) aerosol optical properties of the older MACv1 climatology (Kinne et al., 2013) or (2) aerosol optical properties of the plume approximation for anthropogenic AOD (Stevens et al., 2017, Fiedler et al., 2019) instead. Annual maps for direct (clear-sky and all-sky) aerosol effects at TOA are compared in Figure B1.

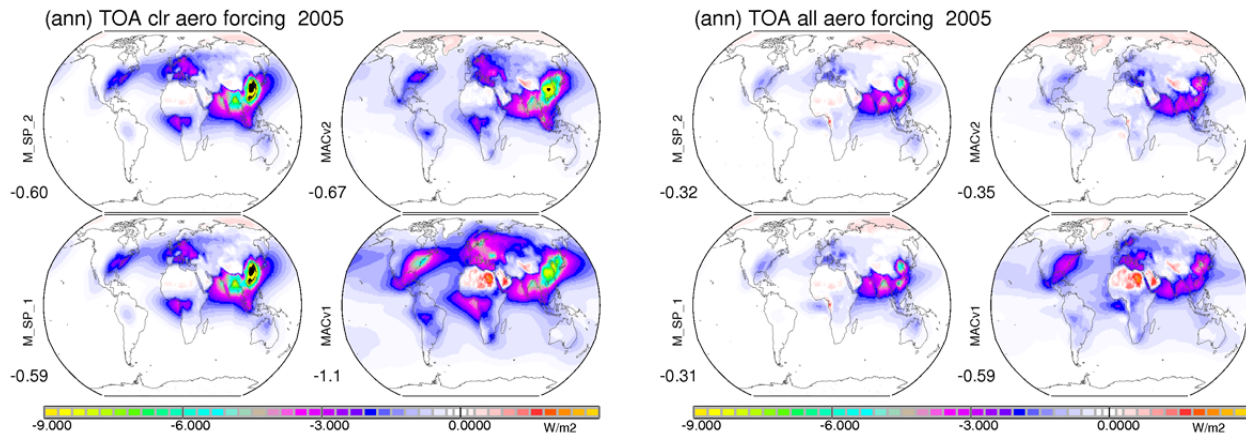


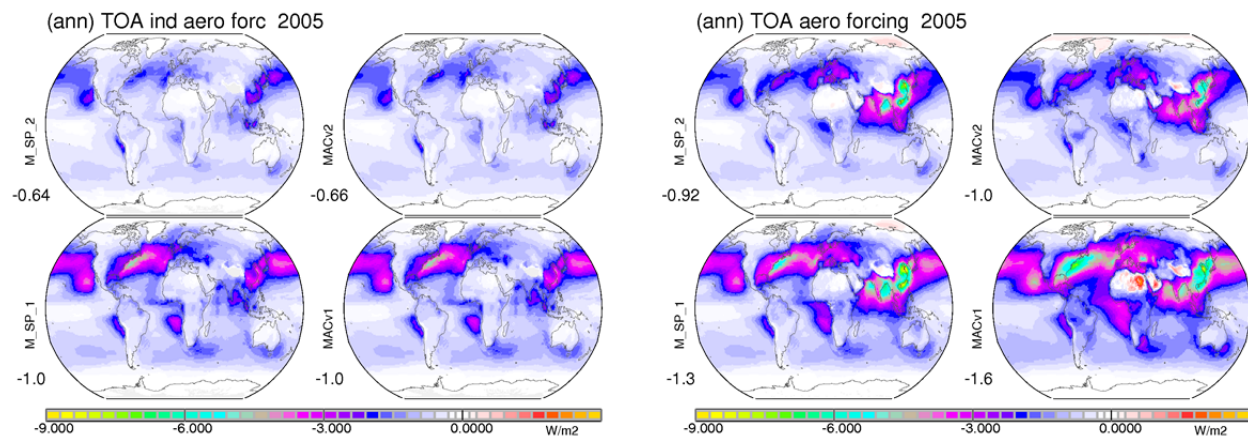
Figure B1. annual maps for present-day aerosol direct radiative forcing in W/m^2 at the TOA at clear-sky conditions (left block) and at all-sky conditions (right block) for MACv2-SP with MACv1 natural background (upper left) and with MACv2 natural background (lower left), for MACv2 (upper right) and MACv1 (lower right). Blue colors represent climate cooling and red colors indicate climate warming. Values below the labels indicate global averages.

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1 For the offline simulations with the MACv2-SP data with two different natural background
 2 conditions are assumed: one with a lower MACv1 natural background (M_SP_1) and one with a higher
 3 MACv2 natural background (M_SP_2).

4 The MACv2-SP plume approximation yields MACv2 similar cooling patterns for present-day
 5 anthropogenic aerosol. However, MACv2-SP maxima are stronger (e.g. China) and weaker (e.g. South
 6 America). In the MACv2-SP plume approximation, contributions in remote regions are lower or
 7 completely missing, as individual plumes with limited spatial domains (even when combined) do not
 8 cover the entire globe. The impact by using different natural backgrounds in MACv2-SP (once using
 9 MACv1 natural aerosol and once using MACv2 natural aerosol) has only a small impact on the direct
 10 forcing. Globally averaged, both clear-sky and all-sky TOA cooling in MACv2-SP are ca 10% smaller than
 11 in MACv2, mainly as the mid-visible global average mid-visible anthropogenic AOD in MACv2-SP is only
 12 0.028 compared to 0.031 in MACv2.

13 In the MACv1 climatology regional contributions for the direct forcing differ from MACv2 mainly
 14 due to differences in anthropogenic AOD regional strengths. MACv1 falsely allowed significant
 15 anthropogenic aerosols over the Sahara (associated with significant warming over the bright desert)
 16 along with too much anthropogenic aerosol over the US and Europe (for extra cooling). In MACv1 also
 17 anthropogenic aerosol over SE Asia is likely too low (for missing cooling). Globally averaged though, the
 18 direct forcing between MACv2 and MACv1 are similar as effects of a larger anthropogenic AOD of 0.040
 19 in MACv1 (compared to 0.031 in MACv2) are compensated by significant warming over the Sahara.
 20 Annual maps for today's indirect effects at the TOA by anthropogenic aerosol are compared in Figure B2.



23 **Figure B2.** annual maps for present-day aerosol indirect radiative forcing in W/m^2 at the TOA at (left
 24 block) and for the combined direct and indirect radiative forcing (right block) for MACv2-SP with MACv1
 25 natural background (upper left) and with MACv2 natural background (lower left), for MACv2 (upper
 26 right) and MACv1 (lower right). Values below the labels indicate global averages.

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 30 In all approaches, only indirect (Twomey) cooling effects at the TOA are considered and
 31 approximated based on the 'observed' satellite relationship between AODf and CDNC as explained in
 32 Appendix A. The fit function is applied twice, for natural AODf and for total AODf (anthropogenic plus
 33 natural), to extract a CDNC increase factor. In MACv2 and MACv1 required AODf values (natural AODf

1 and total AODf) are defined by the climatology. In MACv2-SP only anthropogenic AODf maps and
2 already pre-calculated associated CDNC increase factors are provided with simplified assumptions to the
3 natural AODf background (Stevens et al., 2017).

4 In MACv2-SP the anthropogenic AOD has no global coverage so that in regions with no
5 anthropogenic AOD also no indirect aerosol effects are possible. This is the main reason that the globally
6 averaged indirect TOA cooling in MACv2-SP is only ca 65% of the indirect TOA cooling by MACv2 (despite
7 a much stronger MACv2-SP indirect response over the Pacific). For MACv1, in contrast, the indirect TOA
8 cooling is ca 40% larger than in MACv2. The main reason here is the larger anthropogenic AODf in
9 combination with a reduced natural AODf. Both factors lead to larger CDNC increase factors and
10 stronger indirect TOA cooling when applying the (retrieval based) fit function.

11 In Figure B2 also annual maps for the predicted combined (direct and indirect) TOA cooling by
12 today's anthropogenic AOD are compared. The MACv2 climatology suggests (when globally averaged) a
13 combined present-day climate cooling near -1.0 W/m^2 , with the larger contribution from indirect
14 effects. The associated uncertainty involves a -0.7 to -1.6 W/m^2 range a for the present-day aerosol
15 forcing. To lower that range a more certain the pre-industrial reference and a better representation of
16 indirect effect are needed.

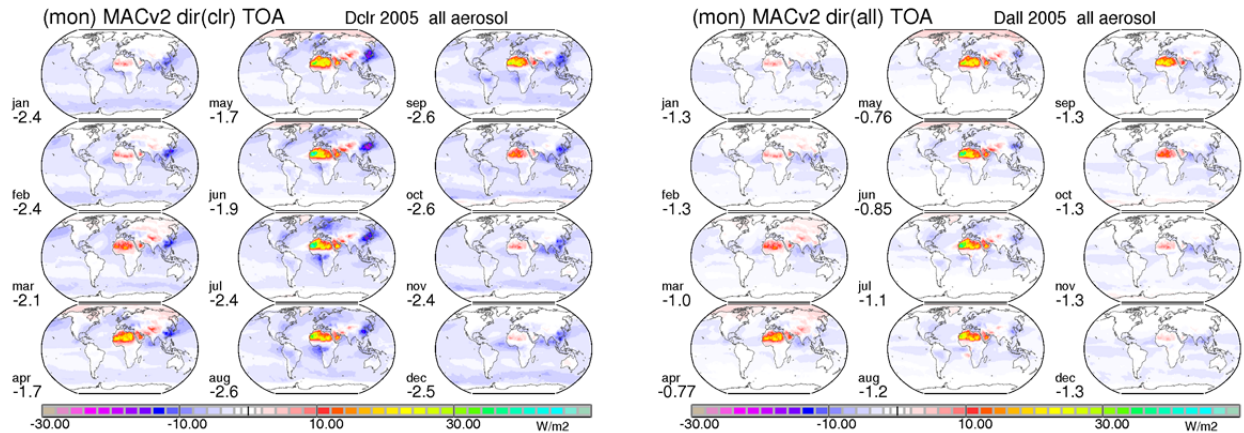
17 The main argument for more negative bound (-1.6 W/m^2) is that the anthropogenic fine-mode
18 fraction could be larger. An alternate anthropogenic scaling based on the AeroCom phase 1 emission
19 increases indirect effects (from -0.65) to -1.1 W/m^2 and direct effects (from $-.35$) to -0.50 W/m^2 for a
20 combined -1.6 W/m^2 cooling. However, the alternate larger anthropogenic fine-mode fraction applies a
21 year 1750 reference. Thus, such a negative aerosol forcing is very unlikely for a year 1850 reference.

22 The main argument for the less negative bound (-0.7 W/m^2) is that the lower bound of the direct
23 effect (which is better constrained than the indirect effect) is estimated at -0.2 W/m^2 and that the lower
24 bound for the indirect effect is estimated at -0.5 W/m^2 . For the present-day direct effect a less negative
25 -0.2 W/m^2 cooling can be explained with a higher anthropogenic fraction of absorbing BC AOD than
26 applied for the fine-mode AOD. For the present-day indirect effect there is a lot of uncertainty. If only
27 the first indirect effect is considered at least a cooling of -0.5 W/m^2 should be expected (in that context
28 it should be noted that the less negative indirect effect MACv2-SP does not qualify as lower bound due
29 to its incomplete global coverage for anthropogenic AOD).

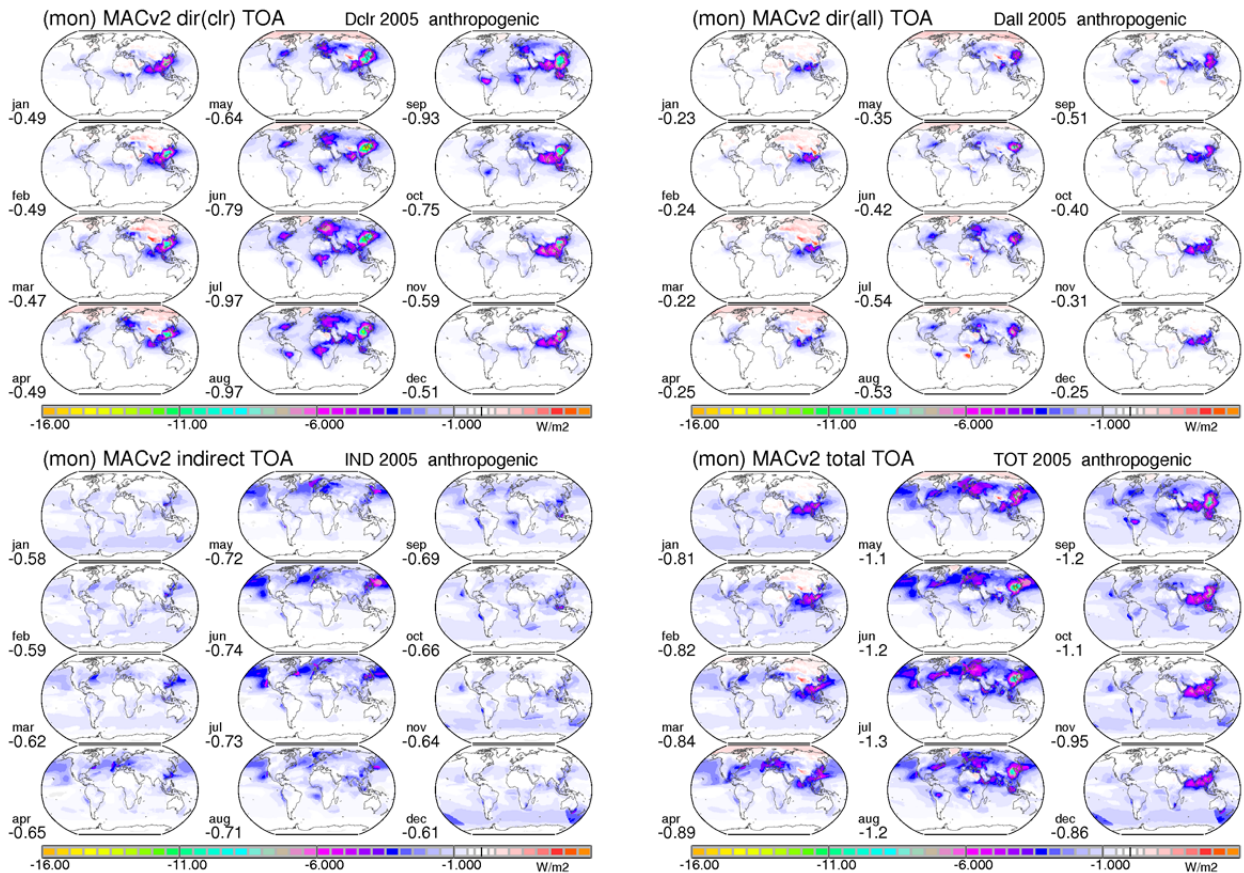
31 **Appendix C monthly TOA forcing**

32
33 Monthly maps illustrate typical variations over the year for present-day aerosol radiative effects
34 at the top of the atmosphere (TOA) for total aerosol in Figure C1 and anthropogenic aerosol in Figure C2.

35 Figure C1 presents for total aerosol the MACv2 associated radiative TOA effects. Monthly maps
36 are presented for clear-sky (cloud-free) and all-sky conditions. Note that the effects in Figure C1 include
37 infrared greenhouse effects by elevated coarse mode dust. For total aerosol, the global average effect is
38 a climate cooling, despite intense climate warming over the Sahara, especially from April to September.
39 The presented aerosol effects at all-sky conditions do not include anthropogenic indirect effects which
40 would increase the TOA cooling by an additional -0.65 W/m^2 .



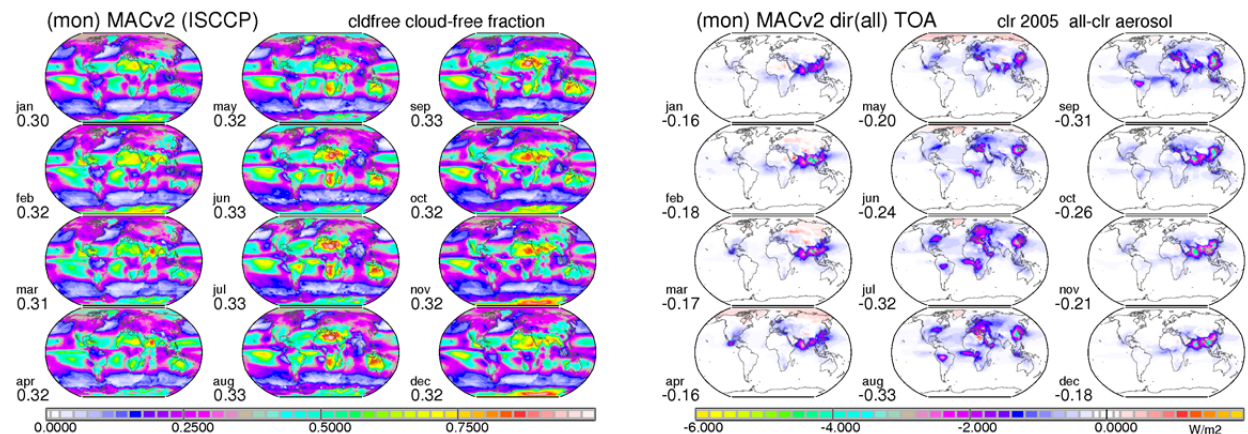
1
 2 **Figure C1.** monthly maps for the direct radiative forcing at the TOA by present-day total aerosol under
 3 clear-sky conditions (left block) and all-sky conditions (right block). Blue colors indicate climate cooling
 4 and red colors indicate climate warming. The infrared greenhouse effects by elevated coarse-mode
 5 mineral dust aerosol are included. Values below the labels indicate global averages.
 6



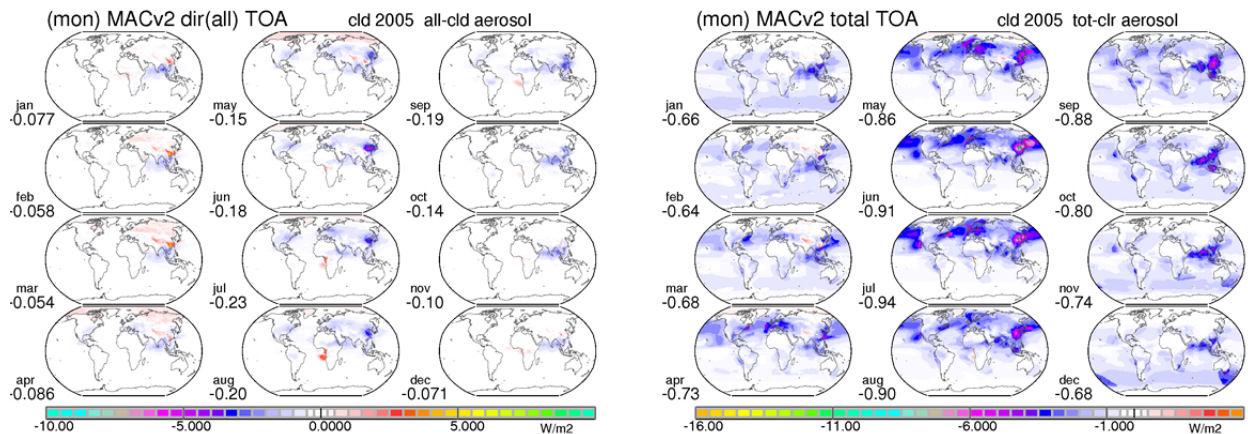
7
 8
 9 **Figure C2.** monthly maps for radiative effects by present-day anthropogenic aerosol. Compared are
 10 direct radiative forcing by today's anthropogenic aerosol under clear-sky conditions (top, left block),
 11 direct radiative effects under all-sky conditions (top, right block), indirect effect (bottom, left block) and
 12 the combined (direct and indirect) effect (lower, right block). Blue colors indicate climate cooling and red
 13 colors indicate climate warming. Scales of all four blocks are identical to better compare contributions
 14 and seasonal dependencies. Values below the labels indicate global averages.

1 Figure C2 presents for anthropogenic aerosol the MACv2 associated radiative TOA effects.
 2 Monthly maps are presented for the direct effect at cloud-free condition (Dclr) and all-sky conditions
 3 (direct forcing, D_{all}), for a (Twomey based) indirect forcing (IND) and for the combined (direct and
 4 indirect) forcing (TOT). In Figure C2 monthly maps were given the same scale for easier comparisons.
 5 They illustrate that for present-day aerosol forcing indirect contributions (IND) dominate over direct
 6 contributions (D_{all}) and that contributions are largest for the NH summer season with indirect effects
 7 globally slightly larger from May to August and direct effects globally largest from July to September.

8 In terms of the aerosol radiative forcing some applications prefer to separate between
 9 clear-sky and cloudy-sky contributions. A requirement for such a separation is the cloud-free fraction of
 10 Figure C3. Figure C3 also presents the clear-sky present-day contributions to the (direct) all-sky or to the
 11 combined (direct + indirect) forcing with MACv2. The corresponding cloudy-sky contributions to the
 12 present-day (direct) all-sky and to the combined (direct + indirect) forcing are presented in Figure C4.



15 **Figure C3.** monthly maps for the clear-sky fraction (left block) and monthly maps for the present day
 16 clear-sky radiative TOA radiative forcing that contributes to the all-sky forcing.
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 18
 19

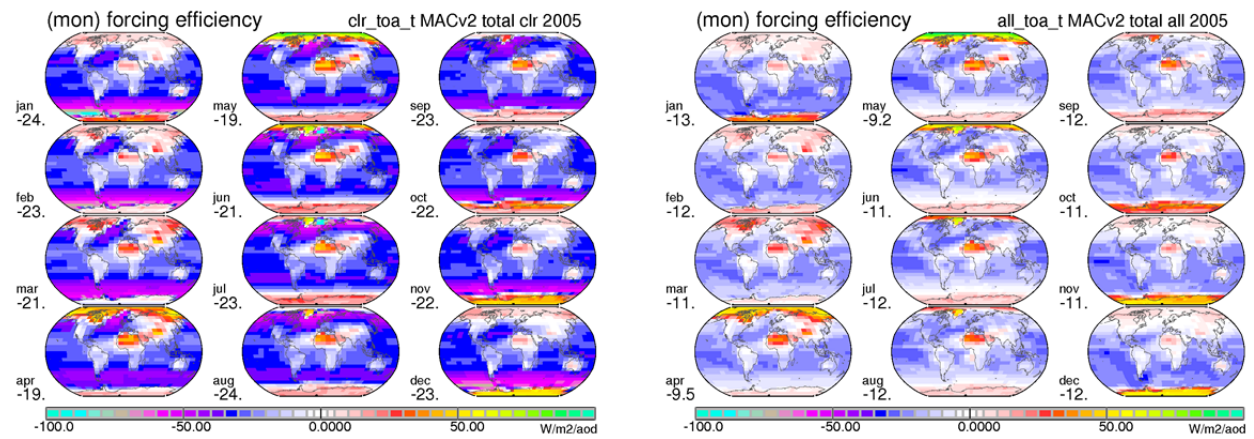


20 **Figure C4.** monthly maps for monthly maps for the present-day cloudy-sky TOA radiative forcing that
 21 contributes to the all-sky forcing (left block) and to the combined (direct and indirect) forcing.
 22
 23
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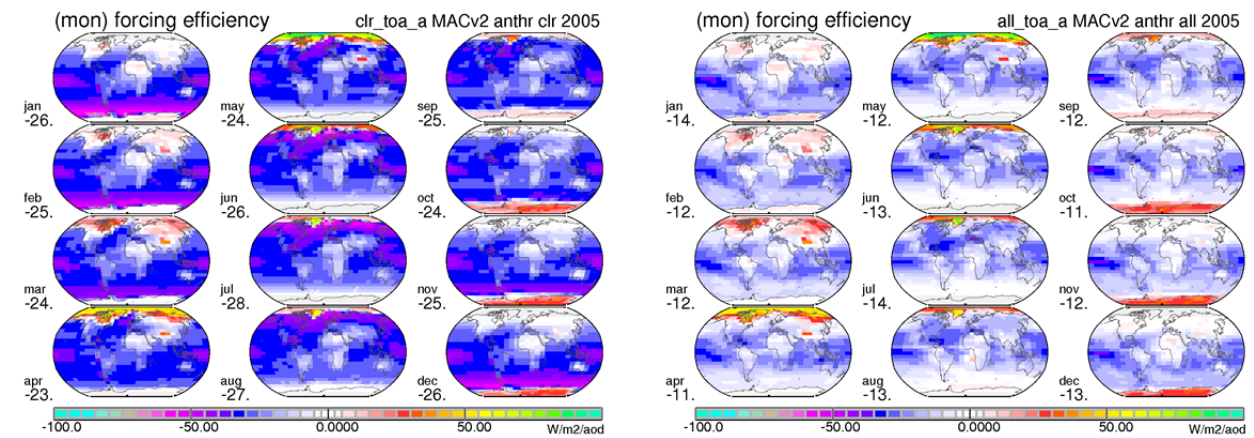
1 The radiative forcing in cloudy regions (both with and without the first indirect effect) is much
 2 larger during the boreal summer season compared to the boreal winter. The absolute global average
 3 seasonal range is similar for the direct and for the indirect effect, with more sunshine, less low altitude
 4 cloud decks and more anthropogenic aerosol loads during higher latitude summer seasons.

6 Appendix D *monthly TOA direct forcing efficiencies*

7
 8 Monthly maps for present-day TOA forcing efficiencies (per unit AOD) at clear-sky and all-sky
 9 conditions are shown for total aerosol in Figure D1 and for anthropogenic aerosol in Figure D2.



12
 13 **Figure D1.** monthly maps for the direct radiative forcing efficiency (per unit AOD) at the TOA by present-
 14 day total aerosol at clear-sky (left block) and all-sky conditions (right block). Blue colors indicate climate
 15 cooling potential and red colors warming potential. Values below the labels indicate global averages.

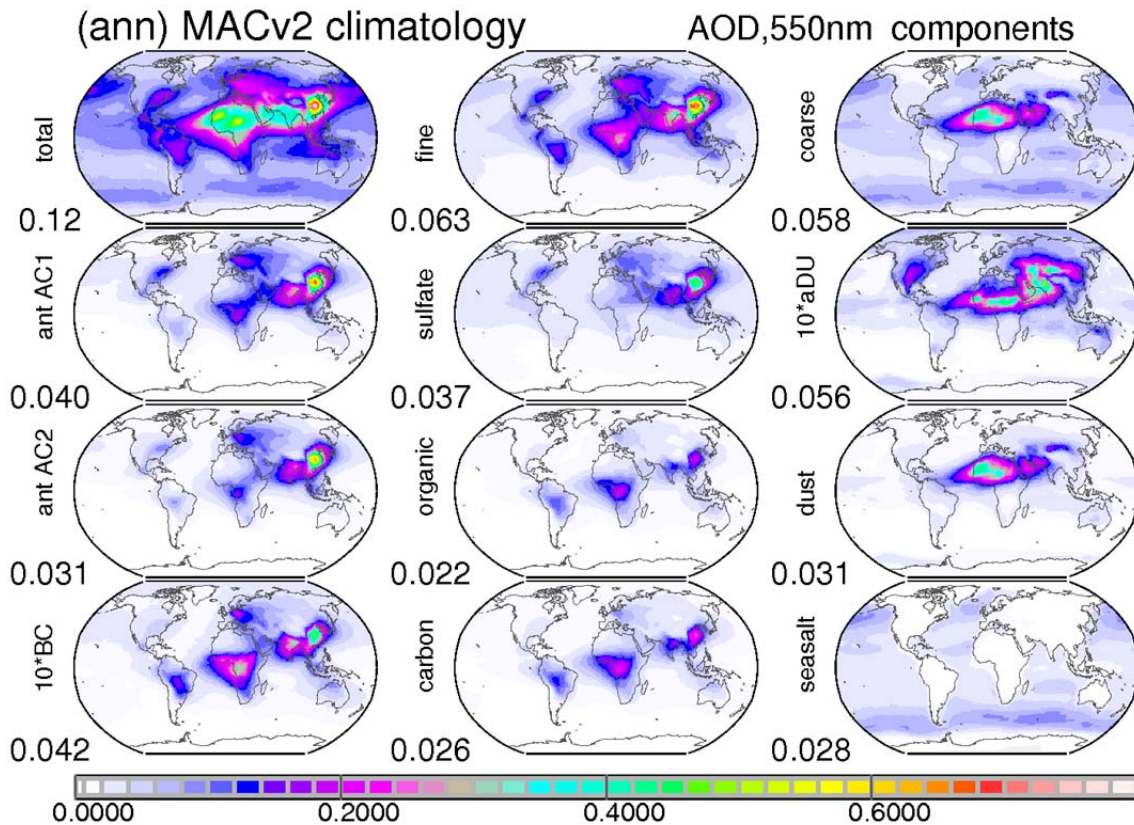


18
 19 **Figure D2.** monthly maps for the direct radiative forcing efficiency (per unit AOD) at the TOA by present-
 20 day anthropogenic aerosol at clear-sky (left block) and all-sky conditions (right block). Blue colors
 21 indicate climate cooling potential and red colors warming potential. Values indicate global averages.

1 The global average forcing efficiencies (per unit AOD) for total and anthropogenic aerosol are
 2 similar and not only in their global averages (near -22 W/m² at clear-sky conditions and near -11W/m²
 3 at all-sky conditions) but also their spatial patterns relatively stable over the year. Noteworthy is the
 4 switch in sign over Asia to positive values during the winter and spring due to snow cover and only for
 5 total aerosol the strong positive values over northern Africa.
 6

7 **Appendix E component direct radiative effects**

8
 9 With the attribution of optical and microphysical properties to (via size and refractive index)
 10 pre-defined aerosol types, type (or component) contributions are assigned such that their mixture is
 11 consistent with (local monthly data for) size-mode associated MACv2 mid-visible aerosol properties for
 12 AOD and AAOD. The resulting MACv2 component AOD distributions are presented in Figure E1.
 13
 14



15 **Figure E1** annual average AOD maps for present-day tropospheric aerosol for total aerosol (top left) and
 16 contributions by fine-mode sizes (top center) and coarse-mode sizes (top right). In addition, consistent
 17 with mid-visible absorption data, component AOD values were assigned for each size mode. Fine-mode
 18 AOD is divided into contributions by BC (soot, here multiplied by 10), OC (organic matter) and SU (where
 19 SU represents non-absorbing fine-mode aerosols). The coarse mode AOD is split into contributions by
 20 sea-salt and dust. In addition, annual AOD maps are presented for total carbon (OC+BC), for present day
 21 anthropogenic dust ('aDU', here multiplied by 10) and estimates for present day anthropogenic (fine-
 22 mode) AOD: 'ant AC2' of MACv2 and 'ant AC1' of MACv1. Lower left values indicate global averages.
 23

1 Considered components for the fine-mode are sulfate (SU - representing the fine-mode non-
 2 absorbing type), organic matter (OC), soot (BC) and for the coarse-mode sea-salt (SS) and mineral dust
 3 (DU). Hereby the size for SU and DU is allowed to vary, to satisfy MACv2 prescribed data for the fine-
 4 mode effective radius and the coarse mode absorption, respectively (Kinne, 2019). Also presented are
 5 two present-day estimates for (fine-mode) anthropogenic AOD based on fine-mode fraction scaling with
 6 processed emissions in global models $[(AOD_{f,pd} - AOD_{f,pi}) / AOD_{f,pd}]$. One estimate (antAC1) applies a
 7 fine-mode AOD fraction map based on AeroCom phase 1 emission data (Dentener et al., 2006), which is
 8 tied to a year 1750 reference and applied in MACv1 (Kinne et al, 2013). The other estimate (antAC2)
 9 applies a fine-mode AOD fraction map based on CMIP5 (AeroCom phase 2) emission data (Lamarque et
 10 al., 2010), which is tied to a year 1850 reference and applied in MACv2 (Kinne, 2019). In addition, an
 11 estimate for present day anthropogenic (coarse-mode) dust (aDU) is presented by applying a present
 12 day anthropogenic dust AOD fraction map based on a satellite data analysis (Ginoux et al, 2012) to the
 13 dust AOD of MACv2. This AOD for anthropogenic dust (multiplied by 10 in Figure E1) is an average
 14 almost an order of magnitude smaller than estimates for the fine-mode anthropogenic AOD. And since,
 15 in addition, solar and infrared contributions to the radiative forcing by dust largely cancel, coarse-mode
 16 anthropogenic contributions were ignored for MACv2. Still, even with anthropogenic dust, the present-
 17 day fine-mode anthropogenic AOD (0.03 to 0.04) is highly uncertain due to differences in choices for
 18 reference strength, including the reference year.

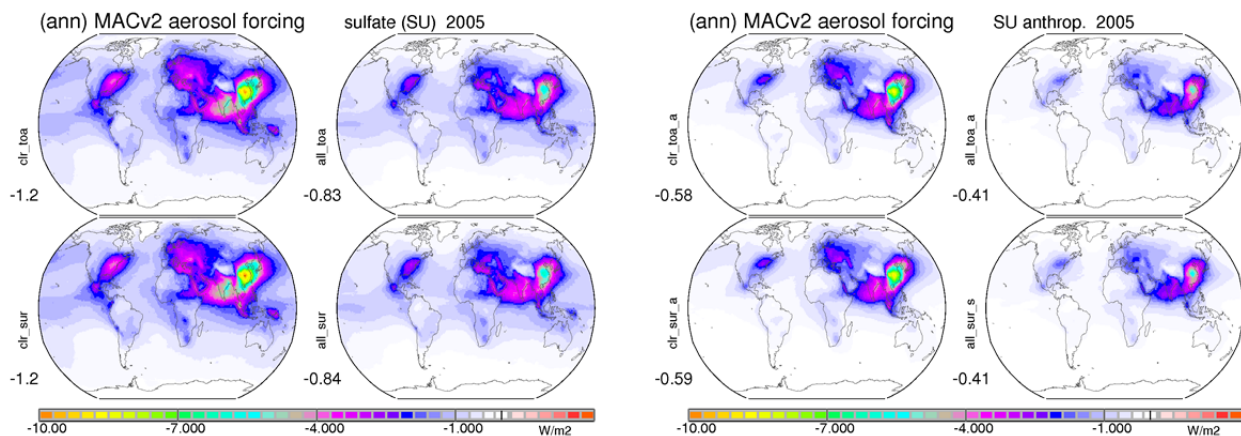
19

20 **SU (non-absorbing fine-mode)**

21 Annual maps by present-day total and anthropogenic non-absorbing fine-mode aerosol (SU - to
 22 cover impacts mainly from sulfate and nitrate, but also from small size sea-salt) are shown in Figure E2.
 23 Anthropogenic SU direct forcing (-.41 W/m²) is unevenly distributed and stronger near sources. Local
 24 radiative effects at TOA and surface are basically identical, due to a lack in relevant solar absorption.

25

26

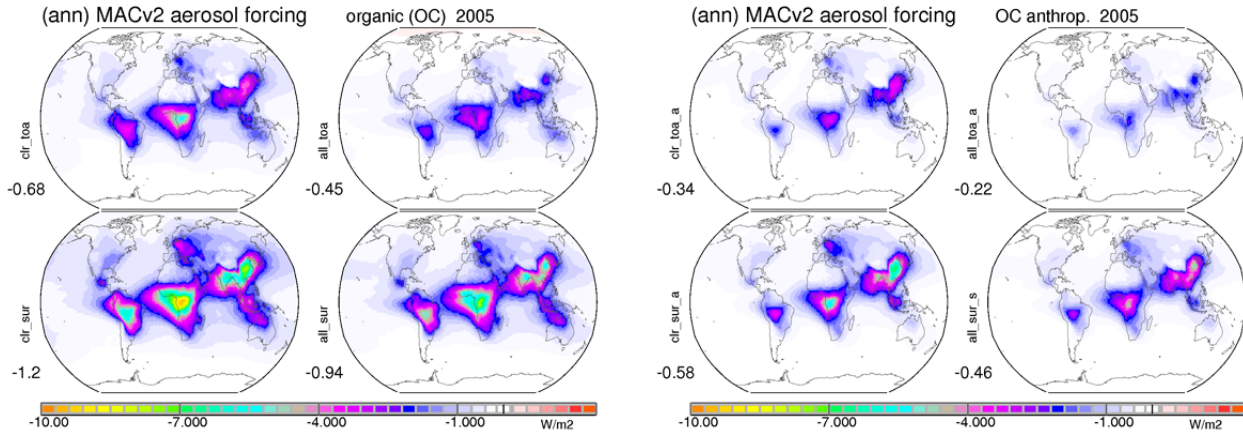


27 **Figure E2.** Annual maps for present-day radiative effects of total (left block) and anthropogenic (right
 28 block) are shown for scattering fine-mode aerosol (SU). Each block displays impacts at clear-sky (left
 29 column) and all-sky conditions (right column) at both TOA (top row) and surface (bottom row). Blue to
 30 purple, green, yellow and red colors indicate an increasing cooling. Values at labels are global averages.

31

1 **OC (organic matter)**

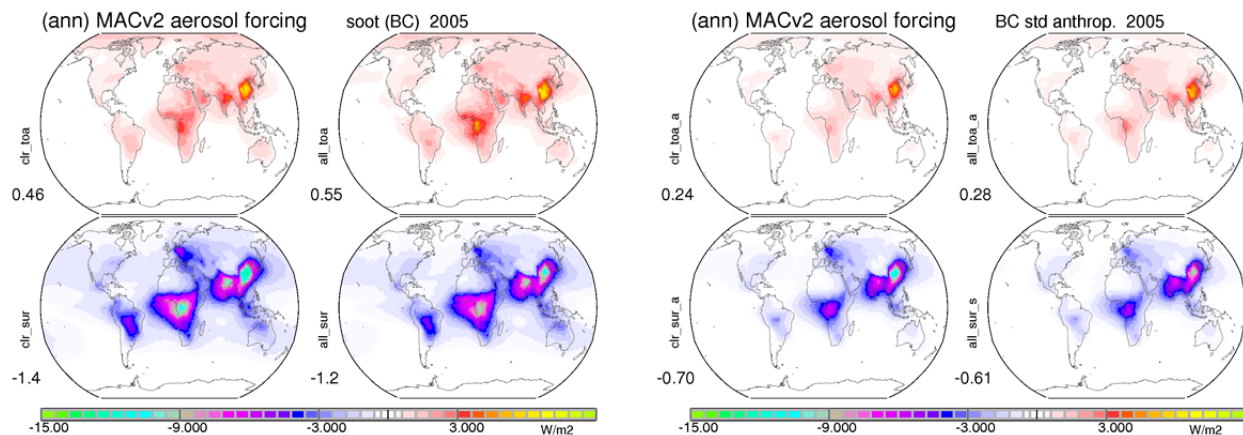
2 Annual maps by present-day total and anthropogenic organic matter are presented in Figure E3.
 3 Anthropogenic OC direct forcing (-0.22 W/m^2) is unevenly distributed and relative strong near sources of
 4 pollution and wildfire regions. Due to assumed solar absorption in the visible and especially towards and
 5 in the UV, the cooling at the surface is larger than the cooling at TOA.
 6



7
 8 **Figure E3.** annual average maps for present-day radiative effects of total (left block) and anthropogenic
 9 (right block) organic matter (OC). Each block displays impacts at clear-sky (left column) and all-sky
 10 conditions (right column) at both TOA (top row) and surface (bottom row). Blue to purple, green and
 11 yellow indicate an increasing cooling. Values below the labels are global averages.
 12
 13

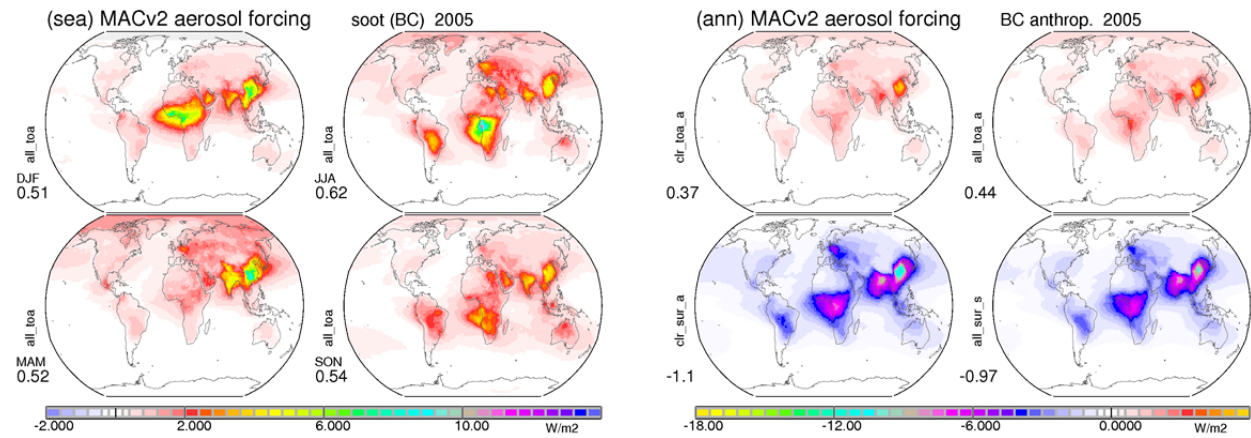
14 **BC (soot)**

15 Annual maps for TOA warming and surface cooling by present-day total and anthropogenic soot
 16 (BC) are shown in Figure E4. Anthropogenic BC direct radiative effects are larger over wildfire and
 17 pollution regions. TOA warming with clouds (all-sky) is larger, as BC dims the reflection of lower clouds.
 18



19
 20 **Figure E4.** Annual maps for present-day radiative effects of total (left block) and anthropogenic (right
 21 block) are shown for soot (or BC). Each block displays impacts at clear-sky (left column) and all-sky
 22 conditions (right column) at both TOA (top row) and surface (bottom row). Blue colors indicate a cooling
 23 (at the surface) and red colors a warming (at TOA). Values below the labels are global averages.

1 The seasonality of the TOA all-sky BC forcing and the BC radiative effects for an alternate larger
 2 (75% rather 50%) for the BC anthropogenic fraction are presented in Figure E5.

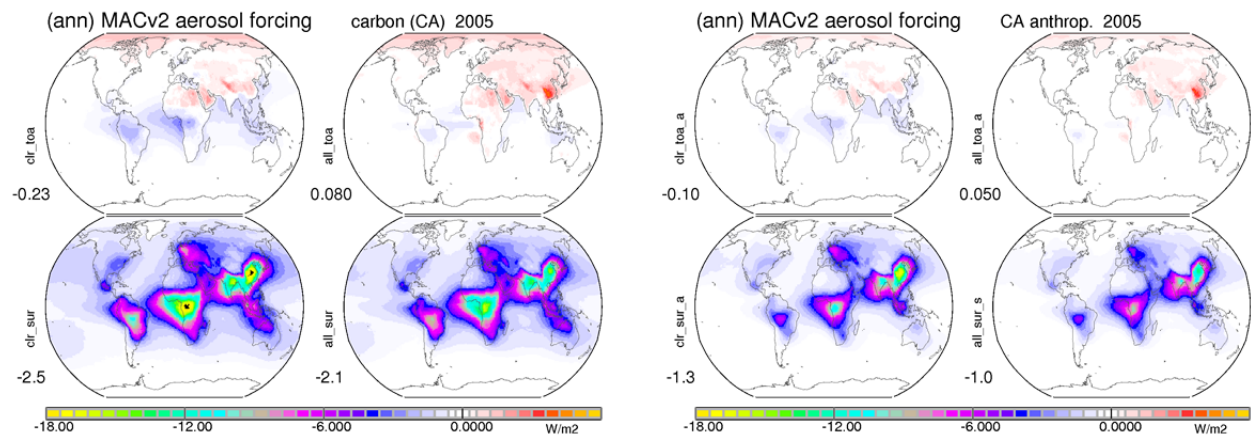


4 **Figure E5** Seasonally average maps for present-day BC radiative TOA all-sky effects at (left block) and
 5 anthropogenic BC radiative effects with a higher (75%) BC anthropogenic fraction compared to the 50%
 6 for BC radiative effects presented in Figure E4. Values below the labels are global averages.
 7

8
 9 Many regional aerosol absorption maxima have a strong seasonal flavor often associated with
 10 wildfire seasons (e.g. DJF over W. Africa, JJA and SON over central Africa and S. America) and pollution
 11 (e.g. MAM over S. Asia and E. Asia prior to the monsoon season). The applied larger anthropogenic BC
 12 fraction raised the TOA forcing by BC from +0.28 to +0.44W/m2.

13
 14
 15 **CA (carbon: OC+BC)**

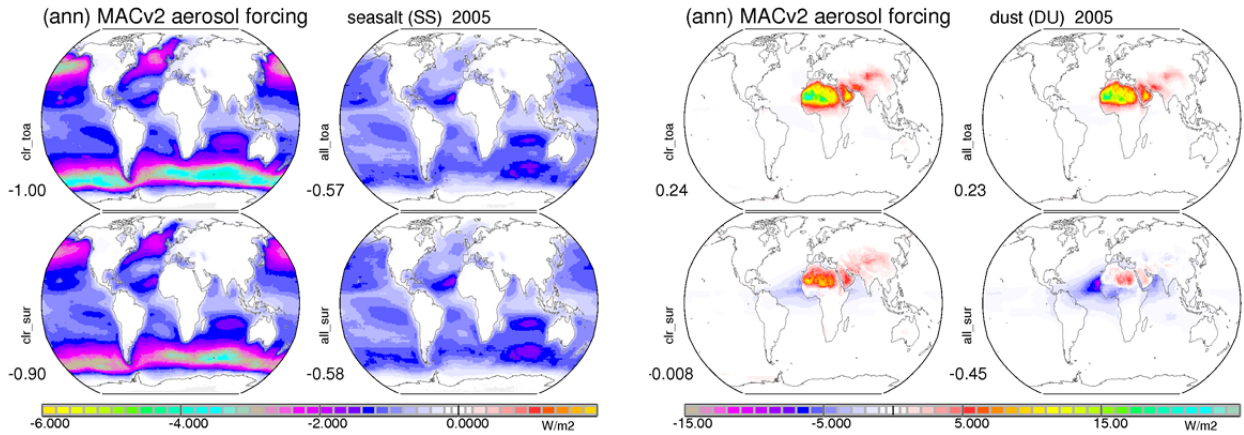
16 Annual maps for present-day direct radiative effects by total and anthropogenic carbon are
 17 shown in Figure E6. The anthropogenic carbon TOA response (+.05 W/m2) is almost climate neutral.
 18



19 **Figure E6.** Annual maps for present-day radiative effects of total (left block) and anthropogenic (right
 20 block) soot (or carbon (BC+OC)). Each block displays impacts at clear-sky (left column) and all-sky
 21 conditions (right column) at both TOA (top row) and surface (bottom row). Blue colors indicate a cooling
 22 (at the surface) and red colors a warming (at TOA). Values below the labels are global averages.
 23

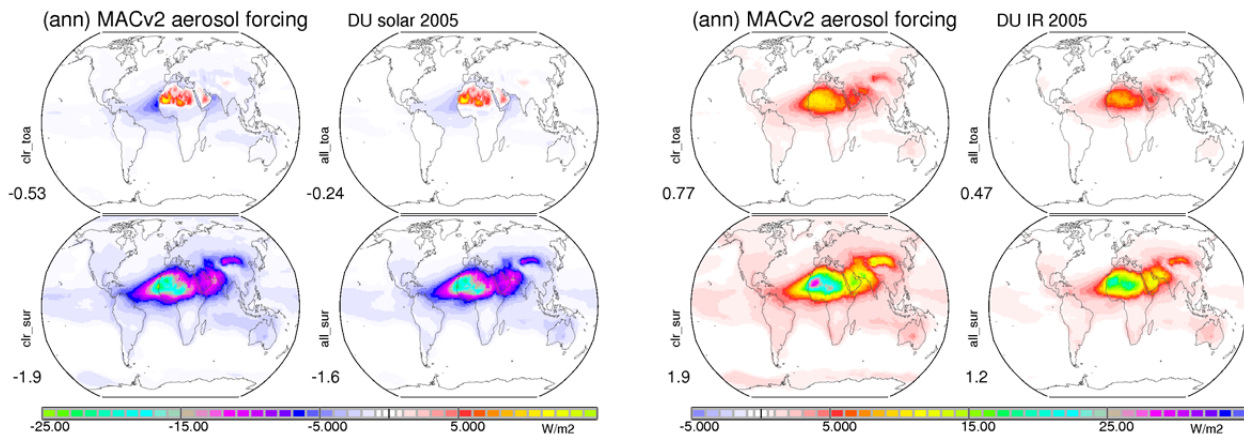
1 **SS (sea-salt) and DU (dust)**

2 Annual maps for direct radiative effects by coarse mode (natural) components of sea-salt and
 3 mineral dust are compared in Figure E7. For sea-salt cooling maxima at TOA and surface are sharply
 4 reduced with the presence of clouds. Thus the direct forcing associated with sea-salt is relatively small.
 5 Mineral dust displays a strong TOA warming over the Sahara. Even at the surface a net-flux increase is
 6 indicated. Both effects are associated with relatively large mineral dust AOD and dust particles sizes.
 7 Mineral dust effects are better understood by looking at solar and infrared contributions in Figure E8.
 8



9
 10 **Figure E7.** Annual maps for present-day TOA radiative effects of coarse mode sea-salt (SS, left block) and
 11 coarse mode mineral dust (DU, right block) at clear-sky (left column) and all-sky conditions (right
 12 column) both at TOA (top row) and surface (bottom row). Blue to purple colors indicate a cooling and red
 13 to yellow and green colors show a warming. Values below the labels are global averages. Note, that the
 14 color scale for sea-salt is 10 times smaller than the color scale for dust.

15
 16
 17 The mineral dust radiative effects are created by partially offsetting solar cooling and infrared
 18 warming effect. Solar warming is only created by larger dust sizes over bright (Sahara) surfaces.
 19

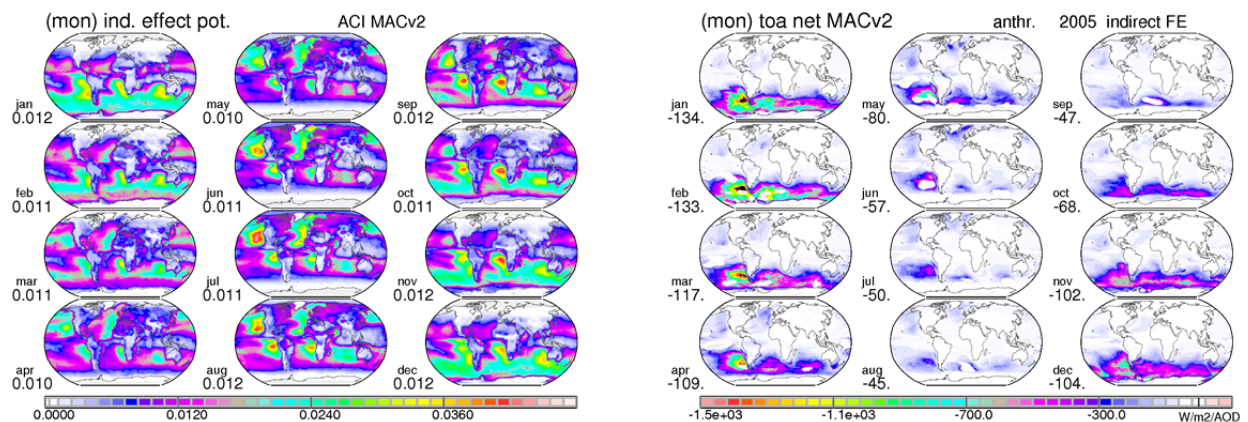


20
 21 **Figure E8.** annual averages maps for present-day radiative effects of coarse mode mineral dust (DU) in
 22 the solar spectral (left block) and the infrared spectral region (right block). Aerosol impacts are shown at
 23 clear-sky (left column) and all-sky conditions (right column) for both TOA (top row) and surface (bottom
 24 row). Blue colors indicate a cooling and red colors a warming. Values at the labels are global averages.

1 For the climatic relevant TOA response at the top of the atmosphere the present-day global
 2 average is a -0.24 W/m^2 cooling, despite the regional solar warming over the Sahara (as stronger
 3 absorbing larger mineral dust sizes dim the bright desert solar albedo). The infrared greenhouse effect
 4 at $+0.47 \text{ W/m}^2$ is much larger, so that the global average response for mineral dust is a $+0.23 \text{ W/m}^2$
 5 warming. The greenhouse effect loses (on average) its dominance for anthropogenic dust, as
 6 anthropogenic aerosol involves more moderate sizes. And also anthropogenic aerosol loads are closer to
 7 the surface. Thus extra anthropogenic dust on average behaves almost climate neutral.

9 **Appendix F aerosol indirect radiative forcing sensitivity**

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 11 Aerosol indirect radiative effects show spatial distribution patterns, which may surprise, as the
 12 strongest impacts (e.g. mainly the increase in planetary albedo) are usually not at locations where
 13 anthropogenic AOD values are largest. The aerosol indirect effects or forcing is strongly influenced by
 14 environmental properties. A lower surface albedo, a high percentage of low altitude cloud cover without
 15 higher altitude clouds, a moderate optical depth for highest susceptibility and the available sun-hours
 16 during a day all favor a stronger indirect response. Based on these factors as summarized in Table F1
 17 monthly indirect forcing potential maps are presented in Figure F1 along with monthly maps for MACv2
 18 indirect forcing efficiency per unit anthropogenic AOD.



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 22 **Figure F1** monthly maps for environmental potential for aerosol indirect effects (left block) considering
 23 low cloud cover, reflection and susceptibility, surface albedo, sunshine hours and sun elevation and the
 24 indirect forcing efficiency (right block).

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 27 The highest environmental potential for indirect effects is at oceanic stratocumulus regions off
 28 western continental coasts in the subtropics and at mid-to-high latitude oceanic regions during spring
 29 and summer seasons with longer sunshine periods. The aerosol indirect forcing efficiency (indirect
 30 forcing per unit anthropogenic AOD) displays a very high sensitivity over the southern oceans, where
 31 anthropogenic contributions, however, are very low. As this high sensitivity is caused by the part of the
 32 logarithmic fit that is not well constrained by observations (see Appendix A) indirect effects are possibly

1 overestimated over aerosol sparse regions, although absolute contributions to the MACv2 indirect
 2 forcing from these very clean regions are relatively small.

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Table F1. assumed properties for the indirect environmental potential

Property	Assumptions
cloud top temperature weight wei	>270K full impact ($wei=1$) <250K no impact ($wei=0$)
solar cloud reflection based R_{cld}	$R_{cld} = (1 - \exp(-COT/(10 + 0.35 * COT)))$ COT-cloud optical depth
Susceptibility S_{cep}	$S_{cep} = R_{cld} * (1.0 - R_{cld})$
R_{cld} , max is 0.67	0.67
consider the background factor B_f	$B_f = 1.0 - surf.albedo$
low only cloud frequency F_{low}	F_{low}
scatter strength /sun-hours	$\sum \{time(u_0) * \{1.0 - \exp(-u_0 / u_{0max})\}^{1.7} * \exp(-0.025 / u_0^{1.5})\}$ with $u_{0max} = 0.25 + 0.75 * (1 - \exp(-0.22 * cot^{1.25}))$
environmental indirect potential	= $wei * S_{cep} * 0.67 * B_f * F_{low} * sun$

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9 Appendix G comparisons to other published aerosol direct radiative effects

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The present-day aerosol properties and associated direct radiative effects of MACv2 are here compared to complementary results from ‘bottom-up’ global modeling. More detail is provided by investigating component contributions in ‘bottom-up’ global modeling with assigned component data of the ‘top-down’ approach in MACv2.

First, mid-visible AOD and AAOD differences between a MACC reanalysis of assimilated MODIS AOD data in an ECMWF simulation (Bellouin et al., 2013) to MACv2 are investigated by comparing annual averages in Table G1.

Table G1. comparison of properties for present-day mid-visible (550nm) AOD and AAOD data between the MACv2 aerosol climatology and the MACC reanalysis data.

	AOD					AAOD			
	<i>total</i>	<i>DU</i>	<i>SS</i>	<i>fine</i>	<i>anthr</i>	<i>total</i>	<i>DU</i>	<i>fine</i>	<i>anthr</i>
MACv2	0.121	0.031	0.028	0.063	0.031	0.0072	0.0021	0.0051	0.0030
MACC	0.180	0.043	0.055	0.081	0.073	0.0080	0.0010	0.0070	0.0070

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With the MODIS data-assimilation in MACC (Bellouin et al., 2013) the global average AOD is 50% higher. Thus is a likely overestimate as updated MISR retrievals (M.Garay, personal communication) and even the ICAP satellite assimilation ensemble (Peng et al., 2013) indicate an upper ceiling of 0.14 for the present-day global mid-visible AOD. The (for anthropogenic impacts relevant) fine-mode AOD MACC has a smaller relative contribution of the total AOD but is still larger than in MACv2. More of a concern is

1 the large anthropogenic fraction of the fine-mode AOD in MACC, so that the present-day anthropogenic
 2 AOD is more than 2.5 times larger than in MACv2 - as all wildfires are incorrectly considered as
 3 anthropogenic in MACC. The MACC anthropogenic AOD has a slightly smaller absorption potential and
 4 the absorption potential of mineral dust is way too small, mainly due to a simplified dust size treatment
 5 in MACC. Differences in associated aerosol radiative effects are expected and shown in Table G2.
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8 **Table G2.** comparison of present-day aerosol associated aerosol radiative effects between the MACv2
 9 aerosol climatology and the MACC reanalysis data (in W/m²)
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	clear-sky TOA solar only direct radiative effect					direct total forcing	
	<i>total</i>	<i>DU</i>	<i>SS</i>	<i>fine</i>	<i>anthr</i>	<i>anthr</i>	<i>corrected</i>
MACv2	-3.5	-0.53	-0.91	-1.5	-0.70	-0.36	
MACC	-7.3	-1.6	-2.8	-2.8	-2.5	-0.70	-0.40

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 13 The MACC total aerosol present-day effect is too large, mainly because the total AOD is too
 14 large but also as the stronger cooling coarse-mode (missing mineral dust absorption/size) has a larger
 15 AOD fraction (too much seasalt).

16 The MACC aerosol present-day direct forcing of -0.7 W/m² is too large as it includes wildfire
 17 contributions (*N. Bellouin, personal communication*). When removing those contributions the aerosol
 18 forcing is corrected downward to an agreeable -0.4 W/m² cooling. Still, the contributing large reduction
 19 from clear-sky to an all-sky (from -2.5 to -0.7) in MACC raises proper cloud impact treatment questions.

20 Focusing on the fine-mode aerosol and its anthropogenic contributions the present-day aerosol
 21 direct forcing, along with component contributions of MACv2 was compared to ‘bottom-up’ ensemble
 22 averages of AeroCom (*Schulz et al., 2006*) and CMIP5 modeling (*Myhre et al., 2013*) in Table G3.
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25 **Table G3.** comparisons of present-day aerosol direct radiative forcing (TOA, all-sky) in W/m²
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	total	non-absorbing		absorbing			
		<i>SU</i>	<i>NI</i>	<i>CA</i>	<i>BC</i>	<i>OC</i>	<i>SOA</i>
MACv2	-0.36 (<i>to -0.20</i>)	-0.41		+0.05 (<i>to +0.22</i>)	+0.28 (<i>to +0.44</i>)	-0.23	
Myhre avg	-0.32	-0.32	-0.08	+0.09	+0.18	-0.03	-0.06
Myhre med	-0.28	-0.32	-0.08	+0.13	+0.18	-0.03	-0.02
Schulz	-0.22	-0.32		+0.11	+0.25	-0.14	

27 *MACv2 values with higher than fine-mode present-day anthropogenic BC fractions are in parenthesis*
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30 There is surprising good agreement for direct forcing estimates, even on a component basis. It is
 31 also shown, that if the prescribed fine-mode anthropogenic fraction in MACv2 is raised for soot (BC),
 32 then extra BC warming shifts the overall aerosol direct forcing to less negative values in MACv2. Thus,
 33 the possibility of a lower MACv2 direct effect cannot be ruled out. Here again, it is the limited
 34 understanding of the pre-industrial reference that introduces uncertainty.