

## Net radiative forcing and air quality responses to regional CO emission reductions

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### Response to Anonymous Referee #2:

We thank Referee #2 for the thoughtful feedback and suggestions for improving this manuscript. 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 italics.

*This manuscript describes a detailed sensitivity study related to CO emission reduction. This kind of study can contribute significantly to understanding behaviors and climate impacts of anthropogenic emissions and their changes, and can give a nice input to the policy relevant issues like co-benefits strategy in terms of global warming and air pollution mitigation (e.g., Shindell et al., 2012). The authors examined detailed source/receptor relationship by performing a sensitivity simulation with respect to regional CO emission reduction. This point can also be regarded as an advantage of the study. I'm, however, a bit concerned about practical feasibility of reducing solely CO emission apart from other emissions such as BC/OC. I also suspect that the authors adopted method to estimate CH<sub>4</sub> concentration may not be suitable.*

*Apart from the above described points, this study can be regarded as a significant addition to the current knowledge on the atmospheric chemistry and emission control strategies. The overall text is competently and clearly written, and reference to related previous studies is appropriate and adequate, several sentences seem to be tediously written, though. Their method to calculate RF/GWP seems to be well organized and systematic. However, their calculated minor values of concentrations and RF for regional CO emission reduction may not be that significant in comparison with the overall/global climate change tendency.*

*The subject of this paper appears to be appropriate to the ACP. However, I would like the authors to consider my questions and revise the manuscript before I recommend the publication of this paper. Details of my comments will be found in the following.*

Thank you for your helpful comments. We have revised the manuscript to better address our study design and motivation for evaluating regional CO emission reductions. We also have added to our discussions of the resulting concentration changes and RFs. More specifically, we respond to the four main comments:

- 1) "Practical feasibility of reducing solely CO" is addressed below where the Referee expands on this idea.
- 2) "Adopted method to estimate CH<sub>4</sub> concentration" is clarified below.
- 3) "Sentences [that] seem to be tediously written" - we have reviewed the entire paper and simplified the wording in several places, so that the ideas are communicated more clearly.
- 4) "Minor values of concentrations and RF for regional CO emission reductions" have been put into context and expanded upon. We have added the following to Page 33461, Line 22 to address the influence of anthropogenic CO emissions on global annual average surface O<sub>3</sub>: Anthropogenic CO emissions overall contribute ~6.1% (1.6 ppbv) to global annual average steady-state surface O<sub>3</sub>, by doubling the change from the 50% global CO reduction (-0.45 ppbv) and scaling for biomass burning emissions.

Our global net RF of CO also has been revised to 0.128 W m<sup>-2</sup>, which is higher than the 0.072 W m<sup>-2</sup> estimate in the previous version of the manuscript, on Page 33458, Lines 26-27:

By doubling the global annual average net RF of the 50% global CO reduction ( $-0.0361 \text{ W m}^{-2}$ ) and scaling for biomass burning emissions (43.6% of global anthropogenic CO emissions), which were excluded in the 50% anthropogenic CO emissions reductions, the global net RF of CO is  $0.128 \text{ W m}^{-2}$ .

In addition, we compare our CO RF estimate ( $0.128 \text{ W m}^{-2}$ ) to the RF of CO+NMVOCs from Shindell et al., (2005; 2009) and Forster et al. (2007). We have also added a comparison of our estimate to Stevenson et al. (2013). To clarify, we revised the following on Page 33459, Lines 1-6:

This is only ~11% greater than the ACCMIP multimodel mean global net RF of CO emissions due to  $\text{O}_3$  and  $\text{CH}_4$  changes alone ( $0.115 \text{ W m}^{-2}$ ) (for 1850-2000) (Stevenson et al., 2013). It is smaller than the RF of CO + NMVOC emissions in previous studies:  $0.21 \pm 0.10 \text{ W m}^{-2}$  (Shindell et al., 2005; Forster et al., 2007) and  $0.25 \pm 0.04 \text{ W m}^{-2}$  (Shindell et al., 2009), and is approximately 8.2% of the global net RF of  $\text{CO}_2$  ( $1.56 \text{ W m}^{-2}$ ). Among the positive forcing agents with short lifetimes (CO,  $\text{CH}_4$ , NMVOCs, and BC), our estimated CO RF is ~8.2% of their total RF ( $\sim 1.57 \text{ W m}^{-2}$ ) (Forster et al., 2007).

On Page 33460, Line 17 of the Conclusions, we also discuss how our present-day CO RF compares to that from  $\text{CO}_2$  and short-lived forcing agents:

The present-day CO RF is estimated as 8.2% of that from  $\text{CO}_2$ , and also 8.2% of the short-lived forcing agents that provide an opportunity to slow climate change in the coming decades.

### **Major Comments**

*\*\* The main subject of this manuscript is to assess the impacts and validities of CO emission reduction. I, however, have to say that there is a big question on how it is realistic and feasible to reduce emission for CO only apart from other components. As the authors state in "introduction", CO is emitted from incomplete combustion of carbon fuels, and should not be independent from emissions of BC/OC. Perhaps it may be also related to emissions of VOCs, NOx/SOx in terms of energy sector. I feel the authors should clarify this point and describe how their results should be interpreted in such context (linkage to other emissions). Please add discussions in the Introduction and Conclusion sections.*

We agree that sector-specific measures to reduce CO emissions would also affect co-emitted species (e.g. BC, OC). However, we do not aim to simulate the impacts of specific emission control measures as part of this analysis. We instead examine the sensitivity of air quality and RF to the location of CO emissions. For determining the net effect of emission control measures affecting multiple pollutants, our results should be interpreted as additive to those for co-emitted species. By simulating the RF impacts of CO emissions alone, we also can estimate the GWP of CO. This approach is in line with that of the IPCC to estimate the RF and GWPs for emissions of different species, and is in line with GHG emission inventories that track individual species.

To clarify, we have added the following to the Introduction, Page 33445, Line 13:

We omit reductions in co-emitted species (e.g. black carbon [BC], organic carbon [OC]) that would be affected by measures to reduce CO emissions, to examine the sensitivity of air quality and RF to CO emissions alone, and to derive CO climate metrics. Future studies may model emission control measures that address multiple species (e.g., Shindell et al 2012), or combine

these results with those for co-emitted pollutants to determine the net effect of emission control measures.

We also have added the following to the Conclusions, Page 33460, Line 21:

While emission control measures would likely affect co-emitted species (e.g. BC, OC), this study focuses on the sensitivity of air quality and RF to the location of CO emissions, which is also relevant for determining the GWP of CO. For measures affecting multiple pollutants, the results reported here can be combined with those for co-emitted pollutants.

**\*\* Page 33448 "2.1 Chemical transport modeling":**

*I found some important information with the model is missing. The authors should show more of configuration of the model. How and how much does it include natural emissions (biogenic/ocean and lightning NO<sub>x</sub>) ? How the stratosphere is treated in the model? Does it simulate full stratospheric ozone chemistry? If yes, are the methane changes (CH<sub>4</sub>) due to CO reduction reflected on stratospheric ozone? For the global tropospheric ozone budget, the authors only discuss the changes/differences in the sensitivity simulations. Please add description on the global ozone budget for the base run. Plus, because the authors discuss the associated sulfate changes, global SO<sub>x</sub> budget in the model should appear in the main text.*

We have added more detail on simulated natural emissions to Page 33448, Line 13:

Biogenic emissions of isoprene and monoterpenes are calculated online in MOZART-4 using the Model of Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther et al., 2006), based on the methods of Pfister et al. (2008), where the simulated global annual isoprene and monoterpene emissions are 738 Tg yr<sup>-1</sup> and 107 Tg yr<sup>-1</sup>. All other natural emissions are taken from the Precursors of Ozone and their Effects in the Troposphere (POET) emissions inventory for the year 2000 (Olivier et al., 2003; Granier et al., 2005; Emmons et al., 2010).

We have also added the following on lightning and soil NO<sub>x</sub> emissions to Page 33448, Line 18:

The global annual mean lightning NO<sub>x</sub> and soil NO<sub>x</sub> emissions (for 2005) are estimated as 2.4 Tg N yr<sup>-1</sup> and 8.0 Tg N yr<sup>-1</sup>, respectively, which are within the range of other modeling studies (Schumann and Huntrieser, 2007; Hudman et al., 2012).

MOZART-4 does not simulate complete stratospheric ozone chemistry, so we have clarified by adding the following to Page 33449, Line 12:

Because MOZART-4 does not have complete stratospheric chemistry (Emmons et al., 2010), three-dimensional monthly mean stratospheric O<sub>3</sub> concentrations from the AC&C/SPARC O<sub>3</sub> database prepared for CMIP5 (Available: <http://pcmdi-cmip.llnl.gov/cmip5/forcing.html>) are merged with the calculated steady-state (short-term + long-term) tropospheric O<sub>3</sub> concentrations for each simulation including the base case. Søvde et al. (2011) estimated that ~15% of the RF of O<sub>3</sub> precursors is due to lower stratosphere O<sub>3</sub> changes, using a single model with both standard and updated chemistry. Since we do not account for lower stratospheric O<sub>3</sub> changes, our RF estimates may underestimate the full effect of CO emissions.

We have added the global O<sub>3</sub>, SO<sub>4</sub><sup>2-</sup>, NH<sub>4</sub>NO<sub>3</sub>, and SOA burdens from the base simulation to the caption of Table 3 (Page 33475):

Table 3. For the global and regional reduction simulations relative to the base, global annual mean burden changes in tropospheric and upper tropospheric (UT) steady-state O<sub>3</sub>, tropospheric CH<sub>4</sub>, SO<sub>4</sub><sup>2-</sup>, NH<sub>4</sub>NO<sub>3</sub>, and SOA. The total global annual average tropospheric O<sub>3</sub> (at steady state), SO<sub>4</sub><sup>2-</sup>, NH<sub>4</sub>NO<sub>3</sub>, and SOA burdens in the base simulation are 352 Tg O<sub>3</sub>, 1788 Gg SO<sub>4</sub><sup>2-</sup>, 457 Gg NH<sub>4</sub>NO<sub>3</sub>, and 237 Gg SOA.

We also added the global O<sub>3</sub> burden and chemical production and loss rates for the base simulation to the caption of Table 7 (Page 33479):

Table 7. Changes in global annual average (short-term) tropospheric O<sub>3</sub> burden (B<sub>O3</sub>), O<sub>3</sub> production (P<sub>O3</sub>), and net O<sub>3</sub> export (X<sub>O3</sub>) from each regional reduction, normalized per change in CO emissions (E<sub>CO</sub>), and the fractions of these above each reduction region and in the upper troposphere (UT). Regional O<sub>3</sub> lifetimes are also shown. For the base simulation, the total global annual average O<sub>3</sub> burden is 352.2 Tg O<sub>3</sub>, and the chemical production and loss rates are 4782.5 Tg yr<sup>-1</sup> and 3975.0 Tg yr<sup>-1</sup>.

*\*\* Page 33448 "Global CH<sub>4</sub> is set to a uniform mixing ratio of... Using our calculated parameters, we diagnose changes in global CH<sub>4</sub> burden for each perturbation. We then calculate long-term O<sub>3</sub> responses offline": I'm a little skeptical of this method. Actual CH<sub>4</sub> abundance in the troposphere should be determined as a result of interaction with O<sub>3</sub>-OH-CO chemistry, which would require an on-line CH<sub>4</sub> calculation. Why don't you use on-line CH<sub>4</sub> simulation in this instance? If you have some reasons for adopting the off-line CH<sub>4</sub> simulation, you should validate this approach in this paper.*

Since our perturbation simulations are only 1.5 years in length, we are unable to fully capture changes in CH<sub>4</sub> lifetime (with a lifetime of ~10 years) due to CO emission reductions. Instead, CH<sub>4</sub> is held fixed in each simulation, and we diagnose changes in global CH<sub>4</sub> abundance, a widely used method documented in several previous studies (Prather et al., 2001; West et al., 2007; Fiore et al., 2009; Fry et al., 2012). This also allows us to calculate long-term O<sub>3</sub> changes from global CH<sub>4</sub> changes, despite the short duration of our simulations.

To clarify this approach, we have revised the following on Page 33448 and Lines 22-29 and Page 33449 Lines 1-11:

Although the perturbation simulations are 1.5 years in length, we account for the influence of CO emissions on CH<sub>4</sub> (via OH), and thus long-term changes in O<sub>3</sub> on the decadal timescale of CH<sub>4</sub> perturbation lifetime, using methods from previous studies (Prather et al., 2001; West et al., 2007; Fiore et al., 2009; Fry et al., 2012). We set global CH<sub>4</sub> to a uniform mixing ratio in the base and perturbation simulations of 1783 parts per billion by volume (ppbv) (WMO, 2006). We then simulate an additional CH<sub>4</sub> control simulation, which reduces global CH<sub>4</sub> by 20% to 1426.4 ppbv. Using the results from the base and CH<sub>4</sub> control simulations, we estimate CH<sub>4</sub> lifetime against loss by tropospheric OH ( $\tau_{\text{OH}}$ , 11.24 years), total CH<sub>4</sub> lifetime based on  $\tau_{\text{OH}}$  and CH<sub>4</sub> loss to soils and the stratosphere ( $\tau_{\text{total}}$ , 9.66 years), and methane's feedback factor (F, 1.29) by the methods of Prather et al. (2001), finding that our  $\tau_{\text{OH}}$  is slightly higher than those estimated by Fiore et al. (2008) (10.3 years) and Emmons et al. (2010) (10.5 years), but within the range of other models: 8.2 to 11.7 years (Stevenson et al., 2006), 10.2 ± 1.7 years (Fiore et al., 2009), and 9.8 ± 1.6 years (Voulgarakis et al., 2012). Prather et al. (2012) also estimated  $\tau_{\text{OH}}$  as 11.2 ± 1.3 years based on observations. Using the offline formulation of West et al. (2007) and our

estimated parameters ( $\tau_{OH}$ ,  $\tau_{total}$ , and  $F$ ), we estimate the steady-state tropospheric  $CH_4$  change for each perturbation. We then calculate long-term  $O_3$  responses by scaling the change in  $O_3$  from the  $CH_4$  control simulation ( $CH_4$  control simulation minus base) in each grid cell by the ratio of global  $CH_4$  burden change calculated for each perturbation to that of the  $CH_4$  control simulation. Long-term  $O_3$  responses are added to the short-term  $O_3$  responses simulated directly for each CO emission reduction (described below) to yield  $O_3$  concentrations at steady state (West et al., 2007; Fiore et al., 2009; Fry et al., 2012).

**\*\* Page 33450 "2.2 MOZART-4 evaluation":**

*As the authors realized, I also found severe overestimates of surface CO by the model at several sites (especially in SH). Aren't those discrepancies attributed to too low OH levels in the model which seem consistent with the lower OH burden relative to Spivakovsky et al. (2000)? The authors should clarify this point in the text. Also, please add some discussions on how the CO biases of the model can affect the later discussions in section 3.*

We have added the following to address the overestimate of CO in the SH, Page 33451, Line 3:  
Low modeled OH concentrations may contribute in part to the surface CO overestimates of CMDL measurements in the SH.

We also added this sentence to Page 33452, Line 19:

Given that modeled OH concentrations are lower than a previous study (Spivakovsky et al., 2000), simulated surface CO concentrations may be slightly overestimated, such as in the SH when compared to CMDL measurements.

**\*\* Page 33452 "BC and OC concentrations are not evaluated further as changes in these species between the base and perturbed simulations are negligible":** *But BC/OC emission could be changeable in accordance with CO emission change in the real world. Is it really proper to consider CO change only? I would like to know the authors intension.*

Our discussion of this issue is included above, under the first response to Referee #2's Major Comments. We added two new statements to the Introduction, Page 33445, Line 13 and Conclusions, Page 33460, Line 21, as noted above.

**\*\* "3 Global and regional air quality responses":**

*The authors discuss impacts of CO reduction on climate and air quality using the results of the sensitivity simulations. Their discussions seem quite detailed and adequate for showing what they obtained in their simulations. However, in fact, climate feedbacks to  $O_3$ -OH- $CH_4$  chemistry should be taken into account. At least the authors should describe how climate change caused by the CO reduction can affect the present discussions in the manuscript. This point seem practically important since the simulated differences in  $O_3/CH_4/SO_4$  in response to CO reduction appear so small/subtle that those are easily subject to change by other factors like meteorology.*

*Particularly, this study ignores indirect effects of aerosols which may largely alter the eventual discussion and conclusions in this paper. Could you include any description on this point in the*

*text? In the simplest way, the authors may very roughly estimate additional changes in RF which would be expected with aerosol indirect effects, using the IPCC's model estimate.*

*In the sensitivity simulation, emissions for species other than CO are kept constant in this study. However, actual emissions should be changing even in near-term future as in most of the IIASA/RCP scenarios. For example, considerable reduction in SO<sub>2</sub> emissions is assumed in each RCP scenario in 2030 or 2050 relative to 2005. Please add some discussions on this point to interpret your results in the context of future emission scenarios.*

Addressing climate feedbacks to O<sub>3</sub>-OH-CH<sub>4</sub> chemistry is beyond the scope of the study, and is not possible using our methods and modeling tools: MOZART-4 (for 1.5 year simulations) and standalone RTM. We also are not able to quantify the contribution of the aerosol indirect effect with the current version of the standalone RTM. Since many different types of responses are possible when modeling effects via atmospheric dynamics, we decided to limit our discussion of the relative importance of the different effects. We have added the following statement on Page 33451, Line 19-20 to briefly mention aerosol indirect effects:

The RTM simulations do not include the indirect effects of aerosols on clouds or the internal mixing of aerosols. Aerosol indirect effects are highly uncertain, and may account for considerable RF contributions beyond the direct effect of aerosols (Forster et al., 2007).

While we focus on present-day (year 2005) CO emissions in this study and do not evaluate climate feedbacks, we added the following to Page 33463, Line 2:

Future work could examine the influence of dynamic climate feedbacks on chemistry, and future changes in emissions that may alter the air quality and RF sensitivities estimated here for present-day emissions.

### ***Minor Comments***

***\*\* Fig.2: In the global chart, could you add a figure (actual number) for each emission sector?***

Given the small size of some of the sectors' emissions contributions in each region, we decided to instead provide the anthropogenic and biomass burning CO emissions quantities by region and sector in a new supplemental table, Table S11.

***\*\* Fig.3-7: Is it possible to show a global mean value in each regional panel?***

Yes, we have added the global annual mean concentration changes to each panel in Figures 3 through 6 (now Figures 3, 5, 6, 7). For Figure 7 (now Figure 8), we have added the global annual average net RF to each panel.

***\*\* Fig. S10: I like this kind of relationship shown in the main manuscript not as the supplementary. Is it possible to include it?***

Yes, we have moved Figure S10 to Figure 4 of the main manuscript. All subsequent figures have been re-numbered.