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# Radiative forcing and climate metrics for ozone precursor emissions: the impact of multi-model averaging

C. R. MacIntosh, K. P. Shine, and W. J. Collins

Department of Meteorology, University of Reading, P.O. Box 243, Reading, RG6 6BB, UK

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Correspondence to: C. R. MacIntosh (c.r.macintosh@reading.ac.uk)

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## Abstract

Multi-model ensembles are frequently used to assess understanding of the response of ozone and methane lifetime to changes in emissions of ozone precursors such as  $\text{NO}_x$ , VOC and CO. When these ozone changes are used to calculate radiative forcing (RF) (and climate metrics such as the global warming potential (GWP) and global temperature potential (GTP)) there is a methodological choice, determined partly by the available computing resources, as to whether the mean ozone (and methane lifetime) changes are input to the radiation code, or whether each model's ozone and methane changes are used as input, with the average RF computed from the individual model RFs. We use data from the Task Force on Hemispheric Transport of Air Pollution Source-Receptor global chemical transport model ensemble to assess the impact of this choice for emission changes in 4 regions (East Asia, Europe, North America and South Asia).

We conclude that using the multi-model mean ozone and methane responses is accurate for calculating the mean RF, with differences up to 0.6% for CO, 0.7% for VOC and 2% for  $\text{NO}_x$ . Differences of up to 60% for  $\text{NO}_x$ , 7% for VOC and 3% for CO are introduced into the 20 year GWP as a result of the exponential decay terms, with similar values for the 20 years GTP.

However, estimates of the SD calculated from the ensemble-mean input fields (where the SD at each point on the model grid is added to or subtracted from the mean field) are almost always substantially larger in RF, GWP and GTP metrics than the true SD, and can be larger than the model range for short-lived ozone RF, and for the 20 and 100 year GWP and 100 year GTP. We find that the effect is generally most marked for the case of  $\text{NO}_x$  emissions, where the net effect is a smaller residual of terms of opposing signs. For example, the SD for the 20 year GWP is two to three times larger using the ensemble-mean fields than using the individual models to calculate the RF.

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opposite signs; therefore the RF, GWP and GTP are smaller residuals of two larger terms (e.g. Fuglestedt et al., 2010).

The dependence on the location of the emissions change (e.g. Collins et al., 2013) has its origins in the spatially-varying background chemistry, and in variations in insolation and temperature which also contribute to non-linearities in chemical behaviour. Model intercomparison studies which perturb, either individually or together, a number of ozone precursor species can provide a useful constraint on our understanding of the effect of changing emissions of SLCF species on tropospheric ozone. The Hemispheric Transport of Air Pollutants (HTAP) project perturbed individual ozone precursor species in different regions with a view to elucidating transport of short lived pollutants (Task Force on Hemispheric Transport of Air Pollution, 2010). Subsequent work by Fry et al. (2012) and Collins et al. (2013) assessed the RF, GWP and GTP for the precursor species. Computational limitations prevented the analysis of the variability in the RF, GWP and GTP using output from individual models in Fry et al. (2012); instead the ensemble-mean  $\pm\sigma$  fields of the ozone perturbation were used as a representative subset of the complete ensemble.

In the present work, we calculate the RF, GWP and GTP using output from each individual model in the HTAP ensemble. We then compare our results to those obtained with the model ensemble-mean ozone and methane  $\pm\sigma$  fields using the method of Fry et al. (2012). This approach assesses the extent to which the RF calculated with the mean ozone fields represents the mean of the RF calculated using the ozone fields from each model individually; it also elucidates whether the uncertainty in RF computed using the  $\pm\sigma$  ozone fields correctly represents the uncertainty that emerges from the calculations using each model individually.

Section 2 introduces the data used in this study, and describes the set up of the radiation code, Sect. 3 presents the initial precursor, ozone and methane fields, Sect. 4 discusses radiative forcing, Sect. 5 presents the metrics, and conclusions are given in Sect. 6.

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## 2 Methods

An ensemble of model runs from the HTAP study provides information on ozone and methane concentrations after a perturbation in an ozone precursor gas has been applied. Initial results and analysis are presented in Fiore et al. (2009). Here the resulting tropospheric ozone (together with calculated methane and ozone primary mode, PM) changes are used to calculate RF for each model and scenario using the Edwards–Slingo radiation code (Edwards and Slingo, 1996). The model ensemble mean and SD fields are calculated both before and after the RF calculations, for the individual ozone, methane and ozone PM fields.

As this study is concerned with understanding methodological uncertainty we do not account for the effects of the precursor emissions on stratospheric ozone or water vapour. In addition, the contribution of sulphate aerosol changes, as a result of the precursor emissions, which were included in the HTAP calculations, are not included here.

### 2.1 Models

The HTAP study perturbation scenarios reduced by 20% emissions of short-lived ozone precursor gases  $\text{NO}_x$ , CO and VOC in four different regions (North America, Europe, South Asia and East Asia), and a further run in which methane concentrations were perturbed globally. There are therefore 13 scenarios. Table 1 shows the HTAP nomenclature for the experiments, and the locations of the source regions. 11 chemistry transport models (CTMs) (see Table 2) produced results for these scenarios. For comparison with previous literature, the 11 models used in our study are the same as those used in Fry et al. (2012) and Collins et al. (2013) (Table 2).

Of the 11 CTMs used in this study, 9 use meteorological background fields from reanalyses to drive the model, while two (STOC-HadAM3-v01 and UM-CAM-v01) are coupled to global climate models (GCMs) and use 2001 sea ice and sea surface temperature data to drive the GCM. The models also use a variety of sources for the base-

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line emissions data, with the result that a 20 % decrease in emissions is not equivalent in mass terms between models. This serves to characterise the uncertainty due to emissions and model differences simultaneously; however, separation of the two effects is not possible here.

The model output is re-gridded to a common resolution of 2.75° latitude × 3.75° longitude, with 24 vertical levels, which is comparable to the resolution of the models on average. Many of the models do not fully resolve the stratosphere; therefore stratospheric changes in all species are neglected, and, above the tropopause, the models share a common climatology.

## 2.2 Radiation code

Model output is at monthly-mean resolution for the year 2001. For each model, January, April, July and October are used as input to the code, in order to reduce run-time constraints whilst remaining sufficient to resolve the annual cycle in transport and RF. Sensitivity tests have shown that the ozone PM and methane RFs are almost completely insensitive to increasing the number of months included (less than 1 part in 1000), and the short-lived ozone RFs have a sensitivity of the order of 0.5% to increasing the number of months. Table S4 provides a brief outline of the sensitivity tests.

The Edwards–Slingo radiation code uses the two stream approximation to calculate radiative transfer through the atmosphere. Clouds are included in the code. Nine broadband channels in the longwave and 6 channels in the shortwave are used. Incoming solar radiation at mid-month, and Gaussian integration over 6 intervals is used to simulate variation in the diurnal cycle.

A common background climatology supplying temperature and humidity are taken from the European Centre for Medium-Range Weather Forecasts reanalyses (Dee et al., 2011). Mean cloud properties from the International Satellite Cloud Climatology Project (ISCCP) are also used for all RF simulations (Rossow and Schiffer, 1999).

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tion for carbon dioxide, and the climate impulse-response function sensitivities from Boucher and Reddy (2008) which is needed for the GTP calculation. The effect of the uncertainties in these impulse response functions are not explored here. The metric calculations require the steady-state RF per unit emission per year, for each pre-cursor and for the short-lived ozone, PM ozone and methane changes individually. Again the technique for deriving these from the steady-state perturbations follows Fuglested et al. (2010). The resulting steady-state RFs for each model and each scenario are given in the Supplement. The nomenclature for the metrics follows that for the RF described in the previous subsection.

### 3 Ozone and methane input fields

The CTMs produce [OH], [O<sub>3</sub>] and associated atmospheric loss rates as 3-D output fields. Short-lived ozone can be used directly as input to the radiation code. Methane fields for each model and each simulation were globally homogeneous, and fixed at 1760 ppbv, except in the CH<sub>4</sub> scenario, when they are reduced to 1408 ppbv. Equilibrium methane concentrations for each scenario can be calculated from the change in methane lifetime,  $\Delta\alpha$ , as  $[\text{CH}_4] = 1760 \times \left(\frac{\alpha_{\text{control}} + \Delta\alpha}{\alpha_{\text{control}}}\right)^f$ , where the methane lifetimes are calculated from the CH<sub>4</sub> x OH fluxes (since the atmospheric OH sink accounts for around 90 % of loss of atmospheric CH<sub>4</sub>, and surface sinks are considered constant). The feedback factor,  $f$  is determined in Fiore et al. (2009) from the change in loss rates between the control and the CH<sub>4</sub> perturbation scenarios, and accounts for the effect of methane change on its own lifetime (Prather, 1996).

Ozone PM changes are dependent on the change in ozone resulting from a change in methane, which in turn depends on the change in methane lifetime for a given scenario. The PM changes for each model and scenario are calculated as described in West et al. (2009) by scaling the ozone change in the CH<sub>4</sub> perturbation simulation by the relative change in methane concentration as given in Fiore et al. (2009).

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change varies almost by a factor of 2, from 7.5 Tg for the MOZARTGFDL-v2 model to 16 Tg for LLNL-IMPACT-T5a, with associated RF of 25.4 and 54.9 mW m<sup>-2</sup> respectively.

The RF due to methane for a given scenario depends on the methane lifetime in the model, and the change in methane lifetime for the particular scenario. The VOC scenarios have the largest relative uncertainty in the methane changes ( $\sigma = 59$  to 104 % of the mean), including one model (GISS-PUCCINI-modelE) which has a small increase in methane due to a decrease in VOC in three of the regions (NA, EA and EU, Fig. 3).

The ozone PM RF depends on the scaling of the ozone PM mass change from the CH<sub>4</sub> perturbation scenario by the relative change in methane for each scenario, and therefore is also related to  $\Delta\alpha$  in addition to the model ozone response to a change in methane; hence it has a slightly larger relative uncertainty than the methane changes due to model differences in ozone PM response.

Short-lived ozone has the largest variability of the three components across the ensemble for any scenario, and the largest normalised variability for NO<sub>x</sub> and CO. For VOC, short-lived ozone is the largest single contributor to the total RF, whereas for NO<sub>x</sub> and CO, methane dominates. The SD in ozone response is between  $\pm 24\%$  in EA to  $\pm 42\%$  (SA) of the mean for NO<sub>x</sub>,  $\pm 58\%$  (SA) to  $\pm 77\%$  (NA) for VOC and  $\pm 27\%$  (SA) to  $\pm 37\%$  (NA) for CO (Table S1). A large relative short-lived ozone response to one precursor species is not a good indicator of the response to other perturbations. For a particular species, however, models with a large response in one region will tend to have a large response in all regions, i.e the models all agree on the order of the regional responses. These depend on the relative size of emissions change in each region and the mass-normalised RF. This is a good indicator of consistency across different emissions datasets and for transport in models, which information cannot be gained by using the model ensemble mean alone.

For NO<sub>x</sub>, there is a positive correlation between the size of the methane response and the short-lived ozone response, with  $r^2$  values between 0.56 (EA) to 0.86 (SA) (Table S1). For VOC and CO, the coefficients are much smaller (0.01 (NA) to 0.52 (SA) for VOC, and close to zero for all CO cases except EU). For NO<sub>x</sub>, there is significant

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correlation between the short-lived ozone and methane responses, even if the emission mass for each model is taken into account, with  $r^2$  values between 0.70 (EA) and 0.86 (NA and SA, Table S2). This suggests that the individual model chemistry may dominate over variability in emissions data.

#### 4.1 Ensemble-mean RF measures

Table 3 compares  $\overline{\text{RF}} \pm \sigma$  with the computationally much less intensive  $\text{RF}_{(\text{EN} \pm \sigma)}$ . Differences between the means are only of the order of a few percent, with the largest differences found for the  $\text{NO}_x$  EA case of 3%. For VOC and CO, the differences between the means does not exceed 0.7%, and for  $\text{CH}_4$ , the difference is negligible. The larger fractional difference in the case of  $\text{NO}_x$  is due to the fact that the means are a small residual of two much larger components. Hence  $\text{RF}_{(\text{EN} \pm \sigma)}$  is representative of the true ensemble mean,  $\overline{\text{RF}}$ . By contrast the SD in the  $\overline{\text{RF}} \pm \sigma$  case is smaller for every scenario relative to  $\text{RF}_{(\text{EN} \pm \sigma)}$ . This is largely associated with the inability of the pre-calculated ensemble mean fields to represent the true model spread, as described in Sect. 3.

Figure 4 separates the total RF into components due to the ozone PM, methane, and short-lived ozone contributions, for each scenario and gas, for the  $\text{RF}_{(\text{EN} \pm \sigma)}$  and  $\overline{\text{RF}} \pm \sigma$ .

In the  $\text{CH}_4$  perturbation case, the absolute methane RFs (red bars) have no uncertainty associated with intermodel differences because the methane concentration change is fixed. The RF calculated using the formula of Ramaswamy et al. (2001) is  $-139.6 \text{ mW m}^{-2}$  for  $\text{RF}_{(\text{EN})}$ , whereas the value calculated by the Edwards–Slingo radiation code for  $\overline{\text{RF}}$  is slightly more negative at  $-141 \text{ mW m}^{-2}$ . Because of the “back-calculation” method described in Sect. 2.2 this results in the estimate of the ozone PM  $\text{RF}_{(\text{EN})}$  being 5% too negative ( $-37.0 \text{ mW m}^{-2}$  vs.  $-35.3 \text{ mW m}^{-2}$ ). The SD of the  $\text{CH}_4$  scenario ozone PM  $\text{RF}_{(\text{EN})}$  is 33% of the mean, compared with 24% for the  $\overline{\text{RF}}$  case. As the  $\text{CH}_4$  scenario ozone PM RF is used to calculate the PM RF for each scenario for

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the EN case, and the PM RF is used to calculate the short-lived ozone RF, this small error will propagate through all the EN RF estimates. The effect is very small however, with the overestimate in short-lived ozone RF being between 1.6 and 2.4 % (NA and EU) in the NO<sub>x</sub> scenarios, between –0.02 and 1.1 % (NA, EU) for VOC, and 1.7 and 2.9 % (NA, SA respectively) for CO.

The relative sizes of the SDs for the methane, ozone PM and short-lived ozone RFs are mostly governed by the increased spread in the input fields as described in Sect. 3. For the methane RF in the NO<sub>x</sub>, VOC and CO scenarios, the SD  $RF_{(EN\pm\sigma)}$  is slightly smaller than the  $\overline{RF}$  SD in all regions, although the difference is less than 1.5 %. This is due to the smaller methane contribution in the  $RF_{(EN)}$  case, resulting in a smaller absolute SD. If the SD is normalised relative to the mean for each scenario, then the  $RF_{(EN)}$  SD is essentially identical to the  $\overline{RF}$  SD.

The ozone PM SD is about 30 % larger for the  $RF_{(EN)}$  case relative to the  $\overline{RF}$  case, reflecting its relatively greater spatial inhomogeneity as discussed in Sect. 3. The  $RF_{(EN)}$  SD is between 35–42 % of the mean (depending on the source region) for NO<sub>x</sub>, 33–106 % for VOC and 37–57 % for the CO scenarios. For  $\overline{RF}$  the values are 26–32 % for NO<sub>x</sub>, 62–96 % for VOC and 26–44 % for CO.

The short-lived ozone  $RF \pm \sigma$  shows the greatest difference between the two methods, again reflecting the difference in the input fields in Sect. 3. The largest absolute reductions, in SD occur for the NO<sub>x</sub> cases, since these are the most spatially inhomogeneous fields; the SD is 45–58 % and 24–42 % (EA, SA) of the mean for  $RF_{(EN)}$  and  $\overline{RF}$  respectively.

The relatively greater uncertainty surrounding the size of the VOC scenario response is reflected in the larger SDs, from 76–94 % and 58–77 % in the  $RF_{EN}$  and  $\overline{RF}$  cases. Finally for the CO cases, the SDs vary between 51–56 % and 27–37 % in the two cases. The difference in the short-lived ozone SDs is the major driving factor of the differences in the total  $RF_{(EN)}$  and  $\overline{RF}$  SDs in Table 3.



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larger than that for the  $\overline{\text{GWP}}$  (Table 4, Fig. 5, top row). Figure 5 also shows the SD for each scenario that is due to ozone PM, methane and short-lived ozone (second, third and bottom row, respectively). The  $\text{GWP}_{(\text{EN})}$  SD is larger than that for  $\overline{\text{GWP}}$ , for most components. The larger SD in the short-lived ozone  $\text{RF}_{(\text{EN})}$  relative to  $\overline{\text{RF}}$  is also apparent in the  $\text{GWP}$  values, and once again is the major driver of the larger uncertainty in the overall  $\text{GWP}_{(\text{EN})}$ .

Uncertainty in methane lifetime is only a minor contributor to the overall uncertainty in  $\text{GWP}_{(\text{EN})}$ . For example, in the  $\text{NO}_x$  NA case, the SD due to the short-lived ozone is  $\pm 44$ , due to methane RF is  $\pm 20$ , due to ozone PM is  $\pm 9$  and due to methane lifetime effects is  $\pm 5$ , with similar behaviour for other regions and species. There is an anti-correlation across the model ensemble between  $\alpha$  and  $f$  of  $-0.72$ , which is significant at the 95 % level. This means that the total methane lifetime,  $\tau$  is smaller than it would be were these quantities to be truly independent, as is therefore the variability attributed to  $\tau$ .

Table 5 gives the 100 year GWP. The magnitude of  $\text{GWP}_{(\text{EN})}$  is again larger for the  $\text{NO}_x$  and CO cases, and smaller for the  $\text{CH}_4$  and VOC cases (except SA), but the difference is much reduced relative to the 20 year GWP, not exceeding 8 % (VOC NA) of the total GWP.

In the case of the 100 year  $\text{GWP}_{(\text{EN})}$ , the long-term component is a simple scaling of the 20 year GWP by the ratio of the exponential terms and by  $\frac{100\text{year AGWP CO}_2}{20\text{year AGWP CO}_2}$ . The normalised  $\frac{\sigma}{\text{GWP}}$  is therefore identical at 20 and 100 year for the  $\text{CH}_4$ , ozone PM and short-lived ozone components, (and is also identical to the  $\frac{\sigma}{\text{RF}_{\text{EN}}}$ ). The total normalised SD for the  $\text{GWP}_{(\text{EN})}$  case is slightly smaller (26–35 % for CO, 59–70 % for VOC and 54–135 % for  $\text{NO}_x$ , relative to 28–36 %, 63–71 % and 93–992 % for the equivalent 20 year GWP values, regions as for 20 years). In contrast, for  $\overline{\text{GWP}}$ , the normalised SD is increased by 2 to 5 % in the 100 year GWP relative to the 20 year GWP for the VOC



and CO scenarios, as differences in the methane lifetime have a larger impact, although there is a substantial reduction in the  $\overline{\text{NO}_x}$  SD (Table S3).

For short-lived ozone, for both  $\overline{\text{GWP}}$  and  $\text{GWP}_{(\text{EN})}$  the scaling factor is simply  $\frac{100\text{year AGWP}_{\text{CO}_2}}{20\text{year AGWP}_{\text{CO}_2}}$ , and the normalised SDs are again identical to those for the 20 year  $\text{GWP}$ . The relative contribution of the short-lived ozone to the  $\text{GWP}$  is therefore slightly smaller at 100 years than at 20 years, being 38–59 % for CO, 97–146 % for VOC and 54–76 % for  $\text{NO}_x$ , relative to 47–73 %, 121–181 % and 67–96 % (all EU, SA) at 20 years. The consistency in the regional response across the scenarios and metrics suggests that the latitude of the emissions affects the relative importance of the short- and long-lived components.

For the 100 year  $\overline{\text{GWP}}$  components (not shown), the normalised uncertainty for the short-lived ozone is identical to that at 20 years, but the  $\text{CH}_4$  and ozone PM  $\frac{\sigma}{\text{GWP}}$  is slightly larger, due to the increased importance of the methane lifetime.

The smaller uncertainty associated with the total  $\overline{\text{GWP}} \text{NO}_x$  results in the values at 100 years being robustly negative, in contrast with the  $\text{GWP}_{(\text{EN})}$  case. This is primarily due to the reduced uncertainty for the short-lived ozone contribution, and the poor representation of uncertainty in RF in the ensemble-mean case.

## 5.2 Global temperature-change potentials

The 20 year  $\text{GTP}$  mean values (Table 6) share many characteristics with the  $\text{GWP}$  values, including the reduction in uncertainty of  $\overline{\text{GTP}}$  relative to  $\text{GTP}_{(\text{EN} \pm \sigma)}$  in most cases.

Figure 6 shows the total, short-lived ozone, methane and ozone PM components for the 20 year  $\text{GTP}_{(\text{EN})}$  and  $\overline{\text{GTP}}$ . One important difference relative to the 20 year  $\text{GWP}$  is that the  $\text{NO}_x$   $\text{GTP}$  is robustly negative in all cases, due to the much larger contribution of the methane component.

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The long-lived components of the 100 year GTP (Table 7) are related to the 20 years GTP in the same way that the 100 and 20 year GWPs are related. The long-lived component is a much larger fraction of the 20 year GTP than it is of the 100 year GTP, since 20 years is close to the methane response time ( $\sim 12$  years). This arises since the GTP is not a time integrated quantity, and the relative contribution of the components to the overall temperature change depends also on the climate response at its timescales.

### 5.3 Comparison of GWP and GTP time evolution for $\text{NO}_x$

Figure 7 shows the time evolution of the GWP (top) and GTP (bottom) for the  $\text{NO}_x$  SA region. Coloured lines show the evolution of each model, with the solid black line and dotted lines giving the true mean and SD. The dashed lines and grey shading give the  $\text{GWP}_{(\text{EN}\pm\sigma)}$ .

Models which have a longer methane lifetime have a steeper GWP gradient at 20 years than models with a short methane lifetime; however, this is not necessarily a good indicator of a more negative  $\text{NO}_x$  GWP at 20 years. Of the four longest lifetime models, three (CAMCHEM-3311m13, UM-CAM-v01 and MOZECH-v16) have GWP values that are more positive than the mean, with the fourth (GISS-PUCCINI-modeLE) lying well within one SD. This indicates that they also have a large short-lived ozone forcing.

$\overline{\text{GWP}}$  has its largest SD between 10 and 30 years, when both short-lived ozone and methane forcings are important. The  $\text{GWP}_{(\text{EN})}$  overestimates the true SD everywhere, but particularly around 10–30 years. At these timescales, the SDs produced in this way lie outside the range of the ensemble members, and therefore are not a good estimate of the uncertainty of the ensemble.

The GTP (lower panel in Fig. 7) does not have the same “memory” of early forcing as the GWP, so that the model spread decreases substantially after about 30 years. The separate effects of a long methane lifetime and a large short-lived ozone forcing can be more clearly seen here for UM-CAM-v01 (yellow line), which has a very neg-

ative minimum GTP value of less than  $-200$ , several years after the other ensemble members.

The largest uncertainty in the GTP is also around 20 years, when both the short-lived ozone, methane and ozone PM RF are important. Again, the  $GTP_{(EN)}$  substantially overestimates the uncertainty between 10 and 30 years. At times greater than about 35 years, however, the  $GTP_{(EN)}$  begins to agree better with the true GTP. The  $GTP_{(EN)}$  may even slightly underestimate the uncertainty at these longer times due to the slightly smaller methane RF estimate calculated in Sect. 4.

## 6 Discussion and conclusions

This study has investigated the derivation of RF and climate emission metrics (GWP and GTP at various time horizons) for emissions of short-lived climate forcing agents from multi-model assessments, using the results of the HTAP ozone precursor emission experiments as an example. Multi-model means and their associated SDs of the ozone perturbations can be used as input to radiative transfer codes, which is clearly more computationally efficient than calculating the radiative forcing for each model individually and averaging the results. Overall, our results indicate that the order of averaging does not have a major impact on the mean values. It does, however, have a larger impact on estimates of the uncertainties.

The global-mean RF from emissions of ozone precursors is only mildly sensitive to using the ensemble-mean input fields with differences in the mean not exceeding 3%. However, the SD of the RF is rather distinct between the two cases. The true SD (using the RF derived from each model individually) is always smaller than the SD when calculating the RF with the ensemble-mean ozone change. This effect is mostly due to the construction of the input ozone fields overestimating the true ensemble spread. In the case of the ozone PM, the  $RF_{(EN)}$  SD is about 30% larger than the true value. For the more spatially inhomogeneous short-lived ozone, the overestimate varies between 20% for the VOC EA scenario to 90% for the  $NO_x$  EA case.

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**Table 1.** HTAP ozone precursor anthropogenic reduction experiments. In the case of SR2, methane mixing ratios are reduced by 20%; for SR3–SR5 emissions of the precursor are reduced. The regions are defined as: North America (NA), 15–55° N, 60–125° W; South Asia (SA), 5–35° N, 50–95° E East Asia (EA), 15–50° N, 95–160° E; Europe (EU), 25–65° N, 10° W–50° E.

Experiment	Region	Description
SR1	Global	Control
SR2	Global	–20 % CH <sub>4</sub> reduction
SR3	NA, SA, EA, EU	–20 % NO <sub>x</sub> reduction
SR4	NA, SA, EA, EU	–20 % VOC reduction
SR5	NA, SA, EA, EU	–20 % CO reduction



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**Table 2.** Methane lifetime ( $\alpha$ ), feedback factor ( $f$ ), and the methane lifetime change due to a 20% global reduction in methane, for each of the 11 CTMs, and the ensemble mean and SD, as calculated in Fiore et al. (2009). Model abbreviations are explained in Fiore et al. (2009).

Model	Methane Lifetime $\alpha$ (years)	Feedback Factor $f$	Lifetime Change $\Delta\alpha_{\text{SR2}}$ (years)
CAMCHEM-3311m13	10.11	1.31	0.51
FRSGUCI-v01	7.72	1.43	0.50
GISS-PUCCINI-modelE	9.39	1.36	0.54
GMI-v02f	9.02	1.31	0.46
INCA-vSSz	8.75	1.31	0.45
LLNL-IMPACT-T5a	5.68	1.39	0.34
MOZARTGFDL-v2	9.06	1.31	0.47
MOZECH-v16	9.63	1.29	0.48
STOC-HadAM3-v01	8.20	1.31	0.42
TM5-JRC-cy2-ipcc-v1	7.98	1.43	0.51
UM-CAM-v01	10.57	1.25	0.45
Mean	8.73	1.33	0.47
SD	$\pm 1.34$	$\pm 0.06$	$\pm 0.05$

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**Table 3.** Total  $\text{RF} \pm \sigma$  ( $\text{mW m}^{-2}$ ) for each scenario. The SD values given for  $\text{RF}_{(\text{EN})}$  are the RF resulting from the mean and standard deviation ozone, methane and ozone PM fields, as described in Sect. 3. The true SD values for RF are calculated after the total RF for each model in each scenario has been calculated; therefore they are not equal to the sum of the SD for each component gas.

Scenario	type	NA		SA		EA		EU	
		mean	$\sigma$	mean	$\sigma$	mean	$\sigma$	mean	$\sigma$
$\text{NO}_x$ (SR3)	$\overline{\text{RF}}$	1.62	$\pm 2.43$	1.02	$\pm 1.76$	1.02	$\pm 1.58$	1.50	$\pm 1.12$
	$\text{RF}_{(\text{EN})}$	1.65	$\pm 3.03$	1.05	$\pm 2.13$	1.05	$\pm 2.27$	1.52	$\pm 1.52$
VOC (SR4)	$\overline{\text{RF}}$	-1.50	$\pm 1.28$	-1.16	$\pm 0.68$	-1.49	$\pm 1.11$	-1.82	$\pm 1.50$
	$\text{RF}_{(\text{EN})}$	-1.50	$\pm 1.41$	-1.15	$\pm 0.80$	-1.48	$\pm 1.25$	-1.82	$\pm 1.69$
CO (SR5)	$\overline{\text{RF}}$	-3.03	$\pm 1.22$	-2.52	$\pm 0.61$	-3.99	$\pm 1.61$	-2.24	$\pm 0.68$
	$\text{RF}_{(\text{EN})}$	-3.04	$\pm 1.46$	-2.53	$\pm 0.87$	-4.00	$\pm 1.98$	-2.24	$\pm 0.88$
Global									
$\text{CH}_4$ (SR2)	$\overline{\text{RF}}$	-176.6	$\pm 8.4$						
	$\text{RF}_{(\text{EN})}$	-176.5	$\pm 12.1$						

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**Table 4.** Ensemble-mean 20 year GWP. The true mean GWP and SD are denoted  $\overline{\text{GWP}} \pm \sigma$ . The GWP calculated using the method described in Fry et al. (2012) is denoted  $\text{GWP}_{(\text{EN} \pm \sigma)}$ . Average methane lifetimes used in the metric construction are given in Table 2.

Scenario	type	NA		SA		EA		EU	
		mean	$\sigma$	mean	$\sigma$	mean	$\sigma$	mean	$\sigma$
NO <sub>x</sub> (SR3)	$\overline{\text{GWP}}$	-9.76	$\pm 15.5$	-27.4	$\pm 34.1$	-2.64	$\pm 20.7$	-20.6	$\pm 7.85$
	$\text{GWP}_{(\text{EN})}$	-11.4	$\pm 41.2$	-30.1	$\pm 98.0$	-4.15	$\pm 41.2$	-21.5	$\pm 20.1$
VOC (SR4)	$\overline{\text{GWP}}$	17.6	$\pm 8.10$	21.2	$\pm 8.20$	16.9	$\pm 7.99$	17.2	$\pm 7.42$
	$\text{GWP}_{(\text{EN})}$	16.3	$\pm 11.7$	22.1	$\pm 13.9$	16.2	$\pm 10.5$	16.0	$\pm 10.6$
CO (SR5)	$\overline{\text{GWP}}$	5.22	$\pm 1.20$	5.59	$\pm 0.98$	5.27	$\pm 1.09$	4.99	$\pm 1.24$
	$\text{GWP}_{(\text{EN})}$	5.32	$\pm 1.86$	5.78	$\pm 1.63$	5.30	$\pm 1.94$	5.03	$\pm 1.47$
Global									
CH <sub>4</sub> (SR2)	$\overline{\text{GWP}}$	64.9	$\pm 4.17$						
	$\text{GWP}_{(\text{EN})}$	64.3	$\pm 5.18$						

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**Table 5.** As Table 4 for the 100 year GWP.

Scenario	type	NA mean	$\sigma$	SA mean	$\sigma$	EA mean	$\sigma$	EU mean	$\sigma$
NO <sub>x</sub> (SR3)	$\overline{\text{GWP}}$	-10.8	±4.77	-23.1	±9.83	-8.62	±6.58	-10.7	±2.67
	GWP <sub>(EN)</sub>	-11.2	±12.0	-23.7	±28.9	-8.75	±11.8	-10.9	±5.86
VOC (SR4)	$\overline{\text{GWP}}$	5.45	±2.54	6.62	±2.57	5.17	±2.54	5.40	±2.41
	GWP <sub>(EN)</sub>	5.04	±3.52	6.86	±4.06	4.94	±3.14	5.05	±3.33
CO (SR5)	$\overline{\text{GWP}}$	1.72	±0.42	1.82	±0.34	1.73	±0.38	1.66	±0.45
	GWP <sub>(EN)</sub>	1.74	±0.59	1.87	±0.49	1.76	±0.62	1.66	±0.47
Global									
CH <sub>4</sub> (SR2)	$\overline{\text{GWP}}$	23.0	±2.41						
	GWP <sub>(EN)</sub>	22.7	±1.56						

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**Table 6.** As Table 4 for the 20 years GTP.

Scenario	type	NA mean	$\sigma$	SA mean	$\sigma$	EA mean	$\sigma$	EU mean	$\sigma$
NO <sub>x</sub> (SR3)	$\overline{\text{GTP}}$	-62.8	±16.6	-122.1	±36.3	-59.3	±19.0	-42.8	±8.38
	GTP <sub>(EN)</sub>	-62.9	±19.1	-122.3	±46.8	-57.8	±17.1	-42.8	±9.5
VOC (SR4)	$\overline{\text{GTP}}$	8.98	±4.61	11.19	±4.31	7.99	±4.49	9.44	±4.68
	GTP <sub>(EN)</sub>	8.25	±5.57	11.54	±5.62	7.66	±4.80	8.93	±6.24
CO (SR5)	$\overline{\text{GTP}}$	3.39	±0.92	3.52	±0.70	3.43	±0.80	3.39	±0.97
	GTP <sub>(EN)</sub>	3.49	±1.16	3.62	±0.79	3.50	±1.21	3.42	±0.90
Global									
CH <sub>4</sub> (SR2)	$\overline{\text{GTP}}$	55.3	±5.49						
	GTP <sub>(EN)</sub>	54.8	±3.77						

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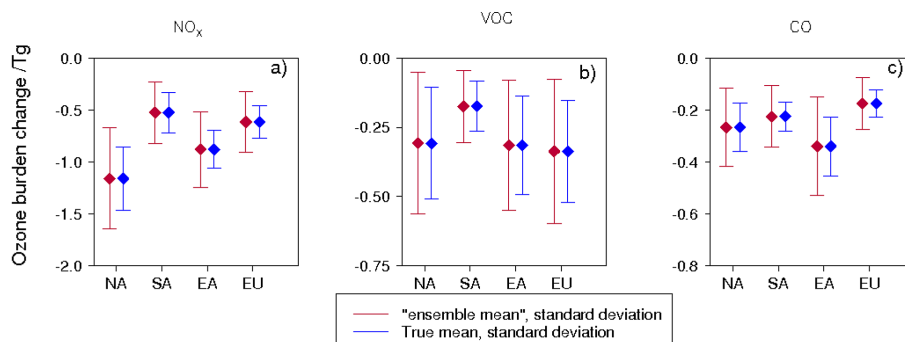
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**Table 7.** As Table 4 for the 100 year GTP.

Scenario	type	NA mean	$\sigma$	SA mean	$\sigma$	EA mean	$\sigma$	EU mean	$\sigma$
NO <sub>x</sub> (SR3)	$\overline{\text{GTP}}$	-2.20	±0.79	-4.53	±1.64	-1.87	±1.04	-1.92	±0.44
	GTP <sub>(EN)</sub>	-2.22	±1.75	-4.55	±4.23	-1.84	±1.71	-1.93	±0.86
VOC (SR4)	$\overline{\text{GTP}}$	0.81	±0.38	0.98	±0.38	0.76	±0.38	0.81	±0.37
	GTP <sub>(EN)</sub>	0.74	±0.51	1.01	±0.58	0.72	±0.46	0.75	±0.50
CO (SR5)	$\overline{\text{GTP}}$	0.26	±0.07	0.28	±0.05	0.26	±0.06	0.25	±0.07
	GTP <sub>(EN)</sub>	0.26	±0.09	0.28	±0.07	0.27	±0.09	0.25	±0.07
Global									
CH <sub>4</sub> (SR2)	$\overline{\text{GTP}}$	3.62	±0.45						
	GTP <sub>(EN)</sub>	3.55	±0.27						

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**Figure 1.** Change in global-mean atmospheric burden of short-lived ozone (in Tg), for **(a)** NO<sub>x</sub>, **(b)** VOC, and **(c)** CO for the emission changes and emission regions given in Table 1. The “ensemble mean” and SD fields (red lines) are constructed by calculating the mean and SD of the model ensemble at each grid point.

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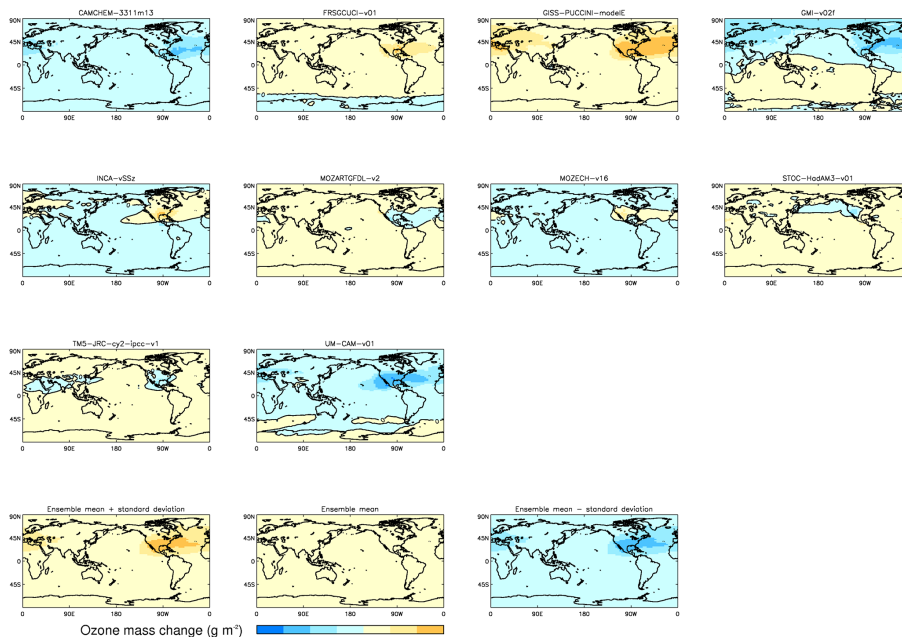
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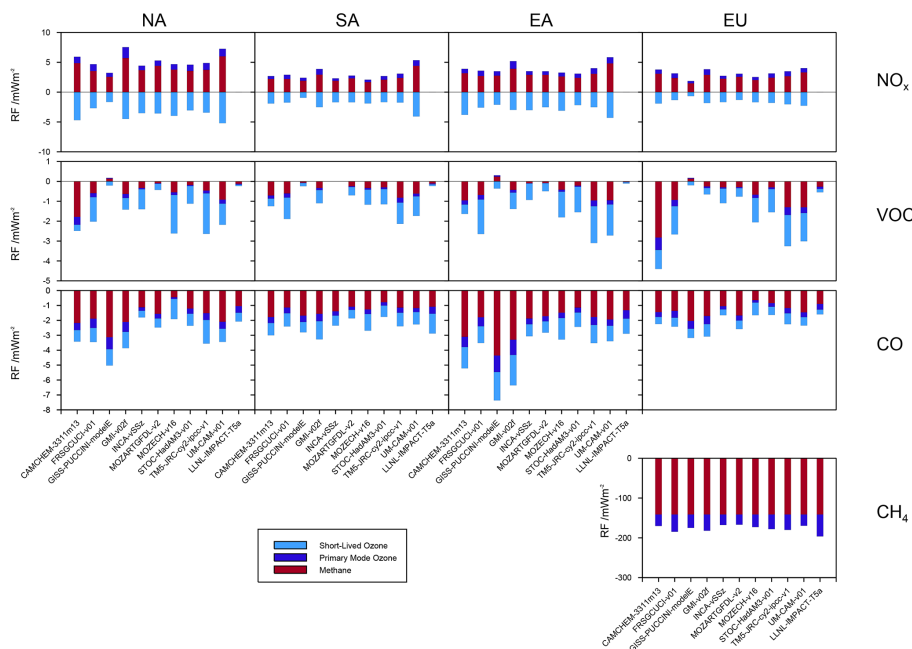


**Figure 2.** Spatial distribution of the deviation from the ensemble mean in annual-mean column integrated short-lived ozone perturbation ( $\text{g m}^{-2}$ ) for the  $\text{NO}_x$  NA case (see Table 1) for each individual model (top three rows). The bottom row shows the ensemble mean deviation (centre, by definition this is zero everywhere) and the plus (left) and minus (right) one SD from this mean.



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**Figure 3.** Radiative forcing for NO<sub>x</sub> (first row), VOC (second row), CO (third row), and CH<sub>4</sub> (bottom), for each of the 11 models, for each of the four regions given in Table 1. Methane forcings (bottom row) are given as if resulting from a perturbation of methane emissions – see text for details. Units are mW m<sup>-2</sup>. Colours show RF due to short-lived ozone (light blue), methane (red) and primary mode ozone (dark blue).

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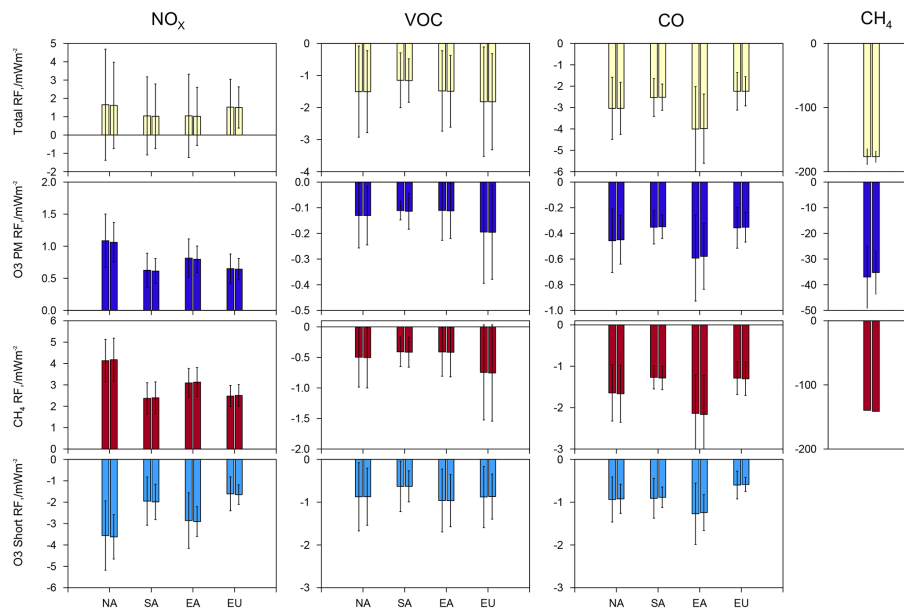
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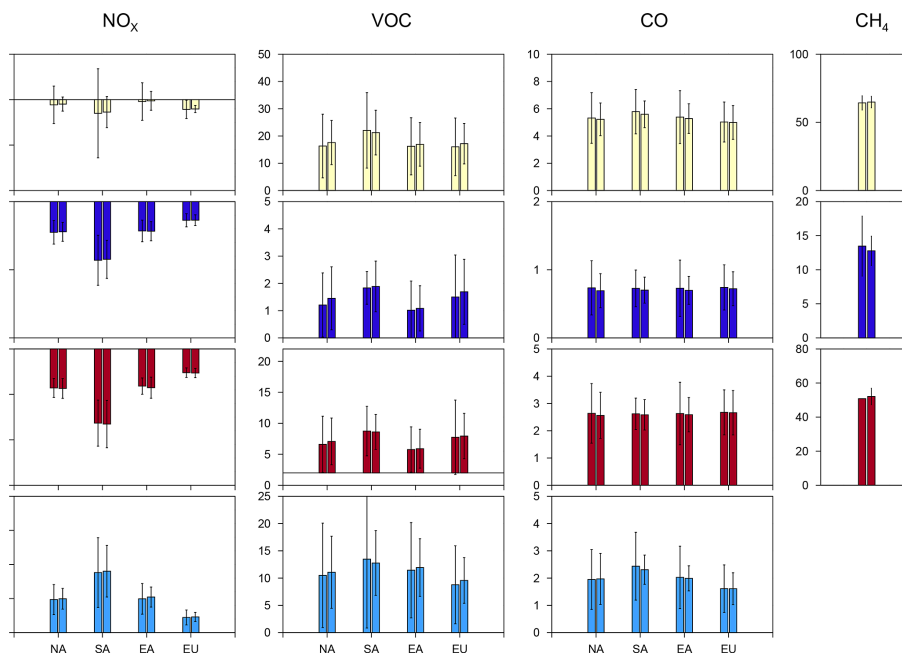
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**Figure 4.** Ensemble-mean radiative forcing for (first column)  $\text{NO}_x$ , (second column) VOC, (third column) CO, and (right)  $\text{CH}_4$ , for (top, yellow) total RF, (second row, dark blue) RF due to ozone PM, (third row, red) RF due to methane, and (bottom row, pale blue) RF due to short-lived ozone. For each pair of bars, the right-hand bar denotes the true mean,  $\overline{\text{RF}}$ , and the left-hand bar gives the ensemble value calculated using the method of Fry et al. (2012),  $\text{RF}_{(\text{EN})}$ . Units are  $\text{mW m}^{-2}$ .

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**Figure 5.** 20 year GWP for (first column)  $\text{NO}_x$ , (second column) VOC, (third column) CO, and (right)  $\text{CH}_4$ , for (top, yellow) total GWP, (second row, dark blue) GWP due to ozone PM, (third row, red) GWP due to methane, and (bottom row, pale blue)  $\overline{\text{GWP}}$  due to short-lived ozone. For each pair of bars, the right-hand bar denotes the true mean,  $\overline{\text{GWP}}$ , and the left-hand bar gives the ensemble value calculated using the method of Fry et al. (2012),  $\text{GWP}_{(\text{EN})}$ .

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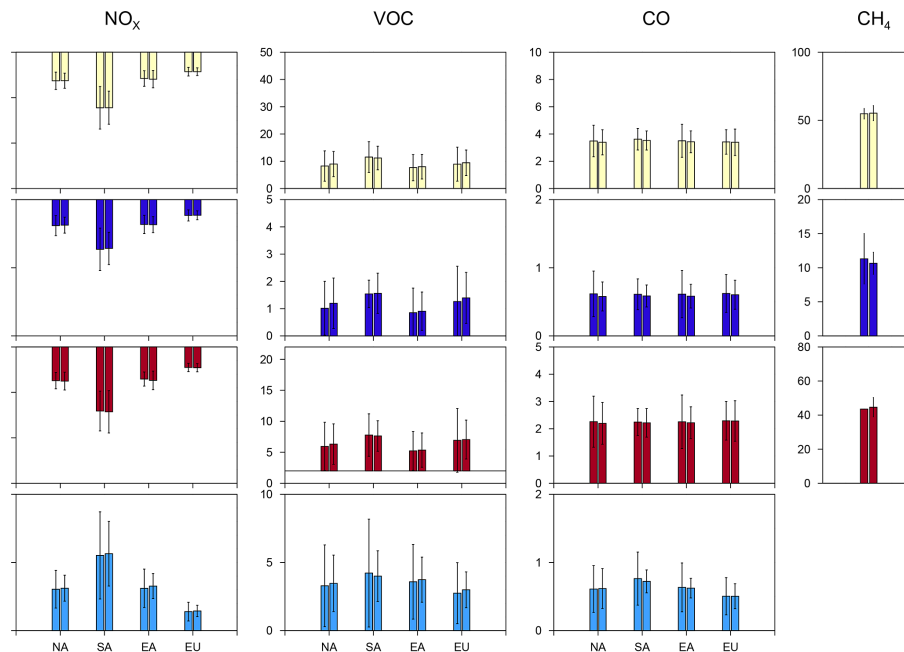
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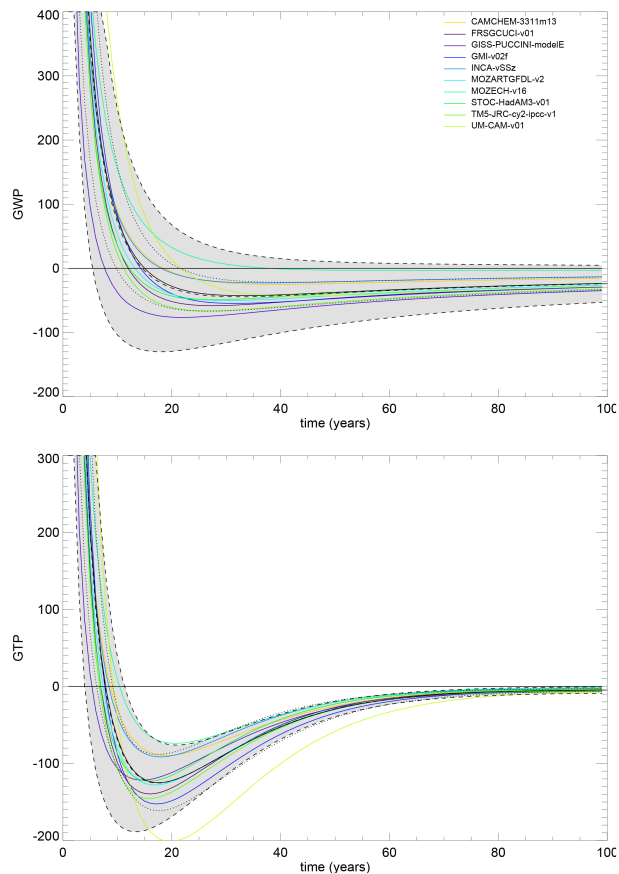
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**Figure 6.** 20 years GTP for (first column)  $\text{NO}_x$ , (second column) VOC, (third column) CO, and (right)  $\text{CH}_4$ , for (top, yellow) total GTP, (second row, dark blue) GTP due to ozone PM, (third row, red) GTP due to methane, and (bottom row, pale blue) GTP due to short-lived ozone. For each pair of bars, the right-hand bar shows  $\text{GTP}$ , and the left-hand bar shows,  $\text{GTP}_{(\text{EN})}$ .

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**Figure 7.** Time evolution of (top) GWP and (bottom) GTP for the  $\text{NO}_x$  SA case, showing each model. The solid black line and surrounding dotted lines represent the model ensemble mean and SD. The dashed lines and shaded area represent the mean and SD using the method of Fry et al. (2012).

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