

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

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### **Response to Anonymous Referee #1:**

We thank Referee #1 for 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 italics.

*Fry et al. present the air quality and radiative forcing effects of halving regional CO emissions, exploring the impacts on CH<sub>4</sub>, O<sub>3</sub>, and SO<sub>4</sub> aerosols. The net result is an annual global mean net radiative forcing of 36.1mWm<sup>-2</sup> and modest changes to surface ozone and aerosol concentrations. A global halving of CO emissions has comparable effect to the sum of individual regional changes. The global warming potential of such reductions in CO is also found to be relatively independent of source region and somewhat smaller magnitude than previous studies. The manuscript is well-written and uses a straight-forward method of modeling to estimate the radiative forcing and air quality effects of CO. I have only minor concerns and comments. I recommend that this manuscript be published after the authors address the comments below.*

Thank you. Your comments and suggestions have been very helpful in improving this manuscript.

### **General Comments**

*- The authors provide a brief evaluation of the model, with much greater detail buried in the supplemental section. As the paper currently reads, I think the authors are not adequately describing the model's successes and failures. For example, "average bias of 4.5 ppbv O<sub>3</sub> across all sites compared to CASTNET." But Figure S1 shows that some sites have a bias of almost 20 ppb in the summer months, which are when ozone and sulfate are at their maximum and their perturbation may be most important.*

We have improved our description of the model's performance on Page 33450, Lines 12-16:

The base simulation produces an average bias of 4.5 ppbv O<sub>3</sub> across all sites compared to the Clean Air Status and Trends Network (CASTNET) (Figure S1), and 0.8 ppbv O<sub>3</sub> compared to the European Monitoring and Evaluation Programme (EMEP) network (Figure S2). MOZART-4 performs the least well during the summer months (June to August) in the US, with biases of nearly 20 ppbv in the Great Lakes, North East US, and South East US regions.

*- In order for this work to be reproducible and fully compared to other models, it is important to list and quantify the relevant "natural" and unperturbed anthropogenic emissions that additionally affect oxidant levels. For example, the authors should include such quantities as lightning NO<sub>x</sub>, which strongly influences tropospheric ozone (especially in the tropics, a region of focus of this work).*

We have added lightning and soil NO<sub>x</sub> quantities 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).

We also added new references to Hudman et al. (2012); Schumann and Huntrieser (2007):  
Hudman, R. C., N. E. Moore, A. K. Mebust, R. V. Martin, A. R. Russell, L. C. Valin, and R. C. Cohen (2012), Steps towards a mechanistic model of global soil nitric oxide emissions: implementation and space based-constraints, *Atmos. Chem. Phys.*, 12, 7779-7795, doi:10.5194/acp-12-7779-2012.

Schumann, U. and H. Huntrieser (2007), The global lightning-induced nitrogen oxides source, *Atmos. Chem. Phys.*, 7, 3823-3907.

In addition, we revised the following on Page 33448, Lines 13-18 to include isoprene and monoterpene emission quantities:

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); global annual isoprene and monoterpene emissions are estimated as 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).

The tropospheric OH burdens by latitude band and altitude are also provided in Table S1, which can be compared to other modeling simulations. Estimated CH<sub>4</sub> loss by reaction with OH (%) is presented in Table S2, as an additional metric for comparison to other models. All anthropogenic emissions are from the CMIP5 RCP8.5 emissions inventory for the year 2005 (Available: <https://tntcat.iiasa.ac.at:8743/RcpDb/dsd?Action=htmlpage&page=welcome>).

*- The authors should at least briefly mention the seasonality of these effects, which are shown in Supplemental Figure 11 for O<sub>3</sub>. I would imagine that seasonal changes in air quality and radiative forcing, while still small, are larger in magnitude and perhaps more relevant.*

On Page 33454, Lines 12-15, we mention the seasonality of O<sub>3</sub> air quality effects (Figure S11) in the following sentence:

As in Fiore et al. (2009), for NH sources and receptors, the greatest transport and intercontinental influences of each region on other regions are mainly from March to June (Figure S11).

We have added a new supplemental figure (Figure S20) showing the monthly global net RF estimates for each of the regional reduction simulations. In addition, we have added the following sentence to Page 33458, Line 26:

Monthly global net RF estimates also vary from ~56% less to ~34% greater than the annual mean, with the greatest RFs from June to September (Figure S20).

*- Seemingly significant changes occur to NH<sub>4</sub>NO<sub>3</sub> and SOA, but little discussion is presented and these species are neglected in the radiative forcing calculation. The authors, at the very least, should add remarks as to the relevance or irrelevance of these species in the ultimate radiative*

*calculations. This is somewhat dealt with in the Conclusions, but could be further elaborated upon and mentioned in Sections 3 and 5.*

We have revised the following sentence in Section 3, Page 33455, Line 24 to mention the magnitude of tropospheric  $\text{NH}_4\text{NO}_3$  and SOA changes:

Global annual average tropospheric  $\text{NH}_4\text{NO}_3$  and SOA changes are likewise small: -21% and 61%, respectively, of tropospheric  $\text{SO}_4^{2-}$  changes, for the global CO reduction.

We also added the following to address the relevance of  $\text{NH}_4\text{NO}_3$  and SOA to the RF calculations in Section 5, Page 33458, Line 9:

While changes in  $\text{NH}_4\text{NO}_3$  and SOA are not accounted for by the RTM, they would likely enhance or offset the localized to regional cooling and warming effects of  $\text{SO}_4^{2-}$ .

### ***Specific Comments***

*Page 33445, Lines 18-20: It is interesting that the uncertainty decreases as more components are added into the calculation. This issue is outside the control of the authors, but it stands out.*

Yes, this is a good observation. These RF estimates were derived in different studies (Shindell et al., 2005; 2009), so the uncertainties may not be directly comparable.

*Page 33454, Lines 3-4: Doesn't this contradict the statement in Page 33453 Lines 14-15?*

On Page 33453, Lines 14-15, we state that the long-term surface  $\text{O}_3$  changes (per unit CO emissions) vary little among regions, while on Page 33454, Lines 3-4, we discuss steady-state surface  $\text{O}_3$  changes, which are the sum of short-term and long-term changes. To clarify, we have revised Page 33453, Lines 9-10:

Global  $\text{CH}_4$  changes are used to calculate long-term tropospheric  $\text{O}_3$  changes, which vary little among regions, that are then added to short-term changes to give steady-state  $\text{O}_3$  responses (Table 4).

*Page 33454, Lines 7-8: 93% is very large, has such an influence been previously noted?*

Fiore et al. (2009) found annual mean surface  $\text{O}_3$  responses to foreign emissions to be as large as ~50% of the response to domestic emissions, such as for East Asia CO emissions on surface  $\text{O}_3$  in North America and Europe. East Asia is the largest CO emitting region that we consider. A 50% reduction in East Asia emissions is much greater than a 50% reduction in North America emissions. East Asia is also upwind of North America, and CO has a mean lifetime of 1 to 3 months (exceeding the time it takes for intercontinental transport), suggesting that large influences of East Asia CO emissions on US surface  $\text{O}_3$  are possible. We have added the following for comparison to Fiore et al. (2009) on Page 33454, Line 8:

Fiore et al. (2009) also found that East Asia CO emissions can influence surface  $\text{O}_3$  in North America by as much as 50% of the response from domestic emissions.

*Page 33455: Additional discussion regarding nitrate and SOA is warranted. Looking at Table 3, the change in SOA is almost as large as  $\text{SO}_4$ . Also, do changes in methane affect aerosols?*

We decided to exclude additional supporting figures and tables showing  $\text{NH}_4\text{NO}_3$  and SOA surface concentration changes and their distributions because the overall  $\text{PM}_{2.5}$  concentration changes (shown in Figure S16) are small. Discussion of these aerosols is included on Page

33455, Lines 2-10, and now on Page 33455, Line 24 (as noted in previous response above). We have also added an additional row to Table 3, to show the changes in  $\text{SO}_4^{2-}$ ,  $\text{NH}_4\text{NO}_3$ , and SOA for the -20% global  $\text{CH}_4$  control simulation (relative to the base simulation).

*Page 33455: The changes in aerosols for a global change in CO listed Table 3 differ from the sum of the regional changes.*

Yes, the sum of the global, annual mean tropospheric aerosol changes from the regional reduction simulations differ from the changes for the global CO reduction. This is in contrast to the  $\text{O}_3$  results. We agree that this is good to mention, so we have added the following to Page 33455, Line 6:

The sums of global  $\text{SO}_4^{2-}$ ,  $\text{NH}_4\text{NO}_3$ , and SOA burden changes for all 10 regional reductions are 0-8% less than those of the global CO reduction, suggesting some dependence on regional conditions and chemistry.

*Page 33459, Lines 1-6: Why is there such a difference?  $0.2 \text{ W m}^{-2}$  vs.  $0.07 \text{ W m}^{-2}$*

Our global net RF of CO has been revised to  $0.128 \text{ W m}^{-2}$ , which is higher than the  $0.072 \text{ W m}^{-2}$  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). Therefore, much of the difference is due to our RF not including NMVOCs. 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).

*Page 33463: Lines 3-5: I am left with the impression that altering CO does not greatly affect air quality or anthropogenic climate forcing. Can the authors add some perspective (1 or 2 sentences) before mentioning international agreements?*

On Page 33460, Lines 17-19 of the Conclusions, we 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.

We have also added the following to Page 33461, Line 22 to address the influence of anthropogenic CO emissions on global annual average surface  $\text{O}_3$ :

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.

*Table 3: I do not understand the units of Tg/yr and Gg/yr since the text and caption presents these quantities as strict changes in burden.*

We have revised the units to Tg and Gg in Table 3, since they represent annual average burden changes.

*All figures with multiple maps: The maps are small, and details, especially at the regional scale, are hard to make out. Please make sure the figures are large and clear in final version. Especially Figure 7.*

The figures with multiple maps will be provided in .eps format, which will allow them to be made larger and clearer in the final version of the paper.

#### Additional Edits:

We updated the References section by re-ordering a few references based on alphabetical order, and adding three new references:

Kopacz, M., D. J. Jacob, D. K. Henze, C. L. Heald, D. G. Streets, and Q. Zhang (2009), Comparison of adjoint and analytical Bayesian inversion methods for constraining Asian sources of carbon monoxide using satellite (MOPITT) measurements of CO columns, *J. Geophys. Res.*, 114, D04305, doi:10.1029/2007JD009264.

Kopacz, M., D. J. Jacob, J. A. Fisher, J. A. Logan, L. Zhang, I. A. Megretskaya, R. M. Yantosca, K. Singh, D. K. Henze, J. P. Burrows, M. Buchwitz, I. Khlystova, W. W. McMillan, J. C. Gille, D. P. Edwards, A. Eldering, V. Thouret, P. Nedelec (2010), Global estimates of CO sources with high resolution by adjoint inversion of multiple satellite datasets (MOPITT, AIRS, SCIAMACHY, TES), *Atmos. Chem. Phys.*, 10, 855-876.

Stevenson, D. S., P. J. Young, V. Naik, J.-F. Lamarque, D. T. Shindell, A. Voulgarakis, R. B. Skeie, S. B. Dalsoren, G. Myhre, T. K. Berntsen, G. A. Folberth, S. T. Rumbold, W. J. Collins, I. A. MacKenzie, R. M. Doherty, G. Zeng, T. P. C. van Noije, A. Strunk, D. Bergmann, P. Cameron-Smith, D. A. Plummer, S. A. Strode, L. Horowitz, Y. H. Lee, S. Szopa, K. Sudo, T. Nagashima, B. Josse, I. Cionni, M. Righi, V. Eyring, A. Conley, K. W. Bowman, O. Wild, and A. Archibald (2013), Tropospheric ozone changes, radiative forcing and attribution to emissions in the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP), *Atmos. Chem. Phys.*, 13, 3063-3085, doi: 10.5194/acp-13-3063-2013.