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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Influence of future air pollution mitigation strategies on total aerosol radiative forcing" *by* S. Kloster et al.

S. Kloster et al.

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We thank the reviewers for their constructive comments and suggestions. We hope to have addressed all raised issues in our comments on the individual reviews.

Anonymous Referee 1:

Specific major comments:

A) P5586: I suggest the authors add a section 5.3 on the additivity of aerosol and oxidant perturbations. Though I hate to suggest an addition to an already long paper, all the material is there and this would be a useful addition to have a short discussion of this aspect on non-linearity. It seems oddly left out at present.

We agree that a discussion of the additivity of aerosol and oxidant changes is interesting and we can address this question with simulations performed for the present study.

We added one figure panel to Figure 3 showing the deviations from additivity and a paragraph to section 5.1 Influence of oxidant concentrations:

As a result the combined response of changes in oxidant concentrations and aerosol and aerosol precursor emission changes are non-linear. Figure 3d shows the additivity of the sulfate burden for the MFR:2030 case. Additivity is thereby defined as:

a = ΔMFR:2030 - (Δ2000:CHEM:2030:MFR + ΔMFR:2030:CHEM:2000)

with ΔX = X - REF and X ∈ (MFR:2030, 2000:CHEM:2030:MFR, MFR:2030:CHEM:2000); REF is the reference experiment 2000. A negative deviation from additivity implies that the decrease in sulfate burden is higher in the sum of the individual experiments (Δ2000:CHEM:2030:MFR + ΔMFR:2030:CHEM:2000), in which aerosol emissions and oxidant concentration are changed separately, than in the combined experiment (Δ MFR:2030). A positive deviation implies that the decrease in sulfate burden is lower in the sum of the individual experiments than in the combined experiment. We find positive deviations reaching up to 0.48 mg(S)/m2 (3% compared to the reference simulation) over Asia and parts of North America. Parts of Europe show slightly negative deviations (-0.16 mg(S)/m2, 4% compared to the reference simulation). Deviations from additivity result from a stronger response of the sulfate burden towards changes in oxidant concentrations in the case aerosol and aerosol precursor emissions remain at their high present day values (2000:CHEM:2030:MFR) compared to the response when aerosol and aerosol precursor emissions are reduced (MFR:2030, compare Fig. 3b and 3c). On the global annual mean the deviation from additivity is rather small (1.8 Gg(S), 0.2% of the sulfate burden simulated for the reference simulation).

B) While Figure 2a shows the zonal mean TOA SW forcings, this is really one of the main results of the study and the paper should include another figure showing the spatial maps of this forcing in the various experiments (at least those with substantial responses over at least some areas) .....

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We added a figure in the appendix (Figure A3) showing the annual mean regional distribution of the TOA SW radiative forcings for the baseline experiments (PI, MFR:2030, CLE:2030, MFR:2030:IP, MFR:2030:DT)

C) Figure A1C shows a fairly substantial positive bias in BC. Its difficult to tell on a log scale, but appears to be around 50-100%. This needs to be explicitly acknowledged in the text as a caveat to the study (the BC responses may be an upper limit?).

The model has a negative bias in BC and to a lesser extent in POM surface concentration when compared to IMPROVE measurements over North America. For BC 45% of the samples (total 115 samples) underestimate the measurements by more than a factor of 2. We added these values (analog for BC and SO4) in the Figure showing the comparison between simulation and measurements (see also referee2 comment 3). The agreement is better when instead of the IIASA emission inventory BC emissions from Bond et al. (2004) are applied (65% of the samples agree within a factor of two with the measurements) as done in the ECHAM5-HAM reference simulation (Stier et al. 2005). A likely reason for the negative bias is thus an underestimation of the present-day emissions over North America in the IIASA emission inventory applied in the current study. Therefore, the BC as well as the OC response towards future changes is likely to be a lower estimate in the current study.

We changed the manuscript to acknowledge this caveat:

While the comparison with BC and POM surface concentration measurements over North America shows in general a good agreement for the ECHAM-HAM5 reference simulation, the surface concentrations in our study tend to underestimate the observed values (most pronounced for BC). The simulated lifetimes for BC and POM are almost identical in both studies. The differences are solely caused by the different emission inventories. Stier et al. (2005) applied the Bond et al. (2004) inventory for anthropogenic BC and POM emissions which are higher over North America compared to the IIASA inventory (23% for BC and 7% for POM emissions). Therefore, the BC as

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well as the OC response towards future changes is likely to be a lower estimate in the current study.

Additional comments:

1. P5568, L5: Section 2.2: While later on the authors explain that oxidants are prescribed with offline fields, I feel this information needs to be in section 2.2 While not part of the aerosol model per se, its intrinsic to aerosol chemical processing. Additionally, the description that is eventually given of offline oxidants is not sufficient. Are all oxidants offline? Including hydrogen peroxide? It may be reasonable for OH and ozone, but for soluble hydrogen peroxide this my cause substantial biases.

We changed section 2.2. 'The aerosol model HAM' to clarify what oxidant concentrations are prescribed to: The main components of HAM are the microphysical core M7 (Vignati et al, 2004), an emission module, a sulfur oxidation chemistry scheme using prescribed oxidant concentrations for OH, NO2, O3 and H2O2 (Feichter et al. 1996), ...

we added a sentence in section 3.2 'Oxidant concentrations' discussing the uncertainties related to prescribing hydrogen peroxide: In addition, prescribing H2O2 concentrations might underestimate the resulting sulfate burden as H202 is strongly depleted in anthropogenic source regions by aqueous reaction with SO2. This will increase the gas-phase production of SO4 which is less susceptible to scavenging and increase the SO4 burden (Barth et al., 2000, Roelofs et al., 1998).

2. P5570, L26: Its not clear what the global mean decrease of 13% refers to here. SO2 oxidation, surface sulfate, or what? Please clarify.

We changed the manuscript to: ... leading to a lower gas-phase production of SO4 and subsequently to lower SO4 surface concentrations (the global annual mean decreases by -13%)

3. P5571, L9: As you've discussed the model underestimate BC over North America,

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it'd be better to say the model shows reasonable good agreement here.

We changed in the manuscript to: Overall the ECHAM5-HAM version used in this study shows reasonable good agreement with observations ...

4. P5571, L23: The authors point out that one of the conventional definitions of RF includes fixed tropospheric temperatures. This is important in the case where adjusted RF is calculated, meaning that stratospheric temperatures are allowed to adjust while those in the troposphere are not. However, I do not believe this is the case in this study. The authors should clarify where they are applying the nudging. If its the entire atmosphere, as I suspect, then they need to clarify that they are not simply holding tropospheric temperatures fixed while allowing the stratosphere to adjust, as the current wording implies. Instead, they are calculating something more akin to instantaneous forcing rather than adjusted, though it is not exactly that either due to the aerosol indirect effects.

We changed the paragraph introducing the section 3 'Simulation Setup' to clarify the applied nudging technique and the radiative forcing calculation to:

We performed a series of experiments applying different future aerosol and aerosol precursor emission scenarios to investigate the associated aerosol radiative effects. In all these experiments the large-scale meteorology is constrained to the year 2000, nudging the ECHAM5-HAM simulated temperature, log surface pressure, vorticity and divergence to the ECMWF ERA40 reanalysis data (Uppala et al., 2005). With the nudging technique the large-scale meteorology is constrained, whereas smaller scale processes, such as cloud formation, respond to perturbations induced into the system (Jeuken et al., 1996). Thus, aerosol effects on the meteorological state are small. The nudging technique allows to a large extent compliance with the definition of the adjusted radiative forcing (RF) as given by (Forster et al., 2007), which is defined as the change in net (down minus up) irradiance at the tropopause after the introduction of a perturbation with surface and tropospheric temperatures and state of meteorology

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held fixed at the unperturbed values, while allowing the stratosphere to adjust. The difference is that in the set-up applied in this study: (i) temperature and state are held fixed for the entire atmosphere which equals instantaneous RF according to Forster et al. (2007). For most tropospheric aerosol forcing stratospheric adjustment has little effect on the RF, and the instantaneous RF at the top of the atmosphere (TOA) equals the adjusted RF; (ii) aerosol-cloud feedback mechanisms are enabled.

5. P5572, L7: It is probably worth noting the the CLE scenario assumes full compliance with legislation (which is clearly optimistic).

We changed in the manuscript to: CLE reflects current perspectives of economic development and takes into account presently decided control legislations for future development assuming full compliance.

6. P5572, L23: Please identify the aircraft emissions used along with the shipping here and in Table1

We do not consider any aircraft emissions. We clarified this in the text:

Emissions from international shipping and aviation were not included in the IIASA emission inventory. We added only international shipping emissions from a different inventory ...

7. P5576, L2-5: this description of sea salt, dust and DMS emissions belongs in section 3.1, not in the results.

They are described in section 3.1. and are mentioned here again in the result section as these emissions are calculated interactively in the model and thus change with different forcings.

8. P5576, L8-9: It sounds odd to say that global emissions decrease over particular source regions. This sentence could be rewritten.

We changed in the manuscript to: BC and POM emission decrease globally with -12%

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and -8%, respectively

9. P5577, L22: Please elaborate on why emissions at a lower latitude would lead to a longer lifetime. One might suppose wet removal would be faster at lower latitudes, hence the opposite response.

We added to the manuscript: The increase in SO4 lifetime is apparent in all experiments, which show a distinct shift of SO2 emissions towards low-latitude regions. Aerosol lifetime is influenced by a number of competing and interacting mechanism. Graf et al. (1997) showed that natural emitted SO2 from volcanoes and DMS in the low latitudes have a longer lifetime compared to anthropogenic emissions in high-latitude regions caused by less effective dry deposition. Stier et al. (2006) showed a similar response as simulated in this study for a shift of anthropogenic emissions towards lower latitudes, going along with a strong increase in aerosol burden over semi-arid tropical regions (Fig. 1(d)).

10. P5578, L3: It would be helpful to the reader if the authors explained more explicitly how sulfate affects carbonaceous aerosols in their model in the interactions referred to here. Thus far they've only said the they are internally mixed, but a short qualitative description of sulfate influences BC/POM hydrophobic/hydrophilic transition would be useful here.

We added a paragraph in the model description (Section 2.1: The aerosol model HAM) explaining the transfer from hydrophobic to hydrophilic: Particles in the hydrophobic modes are transformed to the corresponding hydrophilic/mixed mode by condensation of sulfate on the surface or by coagulation with particles of the hydrophilic modes. The total condensable sulfate and the sulfate added by coagulation are attributed to the number of particles that can be coated with a minimal layer of sulfate in the respective mode. As minimal layer thickness a mono-layer is assumed. ...

11. P5579, L15: The authors use the term RF perturbation. The word perturbation is redundant, as forcing already requires a perturbation. It would be more descriptive to

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use TOA RF instead.

We use the term radiative forcing perturbation to describe future changes in radiative forcing to clearly distinguish between future anthropogenic forcings and present day anthropogenic forcings. We agree that this might be misleading and we changed the formulation as follows:

We calculate the present-day anthropogenic top-of-the-atmosphere (TOA) radiative forcing (RF) as the difference between the present day simulation (2000) and the pre-industrial simulation (PI), hereinafter referred to as TOA RF (2000-PI). Analogously, the future changes are defined as TOA RF (2030-2000).

The revised manuscript we changed accordingly.

12. P5579, L19: The authors say that they diagnosed atmospheric RF, but then proceed to describe that this is atmospheric absorption and not RF at all. Please just call this atmospheric absorption.

We changed the manuscript to:

We also diagnosed the atmospheric absorption, which is the difference in net solar radiation between TOA and surface and the surface RF, which indicate changes in incoming solar radiation at the Earth's surface.

13. P5579, L27: It is unclear to me what is meant by total aerosol RF here.

We refer here to the direct plus indirect effect. We changed the manuscript to clarify this: The global annual mean total (direct plus indirect) aerosol RFs for the different experiments are summarized in Table 3 ...

14. P5581, L7 and L15: I do not see how you can get either of these numbers from the MFR vs MFR EUROPE or MFR vs MFR Asia experiment. ...

We corrected the numbers in the revised manuscript (see also referee2 comment 24 and 25):

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If only Europe will follow a maximum feasible reduction strategy in the future (MFR:2030:EUROPE) the total aerosol global annual mean TOA RF (2030-2000) amounts to 0.00 W/m2 by 2030. Thus, a maximum feasible reduction of aerosol and aerosol precursor emissions over Europe leads only to a small additional negative global annual mean TOA RF (-0.02 W/m2) compared to the case in which worldwide CLE 2030 is applied (CLE:2030). A comparison with the MFR:2030 experiment shows that the TOA RF will be 52% higher in the case MFR 2030 is not only applied over Europe but worldwide.

In contrast, an implementation of a maximum feasible reduction strategy in Asia (MFR:2030:ASIA) leads to a strong positive TOA RF (2030-2000) across Asia (up to +6 W/m2). The global annual mean TOA RF amounts to +0.32 W/m2. This is substantially higher than the +0.02 W/m2 simulated in the CLE:2030 experiment, reflecting the large potential to reduce aerosol and aerosol-precursor emissions in Asia. Compared to the MFR:2030 experiment the positive TOA RF is 23% higher when MFR 2030 is not applied only for Asia but worldwide.

15. P5582, L10: The oxidant concentrations used in the sensitivity studies are assumed to span a realistic range. It seems the may cover the range of changes that might result of anthropogenic emissions, but it is worth pointing out that they do not account for potential changes in natural emissions under a changing climate (e.g. Biogenic VOCs, methane). In addition, the OH changes will be very sensitive to the ratio of NOx to CO+VOC changes, as these push OH in opposite directions, so that oxidant changes could be rather different than those used here if species specific emissions were to evolve differently.

We changed the manuscript to clarify that we only consider changes in oxidant concentrations caused by air pollution mitigation: Abstract: For changes in oxidant concentrations caused by future air pollution mitigation we do not find a significant effect for the global annual mean radiative aerosol forcing. Conclusion: Overall, we conclude that the influence of air pollution mitigation strategies for oxidant concentrations in a realis-

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tic range between present day and 2030 will not significantly alter the SO4 production and thus the influence on TOA RF is rather small.

16. P5582, L27: I do not follow the pronounced differences refers to here. Between no change in oxidants and changing oxidants, or no change in oxidants and present day runs?

The paragraph refers to the impact of changing oxidant concentration (keeping them constant instead of applying air pollution mitigation)

We changed the manuscript to: If oxidant concentrations would remain identical to present day conditions, a situation that would be roughly representative for the absence of further mitigation measures to reduce ozone precursor emissions, we simulate only small impacts in the global annual mean SO4 burden (Table 4) for both aerosol and aerosol precursor emission scenarios (compare CLE:2030:CHEM:2000 and MFR:2030:CHEM:2000 with CLE 2030 and MFR 2030, respectively). However, regionally we find pronounced differences in the SO4 burden (Fig. 3a and b).

17. P5585, L13: The section 5.2.1 discussed the additivity of aerosol RF. This is the whole and only point of section 5.2, so the subsection heading should be removed.

Done

18. P5588, L1-2: The last sentence of this paragraph is repetitive and should be cut. The total and Asian values have been given already, so readers can easily to the math themselves.

Done

19. P5589, L8: A comment should be added at the end of the line about oxidant chances; as a result of air pollution mitigation and climate change, the latter of which was not included here.

We changed the manuscript to: Overall, we conclude that the influence of air pollution

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mitigation strategies for oxidant concentrations in a realistic range between present day and 2030 will not significantly alter the SO4 production and thus the influence on TOA RF is rather small. A recently developed new version of the ECHAM5-HAM model including the full MOZART chemistry scheme as described in Pozzoli et al. (2007) will allow more consistent simulations in the future.

20. P5589, L25: the word globally should be inserted after production. The paper has shown that substantial local changes are possible (Figure 3, for example), which could also be noted if the authors like.

Done

21. P5589, L26: I suggest adding likely to be before rather small here.

Done

22. P5590, L13: The authors should cite previous work on air quality/climate linkages here, such as the recent multi-model study of Shindell et al., JGR, 2008. That study explored the climate impacts of various potential air pollution emission trajectories, so is quite relevant to this work.

We changed the manuscript to: Finally, we want to stress that measures aiming at improving future air quality will have implication on climate change mitigation strategies aimed at maintaining global warming below a specific threshold (Brasseur and Roeckner (2005), Shindell et al. (2008)).

Technical corrections:

all implemented accordingly in the revised manuscript.

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