Reply to reviewer 2

Thank you for your insightful comments on our manuscript. We appreciate your attention to detail and would like to address the concerns you raised regarding the radiative forcing efficiency within the MACv2-SP aerosol prescription. We acknowledge the critical role of parametrisation choices in determining aerosol radiative forcing as well as the limitations of this method, and understand your point about the inherent influence of these choices. It is important to note that MACv2-SP was specifically developed to capture aerosol radiative effects on climate using ground-based and satellite measurements as well as emission inventories of key precursors such as SO2, NH3, and BC. In addition, MACv2-SP has been designed to be lightweight and easy to use in different climate models in the CMIP6 framework. Furthermore, Stevens [2015] argues that this representation successfully replicates key features observed in more complex models, both globally and regionally. To address your comments effectively, we will provide additional details on the construction of MACv2-SP to offer further clarity on our analysis. We also recognize the importance of a thorough understanding of Stevens et al. [2017] for contextualizing our work, particularly Section 4: "Time-varying forcing".

Answers to main comments

1. We understand your concern about the strength of the South Asian plume. MACv2-SP uses CEDS emission inventories for scaling the forcing, including SO2, NH3 and BC for all plumes. Both reflective and absorbing aerosols are considered. MACv2-SP has been designed to fit climatological values of aerosol properties at the year 2005. "High-quality data by ground-based sun-photometer" are merged onto "global model background maps from AeroCom" [Stevens et al., 2017]. Different aerosol properties are considered: AOD at 440, 550 and 870 nm, absorbing AOD at 550 nm and coarse and fine-mode aerosol particles [Stevens et al., 2017]. Using this aerosol climatology retrieved from direct measurements, our calculation indicate a strong direct effect efficiency from South Asia, both against emissions and AOD in 2005 (see Response Table 1). Since that the year 2005 applies aerosol climatology from measurements and AeroCom, we are uncertain why South Asian aerosols would be expected to be strongly absorbing.

   Furthermore, there must have been a misunderstanding about Section 3.5. We do suggest that SSA significantly drives direct effect forcing efficiency (line 174-175: "a new simulation where the SSA was set to the same value for all sources, substantially reducing the spread"). SSA does have an influence on the forcing, which explains the difference between plumes dominated by industrial aerosols and plumes dominated by biomass burning aerosols. But we emphasize that since the recent increases in biomass burning in the Southern Hemisphere have a minor contribution to the total aerosol direct effect, they have a minimal impact on the global forcing-to-emission decoupling.

2. Emission rates are used to scale the 2005 aerosol climatology derived from measurements and AerCom [Stevens et al., 2017]. Analysis of the year 2005, where the measured climatology is applied (without any scaling), already suggests a strong regional dependence of the direct effect (see Response Table 1. Figure 2.c suggests that the difference between regions is in large part explain by cloud masking. Furthermore, Figure 4.a shows that the spread is reduced when considering AOD instead of emissions. To explain this, we do speculate that some aerosol processes are implicitly recorded in the instrumental measurements, such as aerosol removal. We do not completely understand your concern about the influence of residence times on radiative forcing, since we observe this discrepancy between emissions and AOD in 2005. Longer residence time results in greater measured AOD per unit of emission, resulting in greater forcing efficiency per unit of emission. Since the forcing is scaled with emissions inventories, the discrepancy is spread over time. Response Table 1 highlights the emissions and forcing values for each plume in the year 2005. The efficiencies in the Table are directly calculated from 2005 for comparison with the ones calculated in the manuscript figures. While in the manuscript we calculated the efficiencies via linear regression both against emissions and AODs, we acknowledge that we should more explicitly state that these efficiencies are consistent with emissions and thus time. We intent to clarify this in the manuscript.

Other comments

1. Lines 21-23: Both direct and indirect effect and aerosol-radiation (ari) and aerosol-cloud interactions (aci) appear in the literature, and we find that the direct and indirect effect terminology are more intuitive and connect better with the framework of radiative forcing as an instantaneous effect on the radiative balance and subsequent interactions and adjustments within the climate system.

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

2. Line 29: We appreciate that you highlight the relevance of Quaas et al. [2022]. Section 5 of their work emphasizes the close relationship between clear-sky solar ERFaer trends and trends in sulfate precursors, noting significant declines in major source regions from North America, Europe, and East Asia, alongside increases in India and surrounding regions. These findings align with our results across the 4 major industrial plumes and address your concerns regarding the strength of South Asia and the dominance of sulfate in MACv2-SP. Notably, their study supports our results by indicating that trends in sulfate precursors are driving aerosol forcing increases in regions like 'India and surrounding areas' (referred to as South Asia in MACv2-SP) over recent decades. We intend to include this comparison into the manuscript in Section 3.2.

3. Line 73: We systematically employ the two-sided PRP method described in Klocke et al. [2013].To keep our text concise, we opted not to delve into a detailed explanation of the PRP methodology in this paper. The original forward PRP (Wetherald and Manabe [1988]) consists in sequentially substituting specific state variable fields from the current climate state into a reference state, while keeping all other variables constant at the reference level. This partial perturbation approach allows for assessing each variable's contribution to the total radiative forcing. However, the perturbation is sensitive to the state in which it is introduced [Colman and McAvaney, 1997] and de-correlating fields when substituting them introduces unintended perturbations [Klocke et al., 2013]. The proposed method to partially address these approximations is to apply the partial radiative perturbation forward and backward. The backward perturbation consists in substituting the variable fields from the reference state into the current state. By averaging the results of forward and backward computations (two-sided approach), a more accurate estimation of forcing is achieved. We judge that a detailed description of this methodology would be too technical for the scope of this paper, thus we refer the reader to Klocke et al. [2013] and Colman and McAvaney [1997] for a more comprehensive description.

4. Line 103: As mentioned earlier, emissions in Tg of SO2 equivalent are calculated taking into account SO2, NH3 and BC emission inventories from CEDS. We acknowledge that we should more explicitly highlight this in the paper and intent to do so in the revised version, since we use SO2 equivalent as a unit in our main results. For a full description of SO2 equivalent calculation, the reader can refer to Stevens et al. [2017].

5. Line 128: You are right that absorption prevails in the presence of clouds only if the aerosols are above clouds. We will mention this in section 3.3.

6. We use the MACv2-SP version from Stevens et al. [2017] that integrates the CEDS emission estimates used for CMIP6 historical forcing input data. As mentioned previously, the MACv2-SP reference year 2005 uses instrumental measurements of the aerosol climatology. Since our results for the year 2005 already suggest a regional dependence of the aerosol efficiency and imply the decoupling, we do not think that more recent emission estimates would affect the results. The emission estimates are only used to scale the 2005 reference to represent the time-varying forcing. In the context of our study, this helps to clearly observe the decoupling in Figure 1, but our key results are the spatial representation with Figure 2,3 and 4.

   Additionally, the results from Quaas et al. [2022] uses these 2021 updated CEDS emission estimates. Using this new dataset they obtain the results discussed in our answer to your comment on Line 29.

Since their results are consistent with our results, we believe that this update would have little effect on our results, and is unlikely to impact the outcomes of our research.

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 details on the temporal-consistency of the plume efficiencies.
- A more exhaustive description of the MACv2-SP aerosol representation, especially summarizing Section 4 on Time-varying forcing in Stevens et al. [2017] (as formulated in this answer to reviewer 2).
- Clarity on the effect of SSA and on the SO2-equivalent emission unit.

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