Increasing Aerosol Direct Effect Despite Declining Global Emissions

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Abstract

Anthropogenic aerosol particles partially mask global warming driven by greenhouse gases, both by directly reflecting sunlight back to space and indirectly by increasing cloud reflectivity. In recent decades, the emissions of anthropogenic aerosols have declined globally, and at the same time shifted from the North American and European regions to foremost Southeast Asia. Using simulations with the MPI-ESM1.2 global climate model we find that the direct effect of aerosols has continued to increase, despite declining emissions. Concurrently, the indirect effect has diminished in approximate proportion to emissions. The enhanced efficiency of aerosol radiative forcing to emissions is associated with less cloud masking, longer atmospheric residence time, and differences in aerosol optical properties.









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Key Points: 6

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7	• Despite declining global aerosol emissions, the direct effect of anthropogenic aerosols
8	is found to increase in a global climate model.
9	• This is caused by the regional shift of emissions combined with variations of emissions-
10	to-forcing ratios.
11	• Regional variations of the aerosol direct effect are associated with cloud masking,
12	aerosol removal processes and aerosol optical properties.

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13 Abstract

Anthropogenic aerosol particles partially mask global warming driven by greenhouse gases, 14 both by directly reflecting sunlight back to space and indirectly by increasing cloud re-15 flectivity. In recent decades, the emissions of anthropogenic aerosols have declined glob-16 ally, and at the same time shifted from the North American and European regions to 17 foremost Southeast Asia. Using simulations with the MPI-ESM1.2 global climate model 18 we find that the direct effect of aerosols has continued to increase, despite declining emis-19 sions. Concurrently, the indirect effect has diminished in approximate proportion to emis-20 sions. The enhanced efficiency of aerosol radiative forcing to emissions is associated with 21 less cloud masking, longer atmospheric residence time, and differences in aerosol opti-22 cal properties. 23

²⁴ Plain Language Summary

Aerosol particles in the atmosphere from natural sources and human activities have 25 a cooling effect on the climate, and therefore partially offset global warming caused by 26 greenhouse gases. They achieve this through both direct and indirect mechanisms. Di-27 rectly, aerosols cool by reflecting incoming sunlight, while indirectly they alter cloud prop-28 erties, which is thought to increase cloud reflectivity. In this study, we investigate the 29 historical evolution of these aerosol effects using a global climate model. We find that 30 despite the global decline of emissions over recent decades, the direct effect of human-31 made aerosols on the climate has increased, while the indirect effect has decreased. We 32 associate the increase in direct effect with the shift in aerosol emissions from North Amer-33 ica and Europe to primarily Southeast Asia. 34

35 1 Introduction

The state of Earth's climate is determined by the delicate balance between the incoming solar energy and the energy the Earth reflects and radiates back to space. Greenhouse gases, such as CO_2 , act to warm the Earth by reducing the radiation emitted to space (radiative forcing), resulting in an accumulation of energy in the climate system and, consequently, an increase in the surface temperature, T_s . A linear radiative balance framework can be used to study the response in temperature, ΔT_s , to an applied radiative forcing, F, on the net energy balance at the top of the atmosphere, N:

$$N = F + \lambda \Delta T_s,\tag{1}$$

where λ is the total feedback parameter of the system, which must be negative to yield 43 a stable climate. The feedback parameter can be itself divided into the sum of the in-44 dividual climate feedbacks, such as water vapour, surface albedo, cloud and temperature 45 feedbacks. They contribute to enhance or dampen the imbalance induced by an applied 46 forcing. Knowledge of how anthropogenic activity affects the radiative balance is cru-47 cial for understanding how the climate may change in the future, and currently the largest 48 contributor of uncertainty to estimates of the total anthropogenic forcing is aerosols (Forster 49 et al., 2021). 50

Aerosols are small particles, emitted by natural and human sources, which affect the radiative balance of the climate system through direct and indirect mechanisms. Directly, they interact with solar radiation by scattering and reflecting it back to space or absorbing it (direct effect). Indirectly, they interact with clouds, for example through the Twomey effect (Twomey, 1974) by increasing the number density of droplets in clouds, making them more reflective to solar radiation (indirect effect).

⁵⁷ Due to their short lifetime in the atmosphere, aerosols are heterogeneously distributed, ⁵⁸ spatially and temporally. The heterogeneity, together with interactions between aerosols ⁵⁹ and clouds, makes the aerosol forcing difficult to constrain, and thus adds uncertainty to estimates of the total radiative imbalance of the climate system. This uncertainty poses challenges in assessing how aerosols may mask the effects of global warming caused by

⁶² greenhouse gases and the trajectory of future global warming.

It is therefore a community priority to narrow down the uncertainty in aerosol forcing (Bellouin et al., 2020). Multiple approaches to this end have been taken, including process understanding usually in the form of global models (e.g., Fiedler et al., 2023), satellite observational constraints based on internal variability or volcanic eruptions (e.g., Gryspeerdt et al., 2017; McCoy et al., 2017; Malavelle et al., 2017), or top-down approaches based on the observed global warming (Stevens, 2015; Kretzschmar et al., 2017; Booth et al., 2018). In particular for top-down constraints it is essential to relate historical evolution of aerosol emissions to radiative forcing.

Therefore, in the current study we separate the historical evolution of the direct
and indirect aerosol forcing in a climate model, and relate these to global aerosol emissions. The effect of the regional redistribution in recent decades is investigated, and the
underlying mechanisms elucidated.

$_{75}$ 2 Method

We study the historical evolution of the aerosol forcing using the state-of-the-art global climate model Max Planck Institute for Meteorology Earth System Model version 1.2. MPI-ESM1.2 (Mauritsen et al., 2019). MPI-ESM1.2 participated in the Coupled Model Intercomparison Project Phase 6 (CMIP6) and successfully reproduces the observed warming from pre-industrial levels (Mauritsen & Roeckner, 2020). In this study we use the coarse resolution (CR) version of the model to simulate the historical period from 1850 to present day, with input as in the CMIP6 historical scenario.

The radiative transfer scheme of MPI-ESM1.2 uses the Simple Plumes implemen-83 tation of the second version of the Max Planck Institute Aerosol Climatology (MACv2-84 SP) to represent the aerosol impact on the radiation (Stevens et al., 2017). MACv2-SP 85 provides a parameterization of optical properties of anthropogenic aerosols and the re-86 sulting Twomey effect (Twomey, 1974; Stevens et al., 2017). It has been designed to pro-87 vide a uniform and easily controlled representation of anthropogenic aerosol perturba-88 tions for the CMIP6 framework (Eyring et al., 2016; Pincus et al., 2016). In the model, 89 aerosol emissions are represented by nine spatial plumes that are associated with emis-90 sions from major anthropogenic source regions (Stevens et al., 2017). The prescribed aerosol 91 optical properties are based on ground-based measurements provided by the Max Planck 92 Institute Aerosol Climatology, MAC (Kinne et al., 2013), for the present-day (2005) dis-93 tribution of mid-visible anthropogenic aerosol optical depth (AOD). 94

MACv2-SP represents changes from pre-industrial (1850) to 2016, by scaling the 95 present-day distribution of aerosols using historical emission of SO_2 and NH_3 , based on 96 data obtained from the Community Emissions Data System (CEDS). Two types of emis-97 sions are considered: industrial emissions for the plumes in Europe, North America, Aus-98 tralia, East and South Asia and biomass burning emissions for the plumes in South Amer-99 ica, the Maritime Continent, North and South Central Africa. These two types of emis-100 sions differ in their seasonal cycle amplitude, single-scattering albedo, and the strength 101 of the Twomey effect. 102

To model the Twomey effect, Stevens et al. (2017) used satellite observations to derive a relationship between the cloud droplet number density and the fine-mode AOD. With this representation, anthropogenic aerosols cause a greater increase in cloud optical thickness when the atmospheric environment is initially pristine in terms of aerosols. For a complete description of MACv2-SP, refer to Stevens et al. (2017). In the literature, various metrics have been proposed to assess the impact of a radiative perturbation on Earth's energy balance. When a perturbation is introduced into the climate system, rapid adjustments occur due to rapid stratospheric temperature change. Radiative forcing (RF) is used to quantify the radiative imbalance resulting from an applied perturbation, taking these rapid adjustments into account. The effective radiative forcing (ERF) includes further adjustments of the system, accounting for all tropospheric and land surface adjustments.

We use the Partial Radiative Perturbation (PRP) method to calculate the radia-115 tive forcing from aerosols in the MPI-ESM1.2 simulations. In this method the radiative 116 effect of a change in a certain state variable, for example the effect of a change in sur-117 face albedo between a control and perturbed climate run, is calculated as the difference 118 between two radiation calls. The method was first described by Wetherald and Manabe 119 (1988) and Colman and McAvaney (1997), and implemented in MPI-ESM by Meraner 120 et al. (2013) and Block and Mauritsen (2013) as a two-sided version, as described by Klocke 121 et al. (2013). Another version of the PRP diagnostic has also been used by Mülmenstädt 122 et al. (2019) to assess the radiative effects of aerosols in a different version of the atmo-123 spheric model component of MPI-ESM1.2. 124

The PRP method was designed to evaluate the respective contributions of individ-125 ual forcings and feedbacks on the radiative imbalance (Eq. 1). Here, we integrated the 126 anthropogenic aerosol perturbation provided by MACv2-SP into the PRP module of MPI-127 ESM1.2. This enables us to estimate the instantaneous radiative forcing from anthro-128 pogenic aerosols independently of climate feedbacks and atmospheric adjustments. Fur-129 thermore, as MACv2-SP provides two distinct prescribed perturbations for the direct 130 and indirect effects of aerosols, we can substitute them one at a time into the PRP method 131 to evaluate their respective contributions. This approach allows us to conduct regular 132 historical simulations in MPI-ESM1.2 and investigate the past evolution of anthropogenic 133 aerosol effects on the climate system. 134

135 **3 Results**

In the following section we present the simulated anthropogenic aerosol forcing throughout the historical period. We separate the contributions from the direct and indirect effect and show how the forcing varies depending on the location of the aerosol emissions. Finally, we investigate the mechanisms governing regional differences in the relationship between aerosol emissions and radiative forcing.

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3.1 Historical Aerosol Forcing

In the historical simulation, the PRP diagnostic reveals an increasingly negative forcing from aerosols throughout the century, despite the global reduction in aerosol emissions in recent decades, as shown in Figure 1. The persistently negative trend in the total aerosol forcing is primarily driven by the direct effect, which continues to increase even after the implementation of air quality regulations in Europe and North America since the 1970s and 80s. Meanwhile, the indirect effect is reduced in approximate proportion to the decreasing emissions.

In addition to the global decrease in aerosol emissions, the period spanning from the 1970s to 2005 witnessed a shift in aerosol emission patterns. Early in the historical period global aerosol emissions were dominated by emissions from Europe and North America. In recent decades, South and East Asia have become the dominant source regions. The subsequent sections investigate the role played by this geographical shift in explaining the observed inconsistency between global aerosol emissions and forcing.



Figure 1. Historical forcing from anthropogenic aerosols. The top part shows the historical global emissions of aerosols and the bottom part shows the induced radiative forcing in MPI-ESM1.2. Values are global yearly means. Vertical lines indicate the years 1972 and 2005 when emission levels were similar but led to different total aerosol forcing.

3.2 Forcing from Regional Aerosol Sources

As detailed in Section 2, MACv2-SP provides a parameterization for anthropogenic 156 aerosols, incorporating nine distinct plumes that represent various anthropogenic emis-157 sion regions. To assess the aerosol forcing from each of these regions throughout the his-158 torical period, we substituted one plume at a time into the PRP calculation. Figure 2 159 shows the resulting forcing values from each region against the corresponding aerosol emis-160 sions in Tg of SO_2 equivalent. Regressing the induced forcing against the associated emis-161 sion level, we obtain a value of the regional aerosol efficiency in Wm^{-2} per Tg of SO_2 162 equivalent. 163

In Figure 2a, we observe a significant variability in efficiency of the direct effect among major industrial regions, such as Europe, North America, and East and South Asia. Notably, South Asia exhibits an efficiency 20.10 times greater than Europe, representing the most substantial difference in efficiency across these regions. On the other hand, Figure 2b shows a relatively more consistent relationship between forcing and emissions across regions for the indirect effect.

The variation in regional aerosol efficiency explains the persistent increase in the 170 global direct effect despite decreasing emissions. Regions with higher efficiencies have 171 a more substantial impact on the global direct effect despite emitting fewer aerosols. This 172 effect becomes particularly important when considering the shift in aerosol patterns from 173 1980 to 2005. During this period, aerosol emissions shifted from Europe and North Amer-174 ica to South and East Asia, where the efficiency is higher. Consequently, despite reduced 175 global emissions during this period, the global aerosol forcing continued to increase. Sub-176 sequent sections delve into the mechanisms that underlie these regional variations in ef-177 ficiency. 178



a) Aerosol Direct Effect

b) Aerosol Indirect Effect

Figure 2. Aerosol forcing from individual emission regions against regional emission levels. The values are global yearly means for each year from 1850 to 2013 separated into the direct (a) and indirect (b) effect. Panel c) shows the clear-sky aerosol effect. The numbers annotated in panels a) and c) are the efficiencies in Wm^{-2} per Tg of SO₂ equivalent for the major emission regions, based on linear regression.

3.3 All-sky and Clear-sky Aerosol Forcing

We examine the outcomes of the PRP performed under both all-sky and clear-sky conditions. The results show that under all-sky conditions the direct effect primarily causes a radiative forcing in the vicinity of emission sources (see Figure 3c). In contrast, the indirect effect is more pronounced over remote regions (see Figure 3.b.) and is larger than the direct effect on global average. These results are in line with Huusko et al. (2022), who used a different method for estimating the spatial patterns of the aerosol direct and indirect effects in MPI-ESM1.2.

The clear-sky global aerosol forcing surpasses the all-sky global total aerosol forc-187 ing (including direct and indirect effects, see Figure 1). It is essential to note that un-188 der clear-sky conditions only the direct effect applies. Interestingly, the clear-sky aerosol 189 forcing is more than twice as large as the direct effect observed in all-sky conditions. This 190 191 pattern remains consistent across all emission regions, with clear-sky aerosol forcing consistently exceeding the all-sky direct effect (see Figure 2a and c). Under all-sky condi-192 tions, the presence of extensive cloud cover locally results in positive forcing from the 193 direct effect of aerosols (see Figure 3c and e), as the presence of clouds moderates the 194 net effect of aerosol scattering while amplifying the net effect of aerosol absorption (Li 195 et al., 2022; Bellouin et al., 2020). With single-scattering albedo of 0.93 and 0.87 for in-196 dustrial and biomass burning emissions, respectively, in the model (Stevens et al., 2017), 197 absorption prevails in the presence of clouds, resulting in positive direct effect of aerosols. 198

In addition, in regions with persistent cloud systems, the negative forcing from the 199 indirect effect and the positive direct effect tend to balance each other (see Figure 3a and 200 e). This mechanism has significant implications for regional emission efficiency. In par-201 ticular, it explains why Europe, which exhibits a weak efficiency under all-sky conditions (Figure 2a) due to a positive direct effect at high latitudes (Figure 3), demonstrates greater 203 efficiency under clear-sky conditions (Figure 2c). Looking at clear-sky conditions signif-204 icantly narrows the gap in regional efficiencies. In South Asia, the efficiency in clear-sky 205 conditions is only 2.1 times greater than in Europe, which is significantly lower than the 206 factor 20.1 difference observed in all-sky direct effect. The most pronounced difference 207 under clear-sky conditions is seen between South Asia and North America, with South 208 Asia showing a efficiency 3.2 times greater than North America. 209

The effect of cloud cover on the direct effect of anthropogenic aerosols emerges as 210 the main factor influencing regional efficiency. This largely explains the consistent in-211 crease in negative aerosol forcing despite reduced emissions in the last decades. As there 212 is less of a persistent cloud cover at the emission sources in South and East Asian regions 213 compared to Europe, the shift in aerosol patterns results in an enhanced global direct 214 effect. The indirect effect instead induces a forcing in remote regions downstream of emis-215 sions. The strength of this effect globally follows the global emissions despite the shift 216 in the emission pattern. The increase in aerosol efficiency resulting from the shift in aerosol 217 spatial pattern is critical to explaining the increase in aerosol forcing between the mid-218 1970s and the mid-2000s, despite similar emissions levels. However, the disparity in emis-219 sion efficiencies between regions remains substantial even under clear-sky conditions. The 220 following sections investigate other factors contributing to these regional differences. 221

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3.4 MACv2-SP Aerosol Representation and Regional Variation

The MACv2-SP has been designed to simplify the representation of anthropogenic aerosols in climate models through a straightforward parameterization. It provides monthly mean Aerosol Optical Depth (AOD) values based on ground-based measurements, representing the 2005 spatial distribution. To get the spatial pattern in other years the 2005 pattern is scaled with estimates of historical emissions (Stevens et al., 2017). The measured AOD values are influenced by the rate of aerosol removal at a given location, so even though removal processes are not explicitly represented in MACv2-SP the AOD in



Figure 3. Present-day (2005) spatial pattern (yearly mean) of a) the total aerosol forcing, separated into b) the indirect effect and c) the direct effect; d) the clear-sky aerosol effect; e) the cloud cover fraction. Panel f) shows the Column Aerosol Optical Depth at 550nm from the MACv2-SP parameterization, with dashed-line showing low AOD value contour (0.0025). Values annotated on the maps are global means.

the model may not always be directly proportional to regional emissions. For example,
wet deposition is the dominant sink of sulfate aerosols from industrial sources (Textor
et al., 2006), and this deposition mechanism is particularly prominent over the eastern
coasts of North America and East Asia (Rodhe et al., 2002), causing a relatively small
AOD per unit of emissions there.

Figure 4a shows the clear-sky aerosol forcing against the corresponding AOD for 235 each region. This representation noticeably reduces the difference in aerosol efficiency 236 between regions compared to Figure 2c, indicating that AOD is a better predictor of aerosol 237 forcing than emissions. For instance, the efficiency of South Asia is 1.3 greater than that 238 of Europe when measured in Wm^{-2} per unit of optical depth, whereas it was 2.1 greater 239 when measured in Wm^{-2} per unit of emissions (in Tg of SO₂-eq). Interestingly, when 240 considering AOD levels, both Asian regions exhibit similar efficiencies, which is not the 241 case when considering emissions. Strong wet deposition in East Asia contributes to the 242 removal of aerosols (Rodhe et al., 2002), resulting in a lower AOD per unit of emissions 243 and thus a weaker direct effect in this region. Conversely, South Asia exhibits weaker 244 wet deposition (Rodhe et al., 2002), allowing for greater forcing per unit of emissions. 245

This difference in aerosol removal patterns is the second most important explanation for the continued increase in aerosol forcing despite reduced emissions. The shift in aerosol patterns from Europe and North America towards Southeast Asian regions, with weaker wet deposition, prolongs the residence time of aerosols in the atmosphere, consequently enhancing the aerosol efficiency. In the last section, we suggest additional explanations for the remaining minor differences in aerosol efficiency between regions.

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3.5 Aerosol Single-Scattering and Surface Albedo

The distinction between industrial and biomass aerosol emissions affects primar-253 ily the Single-Scattering Albedo (SSA) parameter provided by MACv2-SP, which is 0.93 254 for industrial and 0.87 for biomass burning (Stevens et al., 2017). The greater shortwave 255 absorption by biomass burning aerosols results in weaker efficiencies when compared to 256 industrial regions. Figure 4b is similar to Figure 4a but with data from a new simula-257 tion where the SSA was set to the same value (0.93) for all sources, substantially reduc-258 ing the spread for the biomass burning source regions. It is important to note that this 259 holds true in all-sky conditions as well, as SSA defines the ratio of scattering efficiency 260 to total extinction efficiency. However, it plays a relatively minor role in the overall dis-261 crepancy, given that biomass burning regions have relatively smaller emissions and forc-262 ing compared to industrial sources. 263

Some spread still remains between the regions, suggesting that some other mechanisms also influence the efficiency. For example, the influence of anthropogenic aerosols on the radiative balance depends on the nature of the underlying surface (Li et al., 2022), suggesting that the remaining differences in clear-sky efficiency among emission regions may be partly associated with differences in surface albedo between the regions.

²⁶⁹ 4 Conclusions

We have investigated the relationship between aerosol emissions and radiative forc-270 ing in the global climate model MPI-ESM1.2. Our results reveal an increase in global 271 mean aerosol radiative forcing in recent decades, despite a global reduction in aerosol 272 emissions. This increase is driven primarily by the direct effect, while the indirect effect 273 remains more consistent with emission levels. The increase in the direct effect is asso-274 ciated with regional shifts in emissions. Historically, Europe and North America have 275 been the primary sources of aerosol emissions, but since the 1970s emissions have shifted 276 to South and East Asia, where the radiative forcing per unit of emissions is larger. 277



a) Clear-sky Aerosol Forcing

Figure 4. Aerosol forcing from individual emissions regions against regional column aerosol optical depth. Plots are global yearly means for each year from 1850 to 2013 with panel a) showing the clear-sky aerosol effect. Panel b) shows the same but in a new experiment in which the single-scattering albedo was set to the same value for all sources. Values annotated on the plots are the efficiencies in Wm^{-2} per unit of optical depth of the major emission regions, based on linear regression.

The primary mechanism driving the disparity between global aerosol emissions and radiative forcing is the masking effect of clouds. Mid- to high-latitude regions, characterised by substantial cloud cover, exhibit enhanced aerosol absorption relative to scattering resulting in a weaker negative or even positive direct effect. The recent shift of aerosol emissions to South and East Asia, where the cloud cover is less extensive, has led to a more negative global mean aerosol direct effect.

Other significant contributors include the regional variation in aerosol residence time 284 within the atmosphere. Atmospheric conditions in South Asia with weaker removal pro-285 cesses, such as wet deposition, as compared to North America, allow aerosol particles to 286 stay in the atmosphere longer, causing a greater efficiency in terms of radiative forcing 287 per unit of emissions. Furthermore, the optical properties of the aerosol particles them-288 selves can influence the forcing efficiency per unit of emissions. In recent decades, emerg-289 ing biomas s burning source regions have led to the emission of greater quantities of ab-290 sorbing aerosols, which act to dampen the negative direct effect. Nevertheless, since the 291 biomass burning emissions are relatively small compared to those from industrial sources, 292 this has not offset the global negative increase in the direct effect. 293

When compared to other global climate models, MPI-ESM1.2 has a relatively weak 294 total aerosol forcing (Mauritsen et al., 2019), and Fiedler et al. (2023) have found that 295 the range of aerosol forcing among CMIP6 models is primarily influenced by the strength 296 of the indirect effect. Our findings suggest that models with a weaker indirect effect rel-297 ative to the direct effect may exhibit a radiative forcing less consistent with global emis-298 sions, while models with a strong indirect effect are likely to have greater consistency 299 between their aerosol forcing and emissions. This can help explaining the variety in the 300 301 evolution of the aerosol effective radiative forcing throughout the historical period in CMIP6 observed by Fiedler et al. (2023). 302

303 Open Research Section

The source code for MPI-ESM1.2 can be accessed via https://mpimet.mpg.de/ en/science/models/mpi-esm (Mauritsen et al., 2019). Additionally, the specific parts of the code that were modified and developed for this study, as well as the model outputs and Python scripts used in producing the figures presented in this paper, are accessible through Zenodo at https://doi.org/10.5281/zenodo.10161509 (Hermant et al., 2023).

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Figure 1.



Figure 2.

a) Aerosol Direct Effect

b) Aerosol Indirect Effect





c) Clear-sky Aerosol Effect



- Europe
- North America
- East Asia
- South Asia
- North Africa (biomass)
- South America (biomass)
- Maritime Continent (biomass)
- South Central Africa (biomass)
- Australia

Figure 3.



Figure 4.

a) Clear-sky Aerosol Forcing

