Air quality and radiative forcing impacts of anthropogenic volatile organic compound emissions from ten world regions

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Response to Referee Comments

Response to Anonymous Referee #1:

We thank Referee #1 for reviewing the manuscript and providing thoughtful comments. We have responded to each comment below and have noted the page and line number for each revision to the manuscript. Referee #1's comments are in blue italics.

Fry and co-authors have written a detailed analysis of regional differences in NMVOC impacts on air quality and climate. This manuscript advances beyond past work mainly by analyzing a larger number of regions. Their model predicts smaller RF from NMVOCs than past work, which they attribute mainly to differences in other emissions. The manuscript is thorough and clearly written, emphasizing the regional differences in GWP that may be useful to other scientists and policy makers. I recommend publishing this with the following minor changes.

Thank you. Your suggestions have been valuable toward improving this manuscript.

The estimated present-day net RF, relative to preindustrial, includes all biomass burning NMVOC emissions in the RF calculation. This implicitly assumes that there were no preindustrial NMVOC emissions from biomass burning, which is incorrect. I suggest using the RCP scenario for historical biomass burning to provide a better estimate of the net RF since preindustrial times, with caveats on the inventory accuracy.

In our CTM simulations, we use the RCP8.5 anthropogenic emissions dataset for the year 2005 in the base and perturbation experiments, which includes anthropogenic biomass burning emissions (grassland fire and forest fire emissions), but not natural fires. Our perturbation experiments reduce NMVOC emissions by 50% (in each region) from all anthropogenic sectors except biomass burning (grassland fire and forest fire emissions); biomass burning emissions are left unchanged. As described in the paper, we focus on anthropogenic emissions apart from grassland and forest fires in order to develop policy-relevant GWP estimates, which is the main purpose of the paper.

Nonetheless, we estimate a total RF of NMVOCs for the purpose of comparing with other studies. To calculate the present-day net RF, we scale the RF due to non-biomass burning anthropogenic NMVOCs by the total NMVOC emissions including biomass burning, to get a total net RF for all anthropogenic NMVOCs. In response to Referee #2, we revised our method of scaling to now account for the geographic variability in biomass burning emissions. Please see our response below under Referee #2's comments. Because the RCP8.5 emissions we use only report anthropogenic fire emissions, and we scale to approximate removing these emissions, we believe that we estimate anthropogenic RF appropriately here. There likely were fire-related NMVOC emissions in the preindustrial, which are omitted in our scaling method, but we also do not model 2005 natural fire emissions. Our definition of RF therefore focuses on anthropogenic contributions.

However, the Reviewer is correct that our method differs from the IPCC's standard definition of RF, which is relative to 1750 (in the new AR5 report) and relative to 1850 in the ACCMIP study (Stevenson et al., 2013). As a result, we have decided to remove our total global net RF estimate from the abstract and summary sections, so as not to emphasize our numerical estimate, but we retain our estimate in Section 4 to compare with other studies.

We have removed the following sentence from Page 21126, Lines 18-20:

> The total global net RF for NMVOCs is estimated as 0.0277 W m^{-2} (~1.8 % of CO_2 RF since the preindustrial).

And we have replaced the sentence on Page 21139, Lines 9-10 with:

> The global annual average net RF for the global 50% NMVOC reduction is estimated as -9.73 mW m^{-2} or 0.21 mW m^{-2} (Tg C yr^{-1})$^{-1}$.

We have left our present-day net RF estimate in section 4, but have added text to clarify that our estimate of anthropogenic RF differs from other definitions of RF on Page 21135, Line 25:

> This approach accounts for the geographic variability of biomass burning emissions, yet assumes the same mixture of NMVOC species as anthropogenic emissions. The influence of climate change from the preindustrial to present day is also omitted, as all simulations use 2005 meteorology. While this estimate of total net RF is derived using methods that differ from the ACCMIP and IPCC AR5 standard RF definitions, which are present-day relative to the year 1850 and 1750, respectively, it provides an opportunity to compare with other studies.

Abstract line 16 says RFs are negative (when NMVOC) emissions are reduced, then line 19 says RF is positive (since preindustrial era). While both statements are correct in context, this is confusing. I suggest beginning the sentence on line 19 with, "NMVOC emissions have increased since the preindustrial era, so the global net present-day RF, relative to preindustrial, is ..."

Based on our response above, we have removed the present-day global net RF estimate from the abstract (Page 21126, Lines 18-20).

To address the Reviewer's concerns, we have added a sentence to clarify that GWPs and anthropogenic RF are positive, while the RF impacts of NMVOC emission reductions are negative on Page 21137, Line 24:

> The sign of the total global net RF and GWPs are mostly positive, as they represent the overall warming effect of present-day anthropogenic NMVOCs compared to the preindustrial, in contrast to the negative global annual average net RFs (Table 4) due to reductions in NMVOC emissions.

The abstract attributes the lower RF and GWP values in this work compared to previous estimates to, "differences among models in ozone, methane, and sulfate sensitivities, and the climate forcings included in each estimate." This is vague, whereas section 4 clearly attributes the differences to greater NMVOC/NOx emission ratios in the current work. I suggest using the more clear section 4 claim in the abstract. It would also help to clarify if the emission inventories

used in the earlier work have later been found to be erroneous, or if the inventories differ mainly because they are meant to represent different years (year 2005 in this work vs. ???? for previous work).

The emissions inventory from the HTAP multimodel study (Fry et al., 2012) was not found to be erroneous. However, it did represent a different base year (2001). Here we evaluate 2005 emissions from RCP8.5.

We agree that section 4 provides a better explanation, and have revised Page 21126, Lines 22-24 to clarify our lower RF and GWP estimates:

> The NMVOC RF and GWP estimates are generally lower than previously modeled estimates, due to the greater NMVOC/NO_x emissions ratios simulated, which result in less sensitivity to NMVOC emissions changes and smaller global O_3 burden responses, in addition to differences in the representation of NMVOCs and oxidation chemistry among models.

We also have added that Fry et al. (2012) used 2001 emissions, on Page 21138, Lines 8-10:

> Here total NMVOC/NO_x emissions ratios are 57% greater globally and in NA than the multimodel mean ratios, partly due to greater biogenic NMVOC emissions (calculated online in MOZART-4) for the year 2005. The multimodel ensemble evaluated by Fry et al. (2012) utilizes an emissions inventory representative of the year 2001.

In addition, is the model resolution in the current work higher than in previous estimates of NMVOC RF? Model resolution can alter O3 production and its response to emission perturbations. In general, higher resolution tends to reduce O3 production, which might be consistent with the smaller RF in this work compared to prior papers.

The model resolution used here (1.9° latitude×2.5° longitude×56 vertical levels) is within the range of resolutions used by the HTAP model ensemble in our 2012 paper. We do not believe the resolution differences contributed substantially to decreased O_3 production.

*p3 line 22. More accurately, VOC oxidation *begins* with reactions with OH, as well as O3 and NO3 radicals. Many other reactants enable subsequent oxidation steps.*

We have revised Page 21127, Lines 22-24 to the following:

> NMVOCs are oxidized by the hydroxyl radical (OH) in the troposphere, producing peroxyl radicals (RO_2) and hydroperoxy radicals (HO_2) that then oxidize nitric oxide (NO) to yield O_3. Other reactants (e.g. O_3 and NO_3 radicals) also contribute to oxidation reactions.

Response to Anonymous Referee #2:
We thank Referee #2 for reviewing this manuscript, and providing detailed comments and suggestions. Our responses to each comment are below. In our comments, we reference the page and line numbers for each revision. Referee #2's comments are in blue italics.

This study uses a state-of-the-science chemistry-transport model (MOZART4) forced with short-lived precursor emissions for year 2005 from RCP8.5 scenario to quantify the impacts of reducing NMVOC emissions by 50% globally and for 10 regions. The study quantifies the NMVOC reduction impacts on indirect radiative forcing (for ozone, methane and sulfate), and air quality (ozone only). The results are used to determine the policy metrics GWP20 and GWP100 for NMVOCs. The radiative forcing calculations are performed off-line using the NOAA GFDL RTM. The strength of the study is joint consideration of both the air quality and radiative impacts of the NMVOC emissions reductions. The study is an extension of various multi-model assessments previously published as part of ACC-MIP and HTAP and as such does not include any new or scientifically exciting insights. The manuscript is well written, clear, concise and does have policy relevance. It is important that such results continue to be documented as long as the scientific and policy communities wrestle with identifying win-win strategies for air quality and climate mitigation, and possible multi-gas approaches to climate policy.

Thank you. Your feedback has been very helpful in improving this manuscript.

There are 2 major weaknesses in the paper that need to be addressed quantitatively before the paper could be considered for Atmospheric Chemistry and Physics. Firstly, I agree that neglecting aerosol-cloud indirect effects is justified because these effects are so uncertain (sign not robust across models). However, the scattering effects and resultant radiative impacts of SOA and nitrate need to be included in the analyses and the GWP calculations. The MOZART4 simulations include nitrate and SOA aerosol but then the authors neglect to quantify the radiative forcing of these aerosol changes in the RTM. The hard part is putting these aerosols into the CTM, it should be more straightforward to assess the radiative forcing from the aerosol changes. If nitrate and SOA are included in the simulations and discussion, neglecting their effects on the radiative impacts is not acceptable. Accounting for the nitrate and SOA RF could help elevate the paper to an exciting new level and could affect the sign of the regional net RFs.

We agree that including nitrate and SOA in the RF calculations would strengthen our paper. However, the GFDL standalone RTM had not been developed to calculate the RF due to nitrate and SOA, and while nitrate is likely simulated well by the CTM, we have little confidence in the SOA changes, as these mechanisms are still very uncertain. As we are using the RTM as embedded within the GFDL general circulation model (GCM) – essentially running the GCM for one day each month, iteratively for the stratospheric adjustment – adding new species to the RTM requires coding changes to the GCM that are not trivial. In addition, the simulations completed here are highly computationally demanding and cannot be repeated easily.

For these reasons, we regrettably cannot include nitrate and SOA in the RF calculations at present. We hope that these changes can be made to the GFDL RTM and GCM shortly, for use in future studies.

We have revised the discussion of this issue on Page 21136, Lines 24-27 and Page 21137, Lines 1-2 to:

> Since the RTM does not calculate the RF of SOA and NO_3^- aerosols presently, our simulated net RFs omit these forcing contributions. If changes in SOA and NO_3^- were accounted for by the RTM, tropospheric SOA decreases (greater than SO_4^{2-} changes in some regions) would likely add small regional warming, while tropospheric NO_3^- increases and decreases (mostly lesser than SO_4^{2-} changes) would add slight regional cooling and warming effects. Globally, NO_3^- and SOA would likely contribute small negative and positive RFs, respectively, to global net RF. Previous modeling studies have estimated NO_3^- RF as -0.06 W m^{-2} (Bauer et al., 2007) and SOA RF as -0.06 to -0.09 W m^{-2} (Hoyle et al., 2009), for present day relative to the preindustrial, both of which are small compared to greenhouse gas forcings like tropospheric O_3 (0.41 W m^{-2}) (Stevenson et al., 2013).

Secondly, the paper claims to assess air quality impacts but stops short at ozone. The paper needs to include a quantitative assessment of the surface PM2.5 impacts. All the major aerosol components of PM2.5 are already simulated in the model and available in the model output.

We have already included the spatial distributions of $PM_{2.5}$ changes (Figures S11, S14, S15, S16) and regional and global annual average surface $PM_{2.5}$ changes (Tables S7, S8, S9) in the supplemental material. While the results are interesting, the magnitudes of these changes are quite small (on the order of ng m^{-3}), so the corresponding figures and tables have been placed in the supplement rather than the main paper. However, we have added several sentences to Page 21135, Line 19 to discuss more quantitatively the surface $PM_{2.5}$ impacts:

> Summing the effects of individual aerosol species shows that the greatest changes in fine particulate matter ($PM_{2.5}$, estimated as a sum not including dust and sea salt) occur within the reduction region, and extend intercontinentally in some cases (Figure S16). Halving global NMVOC emissions slightly decreases global annual average surface $PM_{2.5}$ by 28 ng m^{-3} (0.89%), and regional annual average $PM_{2.5}$ by 1.8 ng m^{-3} (0.10%) (AU) to 384 ng m^{-3} (2.3%) (EA) (Table S7). Regional NMVOC reductions also have a small influence on surface $PM_{2.5}$ concentrations in other regions (Table S8, S9), but halving NMVOCs generally does not have a strong influence on $PM_{2.5}$ air quality.

One further issue is the scaling for biomass burning NMVOC contribution to RF. It is rather strange. The author's rationale ": : :.which are excluded since actions to address biomass burning differ from the other anthropogenic sectors, and would likely reduce a suite of emissions simultaneously" is weak and incorrect because actions to address the other anthropogenic sectors (industry, transportation etc.) would also reduce a suite of emissions simultaneously. However, the impact of reductions in biomass burning NMVOC emissions on atmospheric composition is likely to be quite different to industrial NMVOCs, which is why the scaling is dubious. I suggest that the authors either repeat the experiments including biomass burning emissions in the regional 50% NMVOC reductions, or do a separate global experiment reducing

biomass burning NMVOCs by 50%. The CTM simulations in the study are only1.5 years in run length so that is reasonable.

The main purpose of this paper is to estimate regional GWPs for NMVOC emissions from different world regions, for anthropogenic emissions apart from those related to biomass burning. As such, we emphasize anthropogenic emissions apart from biomass burning, and consider 50% reductions. We agree with the reviewer that actions to control these NMVOC emissions will likely also affect other co-emitted species. However, for anthropogenic emissions from industry, transportation, etc., many actions to reduce emissions will have different effects on NMVOCs relative to other co-emitted species. For example, cutting solvent use only reduces NMVOCs, and catalytic converters can have different effects on different species. In contrast, for biomass burning, the only (major) policy options would reduce the amount of burning and therefore the emissions of all co-emitted species by similar percentages. Because of this, previous work has addressed the effects of non-biomass burning anthropogenic emissions (Naik et al., 2005; Fry et al., 2013) separately from the effects of biomass burning emissions (Naik et al., 2007). Similarly, the HTAP study (Fiore et al., 2009) on which our previous work was based (Fry et al., 2012) simulated controls on anthropogenic emissions and excluded biomass burning from the definition of anthropogenic.

In the text, we have strengthened the rationale for focusing on non-biomass burning anthropogenic emissions on Page 21129 Lines 24-26, and Page 21130, Lines 1-2:

> Anthropogenic emissions include all anthropogenic sectors except biomass burning emissions (Figure S2), which are excluded since actions to address biomass burning (i.e. reductions in burning) differ from the other anthropogenic sectors (Naik et al., 2007), and would likely reduce the emissions of all co-emitted species by similar percentages. Actions to reduce anthropogenic emissions from industrial sources, on the other hand, likely have differing effects on NMVOCs relative to other co-emitted species.

In regards to the total anthropogenic RF of NMVOCs that we estimate in the paper, it is mainly for the purpose of comparing with other studies, as more studies have estimated the total anthropogenic RF than have estimated regional GWPs. To facilitate comparison, we scale the normalized net RF (net RF per Tg NMVOC) (Table 4) by the total anthropogenic (including biomass burning) NMVOC emissions that we have simulated. However, there are two major uncertainties involved in our method of scaling: 1) the spatial distribution of biomass burning NMVOC emissions is not the same as other NMVOC emissions, and 2) the species of NMVOC emissions are not the same.

To address the first of these uncertainties and improve upon our estimate of the present-day total anthropogenic NMVOC RF, we have modified the calculation to account for the geographic variability of biomass burning NMVOC emissions. We now present a global net RF (across regions) of 0.0374 W m^{-2}, calculated as the sum of the normalized global annual net RF estimates (Table 4) multiplied by total anthropogenic NMVOC emissions (including biomass burning) in each region (Table S2). We believe this is an improvement, as we now account for the geographic variability in biomass burning emissions, yet we acknowledge that we do not address the second uncertainty concerning the speciation of NMVOCs, as the same speciation is assumed.

As discussed above in response to Reviewer #1's comments, we have deemphasized this estimated total net RF, by removing it from the abstract and summary. We removed the sentence on Page 21126, Lines 18-20:

> The total global net RF for NMVOCs is estimated as 0.0277 W m^{-2} (~1.8 % of CO_2 RF since the preindustrial).

and we have replaced the sentence on Page 21139, Lines 9-10 with:

> The global annual average net RF for the global 50% NMVOC reduction is estimated as -9.73 mW m^{-2} or 0.21 mW m^{-2} (Tg C yr^{-1})$^{-1}$.

We also have revised Page 21135, Lines 22-27 and Page 21136, Lines 1-8:

> The global annual average net RF is estimated as -9.73 mW m^{-2} for the global 50% NMVOC reduction or 0.21 mW m^{-2} (Tg C yr^{-1})$^{-1}$ (Table 4). This estimate for the global NMVOC reduction differs somewhat from the sum of the 10 regions' net RF estimates (12.5 mW m^{-2}) (Table 4), suggesting some level of error in adding the ten regions' net RF impacts to get a global total net RF. However, in order to compare with other estimates of anthropogenic forcing, we estimate the global net RF by multiplying each region's global net RF per unit emissions (Table 4) by total anthropogenic emissions including biomass burning, which were excluded in the 50% reductions (Table S2). We then sum the ten regions' global net RF estimates to yield 0.0374 W m^{-2} as the net RF of anthropogenic NMVOCs.
>
> This approach accounts for the geographic variability of biomass burning emissions, yet assumes the same mixture of NMVOC species as anthropogenic emissions. The influence of climate change from the preindustrial to present day is also omitted, as all simulations use 2005 meteorology. While this estimate of total net RF is derived using methods that differ from the ACCMIP and IPCC AR5 standard RF definitions, which are present-day relative to the year 1850 and 1750, respectively, it provides an opportunity to compare with other studies. This RF is ~66% of the ACCMIP multimodel mean global net RF of NMVOC emissions for 1850-2000 due to O_3 and CH_4 changes alone (0.057 W m^{-2}) (Stevenson et al., 2013), and 15 to 18% of previous CO + NMVOC RF estimates: 0.25 ±0.04 W m^{-2} (Shindell et al., 2009) and 0.21 ±0.1 W m^{-2} (Shindell et al., 2005; and Forster et al., 2007). The RF of anthropogenic NMVOCs is ~2.4% of global net RF of CO_2 (1.56 W m^{-2}), and among the positive short-lived forcing agents (CO, CH_4, NMVOCs, and BC), ~2.4% of their total RF (1.57 W m^{-2}) (Forster et al., 2007).