Reviewer Comments on Lund et al., 2022 - Differences between recent emission inventories strongly affect anthropogenic aerosol evolution from 1990 to 2019

This study presents a comparison of the simulation of aerosols and aerosol radiative forcing over recent decades using a chemical transport model with three different anthropogenic emission inventories. A comparison is made of the simulation using the CEDS emission inventory (used in CMIP6), with two more recently created inventories: ECLIPSE v6b and CEDS21, a recent updated version of CEDS. A comparison is also made to MODIS AOD observations and surface AERONET AOD observations. The results show that simulations using CEDS underestimated magnitude of the total column aerosol burden and aerosol optical depth, particularly over east Asia, compared to the other inventories. In addition, the recent declining in aerosols and aerosol radiative forcing since 1990 is underestimated in these simulations with CEDS. Overall, the study highlights that using the updated anthropogenic emission inventories can better represent recent changes in aerosols, although a model bias in the absolute values of AOD still exists, and their impact on climate, via changes to the radiative balance.

I found this paper well written, with clear graphics and tables. However, it did seem to lack some additional details and more information on the causes and implications of these changes. I have provided some comments below which look to set out these points and where the manuscript could be improved to help the understanding of the topic further.

Major Comments

1. A reoccurring theme within this paper was the influence of biomass burning emissions (and also other natural emissions e.g., sea salt) on particular years when comparing trends between simulations and observations e.g., lines 238-239 for biomass burning and lines 323-234. Since these factors appear to be leading to reoccurring issues in some of the comparisons then it would make the manuscript better if more consideration could be given to dealing with some of these issues and perhaps removing the influence of particular years with high biomass burning emissions e.g. 2019. Or perhaps could a comparative simulation be performed when the biomass burning emissions are set to a fixed climatology to eliminate their influence? A further question arises in that are the linear trends presented in this analysis impacted by the choice of the start year and end year, and could there be a better way of removing the influence on trends of a particular year?

2. Uncertainty is mentioned in the manuscript in terms of observations and model simulations and providing context for the differences between them (e.g. interannual variability of MODIS observations – Fig 3b). I think it would make the figures better if uncertainty could be represented on the figures (e.g. Figure 5), and could better put into context how big or important any differences are. In addition, could some more background on aerosol radiative forcing uncertainty over recent decades (e.g., Regayre et al., 2014) be put into the introduction section to better frame this study.

3. The dipole pattern of radiative forcing differences between South Asia and East Asia is mentioned a number of times in the manuscript but I think the importance of this pattern is not really mentioned. Therefore, I think some text in the manuscript could improve this by identifying what does this pattern mean, why is it important and what are the implications for climate.

4. Would it be best to put the AOD comparison with observations (Section 3.4) at the start of the results section or after the current section 3.2 to show the differences in the model performance over time before then going on to discuss how the different inventories impacts radiative forcing? Also it took me a while to figure out which simulations where used in each section so could this be made clearer throughout the manuscript. Line 305-206 states that the transient simulations are used here but is this the only place they are used?

5. The change of increasing nitrate aerosols in the new inventories in most regions is quite interesting. The text in the manuscript gives a suggestion of why this bit the case but it would be good to have more details on why this occurs and in particularly why does nitrate aerosol decrease over south Asia? This could be of particular interest given that a lot of the CMIP6 models do not include NO3.

6. The manuscript highlights the dominance of aerosol radiation interactions in the radiative forcing calculations. But is this a result of the method that you have use to calculate the radiative effects? If your simulations are nudged and you are using offline radiation calculations, then you wouldn't you expect the calculated aerosol-cloud interactions to be smaller? More comments on the split between the aerosol-radiation and aerosol-cloud interactions would be good in the manuscript. In particular, is the radiative forcing response over South America in Figure S3 due to cloud interactions?

Minor Comments

Line 38 – remove "and" and replace with comma

Line 40 – replace "dominating" with 'dominant'

Line 41 – state year that this plan was put in place and emissions began to decline. Also did the action plan specifically target these gases or was it just a target to reduce PM2.5 concentrations?

Line 45 – Is there not also strong growth in other air pollutant emissions over South Asia?

Line 49-50 – can you give examples of studies showing how aerosol impact regional climate?

Line 58-59 – Are BC and OC emissions lower everywhere in this updated version of CEDS?

Line 60 – Have the reductions in NOx emissions over China (stated on line 42) also been addressed in the revised CEDS emissions?

Line 96 – Perhaps it might be useful to put a small comment here on the evaluation of the model's present-day performance in simulating aerosols with the original CEDS inventory so the reader is aware of existing biases in the model for any particularly aerosol components.

Line 104 – Again perhaps a brief mention of the method used in Quaas et al., (2006) would be useful here for the reader to understand how the change in aerosol in OsloCTM3 is linked aerosol cloud interactions (i.e., an assumed relationship between AOD and CDNC).

Line 132 – Can you state the SSPs used and up to what time period? Also why 3 SSPs are used and are there large differences in anthropogenic aerosol emissions over the time periods of interest?

Line 137-138 – Can you state the natural emissions are kept fixed?

Line 147 – Tables S1 seems to show aerosol burdens and not a list of experiments as stated

Line 160-162 – Is the fact that some regional emissions are quite different pre-2000 important, especially given that your time series simulations with the new inventory only start in 2001 (line 146)?

Line 177-179 – What are the differences between these two sets of numbers? Is it the inclusion of biomass burning emissions?

Line 179 – This is currently Table S1 in the supplement

Line 181 – How small are the reductions in SOA?

Line 184 – Are there differences between anthropogenic VOCs in the inventories which could affect SOA formation? If so the differences in VOCs have not been discussed.

Line 189 – 190 – Is it useful to average the differences in burdens across regions? Doesn't that just take away from the importance of the regional differences? Perhaps best just to identify the largest differences in burden for each aerosol component or give a range of the differences across regions.

Line 199 – There has not been much of a discussion on any differences in NH3 emissions between inventories which could be important for this change in nitrate aerosols.

Line 200-201 – This is true what you have said but I am not sure it explains the increase in nitrate aerosols across nearly all the regions shown on Figure 2. For instance SO2 emissions have been reduced over South Asia in CEDS21 as well but this has not increased nitrate aerosols. This could do with more explanation.

Line 204-206 – Can you expand further on this point as to why it is important. Are you saying that the uncertainty in the inventories are as large as the trend in emissions?

Line 222 – so this means that they are significant differences?

Line 232 – replace "up" with 'higher'

Line 232-233 – If 2019 is a significantly higher biomass burning years does this then create problems when using it as the end year for the calculation of linear trends?

Line 246-247 – I think need to state some evidence to support the statement that "real world emissions have tracked below ssp245" over Asia.

Line 248-249 – Seems a shame to not carry on the discussion of regional trends now.

Line 259 – Is there a reason for the strong negative forcing over South America between 2014 and 1990? I notice AOD has increased but is this related to anthropogenic emission changes?

Line 269-271 – I think you need to refer to Fig S3 in this sentence as well. These are the plots that highlight the shift in emissions with the shift in radiative forcing.

Line 277 – replace "until" with 'by'

Line 297 – Does also this also depend on the speed of the reductions that are occurring in particular regions?

Line 326 – from Figure 5 can you really suggest that there are positive or negative trends in this data? I think you would have to suggest they are very small.

Line 328-329 – what is the trend in the ground based observations and how does it compare?

Line 329-331 – Has there been an observable increase in seas salt aerosol across the oceans across this time period? Since the focus of this paper is on anthropogenic emission inventories is it better to discount the influence of the ocean and take a land-only global mean trend?

Line 332 – is there a reason for the sudden increase in interannual variability in the MODIS observations?

Line 336-341 – There is a strong focus on sea spray changes in studies. Can we be sure that the positive MODIS trend is due to natural aerosols with a contribution from sea salt (and as stated in the conclusions)?

Line 358-359 – Can natural emissions and long-range transport really contribute that much to the large changes seen over Asia between 2018-2020 in MODIS? Is 3 years two short a window for comparison then?

Line 384 – Instead of the global evaluation plots could you also show the AOD evaluation across regions as it would highlight different areas of change and where the change in emissions has caused differences. Also could errors is aerosol process representation also be contributing to these underestimations? I am not sure it is all due to emissions.

Line 416 – “easter” should be ‘Eastern’

Figure 2 – Need to state what year these differences are for.

Figure 3 – Can you make the linear trend lines a bit more distinct? Also panels in c) are currently labelled as b)

Figure 4 – can you make the colour bars wider so it easier to see the different shades