

Reply to reviewer 1

Thank you for your valuable comments. This study indeed aims to explore the potential decoupling between aerosol emissions and their direct effects. We acknowledge your concerns regarding the modelling approach employed to estimate historical aerosol forcing in MPI-ESM1.2 through the use of the MACv2-SP parametrization. In this response, we aim to provide clarity on our methodology by providing a description of the MACv2-SP parametrisation and specific results from our study.

1. MACv2-SP combines ground-based measurements of a 2005 aerosol climatology with emission estimates to represent historical changes in aerosol direct and indirect effects. Emissions estimates of SO₂, NH₃ as well as BC (CEDS) are accounted for in the scaling of the 2005 climatology (See Stevens et al. [2017]: Section 4: 'Time-varying forcing', Table 5,6, Figure 9,10). By weighting the radiative properties of these three compounds with their respective emission estimates, the changes in aerosol properties in time and space are represented. We acknowledge the limitation of this representation, which only takes into account these three (major) species and does not include interactive processes with the atmosphere. Nevertheless, Stevens [2015] argues that this representation of emissions successfully capture the main features seen in more complex models, both in terms of global signal and regional patterns. We intend to clarify and elaborate the representation of the time-varying forcing in the manuscript. While aerosol removal processes are not explicitly represented in MPI-ESM1.2 with MACv2-SP, they are recorded in the in-situ measurements and thus included implicitly in the MACv2-SP representation, and we argue that this provides a sufficient representation of aerosol forcing in the context of this study.

While we acknowledge your concern regarding the time-variation of aerosol forcing, we intend to provide specific results for the reference year of 2005, since it directly applies the measured aerosol climatology. Utilizing the MACv2-SP parametrisation and our PRP approach, we calculate instantaneous aerosol radiative forcing and derive annual means. Notably, in 2005, European, North American, and South Asian sources exhibit similar emission levels (refer to Table 6 in Stevens et al. [2017]), yet significant differences in direct effect efficiency are observed (as shown in Response Table 1). By normalizing the forcing by the respective AOD values (accounting for implicit regional processes), we observe a reduction in the regional efficiency spread. Furthermore, when analysing clear-sky aerosol forcing while accounting for regional cloud-masking effects, the spread is reduced further.

Through our analysis of 2005 forcing and investigation of regional forcing disparities, we infer that a shift in emission patterns could potentially lead to a decoupling between global emissions and direct effect. Our study demonstrates this by the use of parametrised aerosol forcing and PRP approach to effectively distinguish between direct and indirect effects within MPI-ESM1.2. We note that a decoupling between direct effect and emissions does not necessarily imply a strong decoupling in total aerosol forcing, as the indirect effect is usually dominant in ESM [Fiedler et al., 2023] and more consistent with emissions (see Figure 1 and 2b). Such decomposition between direct and indirect effects is not as straightforward in models that use interactive aerosol modules. This complicates a direct comparison with models that explicitly represent aerosol processes. Acknowledging the limitations of our methodology, we constrain the focus of our study to radiative transfer processes in ESM and show that MACv2-SP is a valuable tool in this context.

2. We would like to address the comparison of the present-day aerosol direct effect magnitude

Source region	Emissions	AOD	ADE	ADE/E	ADE/AOD
Europe	16.41	2.79	-0.012	-0.72	-4.22
North America	17.45	1.16	-0.025	-1.46	-22.0
East Asia	37.36	4.26	-0.086	-2.30	-20.18
South Asia	17.17	4.74	-0.152	-8.85	-32.06
North Asia	1.70	0.55	-0.005	-3.09	-9.54
North Africa	4.88	0.24	-0.003	-0.63	-12.64
South America	4.15	0.45	-0.012	-2.83	-26.07
Maritime Continent	3.35	1.38	-0.003	-0.80	-1.94
Australia	1.57	0.56	-0.026	-16.73	-47.01

Response Table 1: Aerosol direct effect efficiencies per source region in 2005. Emissions are Equivalent SO₂ in Tg SO₂, AOD [10^{-3}], Aerosol Direct Effect (ADE) in [Wm^{-2}], ADE/E in [10^{-3}Wm^{-2}] per emission unit, ADE/AOD in [Wm^{-2}] per AOD unit

to the CMIP6 model mean. Referring to AR6, chapter 7, Table 7.6 [Forster et al., 2021], the Direct Effect CMIP6 average and 5-95% confidence range is $-0.25 \pm 0.40 \text{Wm}^{-2}$, Bellouin et al. [2020] reports a present day Direct Effect ranging from -0.37 to -0.12 Wm^{-2} , whereas our study reports -0.324 Wm^{-2} (Figure 3.c). For the total aerosol radiative forcing, AR6 reports the CMIP6 average and 5-95% confidence range of $-1.11 \pm 0.38 \text{Wm}^{-2}$, Bellouin et al. [2020] report a present day total aerosol radiative forcing of -2.0 to 0.4 Wm^{-2} with a 90% likelihood, whereas our calculations stands at -0.76 Wm^{-2} (Figure 3.a). Other recent studies of aerosols radiative effects in CMIP6, such as Fiedler et al. [2023] and Smith et al. [2020], report a present day aerosol Effective Radiative Forcing ranging from -1.47 to -0.59 Wm^{-2} and from -1.37 to -0.63 Wm^{-2} respectively. Results from our PRP calculations align with other studies using different methods (e.g. Mauritsen et al. [2019], Fiedler et al. [2017]). Specifically, our results fall within the ranges cited above, particularly those from the AR6 assessment and Bellouin et al. [2020] study. This indicates that the magnitude of the present-day aerosol direct radiative forcing estimated in our study is not larger than the CMIP model mean and is consistent with the assessment provided in the AR6 report. It is important to clarify that MACv2-SP considers SO₂, NH₃, and BC as precursors. All emissions are presented in SO₂ equivalent units, accounting for the respective contributions of these precursors. We acknowledge the need for clearer explanations in both the manuscript and figure captions about this.

3. We acknowledge your confusion regarding the discussion on SSA and biomass aerosols. While we extensively reference Stevens et al. [2017], we recognize the need for clearer information on the design of MACv2-SP plumes. We thus intend to clarify the effect of SSA in the manuscript as follow. Each plume represents anthropogenic aerosol emissions, accounting for SO₂, NH₃ and BC. The industrial plumes (Europe, North America, East and South Asia) emphasise the industrial aerosols, while the biomass plumes (North and South Central Africa, South America, Maritime Continental and Australia) emphasise aerosols resulting from anthropogenic biomass burning. This distinction is essential to reflect the dominance of specific aerosol species in each plume type. SSA values are uniformly set for both types of plumes based on measurements, as validated by Stevens [2015] to effectively capture temporal trends and regional patterns when compared to models integrating more complex aerosol processes.
4. The aerosol forcing in MPI-ESM1.2 falls within the mid CMIP6 range. In the paragraph starting on line 200 we refer to other studies that did extensive intercomparison between

CMIP6 models (such as Fiedler et al. [2023]); this type of comparison is outside the scope of the present study. As mentioned in Section 2, MPI-ESM1.2 successfully represents the historical surface temperature evolution, in particular it closely matches temperature records during the period 1950-1980 ("dampened warming" period), where the wider spread in CMIP6 models is observed [Mauritsen and Roeckner, 2020, Flynn and Mauritsen, 2020]. This makes us believe that MACv2-SP representation of aerosols is reasonable.

In summary, here is what we intend to include in the revised version of the manuscript:

- Scatter points on Figure 2 and 4 to highlight 2005 values (as presented here in Response Table 1), as well as more details on the temporal consistency of the plume efficiencies.
- A more exhaustive description of the MACv2-SP aerosol representation in the method Section, especially summarizing Section 4 on Time-varying forcing in Stevens et al. [2017].
- Clearer description of the effect of SSA and of the SO₂-equivalent emission unit.

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