

This manuscript reports the trends of aerosol optical depth, cloud properties, and top-of-atmosphere radiative fluxes in last two decades (2000-2019), mostly from satellite retrievals, to assess the anthropogenic aerosol radiative forcing trends. It also examines the consistency of the trends among AOD, clouds, and radiation. The paper concludes that the anthropogenic aerosol radiative forcing has become globally less negative in this 20-year period, which is consistent with the declining trends of anthropogenic aerosol and precursor emission, aerosol burden, fine-mode aerosols, cloud droplet number concentrations, and TOA fluxes. Based on the findings, it is concluded that the reduction of anthropogenic aerosol leads to an acceleration of the forcing of climate change through both aerosol-radiation and aerosol-cloud interactions.

I find that the manuscript provides an extensive measurement-based information to assess the aerosol radiative forcing on climate, but there are several major issues in synthesize the information to draw the conclusions. Several major issues and specific comments are listed below, and they should be addressed and clarified before the manuscript can be accepted for publication.

On behalf of the co-authors I would like to thank the reviewer for the thorough review of our manuscript.

Major Issues:

Definition of ERF: It is not clear what the definition of aerosol ERF is – is it (a) aerosol radiative effects from anthropogenically emitted aerosols and their precursors? Or (b) the ERF from present-day aerosols minus preindustrial aerosols (e.g. 1750)? Or (c) just the radiative effects of total aerosol? Using modern satellite data implies (c), which is present-day total aerosol effects, but in the paper, it is often casually refer that as aerosol climate forcing or anthropogenic aerosol forcing. Clarification is needed.

The reviewer raises an important point. We always consider both, aerosol-radiation and aerosol-cloud interactions, and as such, a baseline is necessary (no present-day total aerosol effect can be defined for aerosol-cloud interactions, which always require a non-zero baseline). In fact, we have two variants of what the reviewer lists as (b). At some instances, we report the ERF with respect to 1750, in which case we name the baseline. At other instances, we report changes in ERF between two time periods, or as trends between these (specifically, for the period 2000 to 2019), in which case no explicit baseline is required. We now clarify this early on in the Introduction: “Throughout this manuscript, we consider ERF with 1750 as baseline, or changes in ERF over certain periods (such as 2000 to 2019).”

Causality: Even if the trends among aerosol, clouds, and radiative fluxes are “consistent” from satellite observations, it does not mean that the trends can be explained by the reduction of anthropogenic aerosols. There is no effort shown in the paper to separate causality with association. By showing the similarities among the variables is not enough to attribute the trends to the cause. CMIP6 or RFMIP models should be able to provide some insights.

This is right, and it is especially difficult for the cloud quantities. Unfortunately current GCMs are not very reliable in simulating the cloud response to aerosols, so no clear detection-and-attribution study is possible either. In order to respond to the reviewer concern, we now make it more clear still, in the Conclusions section, that the interpretation of causality is questionable in particular when it comes to cloud fraction trends (cloud liquid water path trends were anyway inconclusive): “It is to be noted that the spatial consistency of the trends in cloudiness are not a clear proof of causality.”

“Consistency” between trends in Fig. 1-4: Global map of trends shown in Figures 1-4 are informative, but more in-depth analysis is needed to not only better convey the consistency (or inconsistency) among the trends of AOD, clouds, and radiative fluxes but also different trends of those quantities in various regions. I would suggest show the 2000-2019 time series of each quantity averaged over selected regions (e.g., major pollution source regions, continental outflow regions, and remote regions) to reveal how linear the trends are and if they are indeed consistent with the change of anthropogenic emissions.

This is indeed a useful suggestion. We did the analysis the reviewer suggested, but (as expected) the time series are (as expected) noisy and far from straight lines. This is now reported in the revised manuscript. We also now make clear what exactly we mean by “consistency” where we first refer to it.

Significance of the trends: Areas with “substantial” positive and negative trends are defined as those where the clear sky ERF trends are larger than 0.05 W/m²/year from RFMIP multi-model ensemble mean. According to the caption of Table 1, regions with negative trends cover just 7.3% of the Earth’s surface and that with positive trends covers 1.1%. That implies no trends or weak trends over 91.6% of the Earth’s surface area. How do you explain the significance of global changes of these quantities if the substantial trends are only confined in ~8% of the area?

The reviewer again highlights one of the key issues. The idea is that trends in forcing are isolated, and not so much trends in natural variability. In order to address the reviewer’s remark, we now also report and discuss the global mean trends in Table 1. There is an inconsistency in the AOD trends from MODIS with the other quantities, but overall there is clear support for the main conclusion of reduced aerosol forcing also at the global level. This is now reported in the revised text.

Specific comments:

Line 8: “consistent” with what? With anthropogenic aerosol trend?

Indeed this was poorly formulated. We now expand: “cloud droplet numbers show trends in regions with aerosol declines that are consistent with these in sign.”

Line 16: “ERFari occurs through the scattering and absorption of sunlight by aerosols”: This is the aerosol radiation interaction, which referred to as “RFari” according to IPCC AR5. The ERFari includes additional “semi-direct effects”. Please use the terminology more carefully.

The reviewer is of course right. We now explicitly mention the semi-direct effect.

Line 22-25: If +1.01C temperature change is due to CO₂ and -0.51C due to aerosol, should the net temperature increase be $1.01 - 0.51 = +0.5C$? Or, in other words, it would have reached +1.01C temperature increase without aerosol cooling.

The reviewer again has a good point! We now clarify also the effect by the all anthropogenic greenhouse gases that allows to better compute the full warming number.

Line 38: Do you see a turning point from the 20-year data record when aerosol forcing became substantially less negative? That is why plotting time series is very helpful, as I mentioned in “Major issues” #3, to see if the trends are linear or showing a turning point.

This is a good idea by the reviewer, but the time series are noisy due to lots of interannual variability. It is thus not easy to clearly identify (piecewise) linear trends from such a still rather short period. We will do more in-depth follow-up analysis to seek insights into this point.

Line 58-59: “anthropogenic aerosol emissions over China have been increasing until ~2010 and decreasing thereafter”: That means the emission trend is not only non-linear but also has shifted directions during the past two decades. It will be interesting to see if AOD, clouds, and radiative flux shows similar or different decadal variations.

The reviewer is right, but for these still shorter periods one would expect the signal to be too noisy to draw firm conclusions. But here again, the reviewer raises an important point which a sub-team (the Univ Leipzig group) will investigate in follow-up work.

Line 68-69: CMIP6 used the CEDS_v2017 described in Hoesly et al. 2018, not the newest CEDS version (2021 version).

Indeed this was formulated in a sloppy way and is corrected now.

Line 74-75: Not very clear what you mean “mirror” here, which usually means opposite direction. Do you mean that OC and BC emissions have an increasing (decreasing) trend that mirrors the decreasing (increasing) trend of sulfur emission in the same region? In Figure 1, the regional trends of SO₂, OC, and BC are similar in the same directions, though.

Sorry for poorness in language. We meant, they show the same. The formulation is now corrected.

Line 107-108: Over most oceanic area, MODIS and MISR have opposite AOD trends, especially the fine-mode AOD. What is the implication for global aerosol forcing since ocean covers 70% of the surface area?

First of all, the reviewer has an important point in that it is necessary to discuss this discrepancy in detail. This is now done in the revised manuscript, putting the result reported here into the context of previous studies. Also the relevance for the forcing is now discussed in the new section that reports the global trend numbers.

Line 111 and 112: Is it Metop-A or Metop-B you are using?

We are very sorry for the confusion we created. Metop-A is correct and this is rectified now throughout the manuscript.

Line 114-115: GOME-2 shows different trends over most land regions. Can you be more specific about what the "expected behavior" is? How can the opposite trends in some regions be described as "consistent"?

This statement explicitly is meant only for the highlighted regions with clear anthropogenic trends. We now clarify that we mean the "same", rather than "expected" behaviour.

Line 115, "These trends are largely consistent with those from AERONET data": You have not shown any AERONET data here.

We now report also in the body text at the instance the reviewer highlights that the Aeronet data are shown in Fig. 2.

Line 131-133: Is CDNC less variable than cloudiness and cloud radiative properties? How large should the variability be to prohibit the detection of trend? Again, time series plots may better convey the story.

We now make clear that here we mean the variability in cloud fraction and LWP beyond the one driven by aerosols. CDNC responds to aerosols much more directly than these two quantities.

Line 147-151: There are clearly several regions that the directions or magnitudes of changes between aerosols and cloud properties are not in sync. Can you make more quantitative analysis of different regions, e.g., major pollution regions, immediate downwind regions, and more remote regions, and explain, to the degree you are able, the reason for the consistency or inconsistency between the changes of aerosols and clouds?

This is indeed an important issue, and perhaps a central reason why it is complicated to infer clues about aerosol-cloud interactions: aerosols are not the first-order determinant on most cloud properties, and so there is not a one-to-one relationship. So for the present analysis, it is proposed to make the link where it is possible (i.e. where there are regionally contingent and consistent changes in the aerosols that may be reflected in changes in cloud properties) in the current study.

Figure 4: The terms in Figure 4 are confusing. For example, the caption of Fig 4a says "net broadband solar flux for clear-sky", but the figure title indicates it is "rsutcs", which is defined as "radiation shortwave upward TOA clear sky", not "net". Also it seems the quantities from RFMIP in Fig. 4d-f do not corresponding to the quantities from CERES in Fig. 4a-c: CERES data are the radiative fluxes whereas the RFMIP the effective radiative forcing (meaning either PD – PI, or anthropogenic aerosol only). Lastly, rsut + rlut is total (shortwave + longwave), not net. Please get the terms straight and clarify if you are compare the same or different quantities between CERES and RFMIP.

We clarify now the definitions at the end of the caption and also specify that model and satellite data are comparable. "net" means here, incoming plus reflected (both defined positive downward), and trends for the ERF are shown that are comparable to the trends in ToA net flux changes.

Figure 4 caption, third line from the bottom, the sentence started with "For the emissions...". What is the context of emission here? Besides, there is no grey shading anywhere in all panels.

This is a well-spotted mistake that is a remnant after splitting the figures into four. It is now corrected.

Line 172: Delete "year" in "year0.32 W m⁻²".

Corrected.

Overall, this section is confusing. As I mentioned in "Major Issues" #1, it is not clear whether the discussion is about TOA upward flux, or net flux, or shortwave + longwave flux, or surface downward flux, or if preindustrial condition is considered in the model, or how the RF is defined - is it PD - PI? or is it by anthropogenic aerosol?

In order to make things very clear, we now start this section with the explanation on what exactly is discussed: “Changes in net top-of-atmosphere radiation fluxes in a period correspond to the changes in ERF in that period, but also include the signal of natural variability and of feedbacks to changing climate. “ Also, to further make this clear, we now use the same labels above the panels.

Line 239: delete “but also”.

Done

Table 1 caption, 2nd line: Is CMIP6 or RFMIP models used in Fig. 4? Are they the same suite of models? Please be consistent.

The reviewer is right – it is much better to specifically write RFMIP.

Line 257-258: CEDS emission was not consider any aerosol satellite retrievals.

We meant the opposite, namely that in the construction of the emissions inventories, satellite data are considered, but indeed in a mis-leading and too superficial way. It is now clarified.

Line 269: Remove “aerosol” in “all three aerosol species”. SO₂ is not aerosol, but an aerosol precursor gas.

The reviewer is right, this is removed.

Line 269-270: it is self-repeating that MODIS and MISR AOD increase or decrease at regions aerosols increase or decrease. AOD is a measure of aerosol. Maybe you mean at regions aerosol and precursor emission increase or decrease?

This is true, corrected.

Line 272: What are the expectations? Are the expectations consistent with the aerosol trends or not and why?

The reviewer is right, the formulation was too sloppy. It is now made explicit.

Line 282-286: Again, it seems the terms you compare between CERES and CMIP6 (or RFMIP?) are not the same terms.

They are comparable. We now state this at this point of the text: “The changes in net radiation retrieved by CERES should reflect the trends in ERF, but also natural variability and feedbacks to climate change.”

Figures 1-4: Since the color scales are not linear, it is hard to tell the data range covered by the color bars. Please add numbers for each color interval to help quantify the range.

This is a very good suggestion by the reviewer which we followed.