

Interactive comment on “Historical and future changes in air pollutants from CMIP6 models” by Steven T. Turnock et al.

Anonymous Referee #1

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This comprehensive manuscript interrogates past and future changes to surface ozone and PM_{2.5} air pollution in state-of-the-science multi-model simulations from AerChem-MIP/CMIP6 using updated historical and future emissions datasets. The manuscript is thorough and extremely clear and represents a very large simulation and analyses workload involving multiple international institutes. It is important to document the validation of the state of the science global Earth system models and assess the surface air quality responses to past and future global change for new updated emission scenarios. The methodology is sound and the Figures are clear. It may be possible to slightly reduce the number of Figures in the main manuscript further. The multi-model evaluation of surface ozone and PM_{2.5} is highly valuable to the entire chemistry-climate scientific community. The manuscript discusses changes to both emissions versus

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climate, but these are mostly qualitative, and even intuitive, rather than quantitative because none of the applied simulation protocols formally separate out climate change versus emissions change impacts. The authors have done an excellent job with the available datasets and from this perspective the paper is appropriate for publication. However, the results raise some challenging questions about the usage of these global models for surface air quality research. For instance, human health effects calculations depend explicitly on absolute concentrations for exposure. There are some more detailed comments/questions to consider below.

1. The systematic model overestimate of surface ozone across all models is striking (e.g. Fig. 3(c) and (f)). From Fig. 4 for the NAM and EU where there is by far the most data, all models are unable to reproduce the seasonal dynamics (maximum in NH spring and gradually decreasing through the summer months). The authors offer some possible explanations: “The overestimation in the CMIP6 models analysed here could be due to the coarse resolution of the ESMs, an excess of O₃ chemical production (potentially due to an overabundance of NO_x and/or VOCs) and weak O₃ deposition.”. If possible, it would be good to have a more robust and clear explanation and understanding of the systematic overestimate and poor seasonal dynamics? Is the coarse resolution problem related to directly injecting the NO_x emissions across the large spatial extent $\sim 2\text{deg} \times 2\text{deg}$ ($\sim 200\text{km}$) grid cells? Where the ozone production regime will be highly NO_x-limited at this scale? What is needed from the community to improve/address the systematic positive bias in surface ozone simulations in global models?

2. The systematic underestimate in monthly PM_{2.5} in NAM, EU and EAS (Fig. 6) is troubling. Can it really be explained only by the missing nitrate component? Are there other fundamental missing or misrepresented processes? Output from these models is more frequently being used to assess health impacts, for example, premature mortality due to outdoor air pollution exposure (PM_{2.5} and ozone) but such application would not be justified based on the model/measurement comparison here. It could be argued

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from the model/measurement evaluation that the models cannot be applied as tools to study the surface air quality?

3. How reliable are the model simulations of past and future changes when the monthly mean surface air quality concentrations cannot be reproduced by the models and there are clear systematic biases?

4. Fig. 9. I find this Figure also striking in the diversity of model results for historical surface ozone evolution. Why does the GISS model have such large changes/sensitivities to the PI-PD? Esp. for Europe, S. Asia and E. Asia (but not SE Asia + less polluted SH regions)? Does the GISS model gas-phase chemistry have a larger sensitivity to NO_x changes than other models and why? The GISS model is also an outlier in Fig. 10 for evolution of PM_{2.5} over S. Asia region specifically? What is the value of the multi-model mean in e.g. Fig. 13 when there is such large diversity of sensitivities shown in Figs. 9&10?

5. "Surface O₃ increases across most world regions in this scenario can be attributed to the large increase in global CH₄ abundances (80%) and the large predicted increase in surface temperatures". Why do increases in surface temperature increase surface ozone concentrations independent of emissions? What is the mechanism? Is it temperature, or co-varying stagnation or light/downward SW? How do we know it is temperature with 100% certainty as stated here?

6. "across East Asia the additional precursor emission reductions in ssp370-lowN₂O have made little difference to surface O₃ concentrations predicted by the CMIP6 models, indicating that other factors are more important over this region (chemistry or climate change)." This result is critically important. So, aggressive mitigation of ozone precursors has no impact on the surface ozone concentrations in this region relative to a scenario with those precursors? What is the reason for surface ozone in East Asia to be independent of ozone precursor emission changes under this level of global change? Further explanation is needed. Are there climatic feedbacks from the precursors themselves that are offsetting the changes?

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sors themselves that are offsetting the changes?

7. "Discrepancies in the magnitude of change in these emissions due to climate and *land-use change*". Please specify in similar to Table 1 the models for which the natural emissions and atmospheric chemistry are actually dynamically coupled with the climate model's land surface scheme and vegetation cover / Plant Functional Types (that are dynamically changing in the simulations due to human land use change). Which models have the BVOC emissions actually coupled to the climate model's internal land surface scheme? If uncertainty in the changes to natural emissions is an important conclusion of the paper, there needs to be a separate Table describing the representation of those emissions in each model.

Minor comments

I find Fig. 2 challenging to look at and wonder about for other readers. I appreciate it is difficult to show this Fig. 1 type information across multiple regions.

Is it necessary to have both Fig 6 and Fig 8 i.e. for the 2000-2010 and 2005 and 2014 periods? Could one of the plots go into SI?

"Large regional historical changes are simulated for both pollutants, across East and South Asia, with an increase of up to 40 ppb for O₃ and 12 μg m⁻³ for PM_{2.5}." and similar sentences in abstract. Need to include the temporal averaging associated with those values in abstract (annual).

"Near Term Climate Forcers (NTCFs)." IPCC AR6 uses "Short-lived Climate Forcers (SLCFs)".

"Initial assessments have been made of future changes to air pollutants in the SSPs using simplified models." Need to add references here.

"A particular climate mitigation target, in terms of an anthropogenic radiative forcing by 2100, is included on top of each SSP" What does "on top of" mean exactly?

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“However, scenarios with large increases in global CH₄ abundances, a large climate change signal and limited control of precursor emissions fail to restrict regional increases in surface O₃, leading to poor future air quality and potential human health impacts (Silva et al., 2017).” Is this statement redundant/obvious? Where is the new science?

“Whilst there is disagreements” sp. there are

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