

The authors lay out a series of trends in anthropogenic aerosol and precursor emissions, column aerosol burdens, aerosol-influenced cloud properties, and top-of-atmosphere radiation that together provide consistent evidence of a reversal in the aerosol radiative forcing trend from more negative values over the twentieth century to less negative (positive trend) over the twenty-first century. This is mainly driven by trends in North America, Europe, and eastern Asia (especially since ~2010) and is somewhat offset by trends in south Asia. The manuscript is a useful review of the trends and related literature and is particularly helpful in putting everything together in one place (e.g., Table 1 and the Supplemental Figure). I believe that some further reporting of the regional breakdowns and of absolute (in addition to relative) changes would strengthen the paper. I recommend prompt publication of a suitably revised manuscript. -MD  
*On behalf of the co-authors I would like to thank Michael Diamond for the thorough review of our manuscript.*

#### General comments

A. Relative versus absolute trends. I understand why the authors chose to report all trends (except for radiative fluxes) in relative, rather than absolute, units. Unfortunately, this choice would make it a bit difficult for someone not already familiar with the spatial pattern of aerosol burden to see the bigger picture. For instance, in Figure 2, one might think the global average trend is of opposite sign between MODIS and MISR just based on the maps shown, although if you were to take the global average, my impression is that both would show a decrease in AOD(f). Perhaps an additional supplemental figure, like the one already included but with absolute units, would be helpful?

*This is indeed a very good suggestion and in the revised version a second variant of Fig. 1-3 is included as supplementary material that shows the trends in absolute units.*

B. Regional breakdown. Table 1 has a nice breakdown of the increasing versus decreasing areas, although I would be interested in seeing a finer regional breakdown (i.e., North America, Europe, east Asia, south Asia, all other). Waterfall plots showing the global change between 2000 and 2019 and the components related to each region for some key variables (e.g., AOD, CDNC, rsutcs) could be really nice, although even just another table or an expansion of Table 1 would suffice.

*The reviewer raises a good point here. We examined thoroughly the possibility to seek reliable results also for smaller regions, but it is, as the reviewer suspected, rather difficult and noisy. It will certainly be of interest to follow this up more closely in future studies. It is also a very good point that it is useful to visualize the results in Table 1, which we do in the revised version along with the table.*

C. Global results. More generally, I think it would be worth reporting globally-averaged values for each variable of interest. It is clear that the authors believe the global trends are positive (decreasing magnitude of ERF<sub>aer</sub>; e.g., Figure 5). This is also clearly implied by the title. It seems clear that the decreasing aerosol regions dominate in the global average over the increasing region(s), so why not just show this directly?  
*Again we agree, although also here there is the “noise” due to the natural-aerosol variability. The numbers are now included in the Table.*

#### Specific comments:

Line 15: ERF<sub>aer</sub> also includes semi-direct effects.

*Indeed! We thank the reviewer for the clarification and include this in the revision.*

Line 16: If you want a classic reference for ARI as well, I'd recommend Chýlek & Coakley (1974).

*This is a very good suggestion and now included in the revision.*

Lines 22-25: As written, this would imply the world has only warmed ~0.5 K since the pre-industrial, when the true value is closer to 1 K. Instead of just citing CO<sub>2</sub> perhaps it'd be better to cite the value for all well-mixed GHGs (sum of ~1.5 K), or state that the aerosol forcing essentially offsets the non-CO<sub>2</sub> well-mixed GHG forcing.

*The reviewer again has a very good point. We now clarify the effects of all major anthropogenic forcings.*

Lines 45-47: This sentence could be simplified or broken up. Also, isn't the claim global, not just regional?  
*The sentence is revised for readability and removed redundancy. Good point again!*

Line 117: Is significance tested using a t-test? Do you account for temporal autocorrelation?  
*Yes, and indeed this information was missing it is now inserted.*

Lines 107-109: Could you provide some more discussion of the differences between the MISR and MODIS trends? Even some statistically significant pixels have opposite trend signs between (a) and (c). Are there differences in the retrievals and their relative strengths/weaknesses or in what conditions retrievals are possible that could help explain this?

*The reviewer indeed has an important point, it is a negligence to not discuss in detail the discrepancy between the MISR and MODIS trend results in particular in the Southern ocean. We do so now and also provide a literature overview over previous studies that already identified (for shorter periods mostly, though) the positive trend in AOD as retrieved from MODIS. One such study proposes it might be due to increases in sea salt emission, but two others find that MODIS stands out with the positive trends: the other products show little or slightly decreasing trends.*

Lines 112-113: If you subset the MODIS and MISR trends to the same period as PMAp, do things look more consistent?

*There are previous studies that look at shorter periods in MODIS as well, and they also show the increasing trends in the Southern ocean. So rather than presenting the new analysis proposed by the reviewer, we now rather report the results of these previous studies.*

Lines 129-130: Especially for LWP, bidirectional changes in response to Nd are now widely acknowledged, so I'm not really sure what the "expected" changes should be in this case.

*The reviewer is right. To make this point clear, we add "not necessarily what is expected".*

Lines 132-133: Similarly, different senses of change in macrophysical cloud properties are possible for different cloud regimes or under different meteorological conditions in the same regime (e.g., Zhang et al., 2022), so it really isn't clear that should be one "expected" change.

*The reviewer is right and this is now explicitly stated at this point of the revised manuscript.*

Line 144: I was a bit surprised by the Gryspeerdt et al. 2016 reference here, as the main point of that paper in my reading is how misleading such correlation analyses can be without the proper statistical controls.

*The reviewer is right, but still Gryspeerdt et al. (2016) concluded that there is a remaining positive relationship between Nd and cloud fraction.*

Lines 158-164: How were model variants treated? Is only one used per model, or do you average all variants for each model, etc.?

*This is an important point! We now clarify that we use the simple arithmetic average.*

Lines 172-173: I'm confused about what the IPCC assessment is referring to here.

Line 174: The emulator ensemble is not introduced.

*We agree that this entire bit on comparison to the IPCC assessment was written in a confusing manner. It is reformulated now: "This result can be compared to the assessment by IPCC AR6 (Forster et al., 2021). Their assessment is based on multiple lines of evidence that are incorporated in an emulator ensemble simulation. The time series of the diagnosed ERFaer is available via the IPCC web site and at <https://doi.org/10.5281/zenodo.5705391>. Computing the linear trend between 2000 and 2019 yields 180 an increase by  $+0.0145 \text{ W m}^{-2} \text{ yr}^{-1}$  between 2000 and 2019 (5 to 95% confidence interval of  $+0.0068$  to  $+0.0253$ ), i.e. by  $+0.29$  ( $+0.14$  to  $+0.51$ )  $\text{W m}^{-2}$  over the full period (Gulev et al., 2021; Forster et al., 2021)."*

Figure 4: It might be worth having another figure (perhaps in the supplement) showing each model individually, and perhaps the radiation fields directly (rsutcs, rsut, rsut+rlut) instead of ERF, for a more apples-to-apples comparison with the CERES record.

*Again a very useful suggestion by the reviewer. We split this proposed modification into two. One is to assess the change in radiation vs. the change in aerosol ERF. For this, we now analysed the piClim\_histall results*

*that include all changes, and analyse the trends in TOA fluxes. In the solar spectrum, it is evident that the aerosol signal dominates. In the all-sky fluxes, however, a signal by the greenhouse gas forcing is superimposed. This result is reported in the main text.*

*The new Supplementary Figures S6 to S8 now examine the trends for the individual models. No surprises are found, and this result is now also reported in the main text.*

Figure 4: It also may be worth looking at variants versus ensemble average for models like NorESM with several variants to explore how much of the noisiness is due to internal variability.

*This is now also done exemplarily for an individual ESM (NorESM was selected) and shown as Supplementary Figure. The result, namely that the pattern of changes is robust, is reported in the main manuscript.*

Figure 4 caption: The gray shading note is for the wrong figure.

*The reviewer is right, this is corrected now.*

Figure 4: The labels for CERES (rsutcs, etc.) are clear to those familiar with climate modeling but aren't obvious otherwise. Please introduce the labels or use another descriptor.

*This is a good suggestion which we follow in the revision.*

Line 221: More explanation of the Smith et al. (2021a) method would be helpful here, and below for Alright et al. (2021) as well.

*We now added a short paragraph on this in the revised text.*

Line 222: What is the range quoted? I'm guessing 5-95% confidence?

*The reviewer is right and this is now clarified in the text.*

Figure 5: Please explain the colors on the x labels. I think I figured it out after staring at it for a bit, but it would be much easier on readers if the information were in the caption.

*Of course, very good point! It is done now.*

Line 240: Zhou et al. (2021) would also be appropriate to reference here.

*This is an excellent suggestion which we follow.*

Line 245: No strong trends in volcanic aerosol, or eruptions, etc.?

*Indeed, the data shown by Carn et al. (2017) suggest no systematic trends. More detail on their study is now provided in the revised manuscript.*

Lines 245-248: Not only wildfires are relevant here but also agricultural burning, especially in Africa. Andela et al. show that burned area has actually been decreasing on average due to human activities, although there isn't a one-to-one correspondence between burned area and smoke emissions.

*This is a very good point, many thanks to the reviewer for pointing to this reference!*

Table 1: See general comment above, at minimum I would add an "all else" column. I also think it would be helpful to have some indication of how things look in absolute, not relative, units, as spatially averaging the percentage changes doesn't necessarily lead to meaningful values given the differences in the absolute amount of aerosol, etc., involved. For the reported values, are you averaging the percentage values from the maps in the figures in space, or taking the absolute values and calculating the percentage trend for the full region?

*As suggested, a column with the global values is now added. The trends in absolute numbers are now reported as supplementary material.*

Table 2: I believe this table is never introduced?

*The reviewer is right and this mistake is now corrected. We now added a short paragraph on this in the revised text.*

References:

Andela, N., Morton, D., Giglio, L., Chen, Y., van der Werf, G. R., Kasibhatla, P. S., DeFries, R. S., Collatz, G. J., Hantsson, S., Kloster, S., Bachelet, D., Forrest, M., Lasslop, G., Li, F., Mangeon, S., Melton, J. R., Yue, C., and Randerson, J. T.: A human-driven decline in global burned area, *Science*, 356, 1356-1362, 2017.

Chýlek, P. and Coakley, J. A.: Aerosols and Climate, *Science*, 183, 75-77, 1974.

Zhang, J., Zhou, X., Goren, T., and Feingold, G.: Albedo susceptibility of northeastern Pacific stratocumulus: the role of covarying meteorological conditions, *Atmos. Chem. Phys.*, 22, 861-880, 10.5194/acp-22-861-2022, 2022.

Zhou, X., Zhang, J., and Feingold, G.: On the Importance of Sea Surface Temperature for Aerosol-Induced Brightening of Marine Clouds and Implications for Cloud Feedback in a Future Warmer Climate, *Geophysical Research Letters*, 48, e2021GL095896, 10.1029/2021gl095896, 2021.