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Implications of model bias in carbon monoxide for methane lifetime

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Abstract

A low bias in carbon monoxide (CO) at high northern latitudes is a common feature of chemistry climate models (CCMs) that may indicate or contribute to a high bias in simulated OH and corresponding low bias in methane lifetime. We use simulations with

⁵ CO tagged by source type to investigate the sensitivity of the CO bias to CO emissions, global mean OH, and the hemispheric asymmetry of OH. Our results show that reducing the hemispheric asymmetry of OH improves the agreement of simulated CO with observations. We use simulations with parameterized OH to quantify the impact of known model biases on simulated OH. Removing biases in ozone and water vapor as well as reducing Northern Hemisphere NO_x does not remove the hemispheric asymmetry in OH, but brings the simulated methyl chloroform lifetime into agreement with observation-based estimates.

1 Introduction

Carbon monoxide (CO) is an ozone precursor and the primary sink of the hydroxyl radi-¹⁵ cal (OH) in the troposphere. Consequently, CO indirectly impacts climate by increasing tropospheric ozone and increasing the lifetimes of methane and other short lived greenhouse gases (GHGs), as well as eventually oxidizing to CO₂ (e.g. Prather, 1996; IPCC, 1990). The effect of CO on OH also leads to impacts on oxidation of SO₂ to sulfate, providing another climate forcing (Shindell et al., 2009). Previous studies calculated

- ²⁰ global warming potentials due to these effects using box models (Daniel and Solomon, 1998), 2-dimensional models (Fuglestvedt et al., 1996; Johnson and Derwent, 1996), or 3-dimensional models (Derwent et al., 2001; Fry et al., 2013, 2012; Berntsen et al., 2005; Shindell et al., 2009). Since neither CO nor ozone is well mixed in the atmosphere, the location of the CO perturbation affects its climate impact. CO emissions
- ²⁵ in the tropics have a greater impact on ozone radiative forcing than emissions at high latitudes (Bowman and Henze, 2012) due the intense photochemistry in the tropics as



well as the presence of deep convection, which can loft ozone precursors to the upper troposphere where the ozone radiative forcing is greatest (Fry et al., 2013; Naik et al., 2005).

- While a large number of modeling studies have investigated the sources, transport,
 and distribution of CO, global models often show major biases compared to observations. A study of 26 atmospheric chemistry models found that the simulated CO was biased low in the extratropical Northern Hemisphere (Shindell et al., 2006) compared to satellite observations from MOPITT (Emmons et al., 2004) and surface observations, especially during spring. Shindell et al. (2006) attribute this bias primarily to an underestimate in Northern Hemisphere (NH) CO emissions, particularly from Asia. Monks et al. (2015) found that arctic CO is biased low in the multi-model POLARCAT Model Intercomparison Project (POLMIP), and identified differences in global OH concentrations as a major driver of the inter-model differences in arctic CO. The multi-model
- mean of the Atmospheric Chemistry and Climate Model Intercomparison Project (AC CMIP) simulations also shows a negative bias compared to both MOPITT and surface observations in the northern extratropics (Naik et al., 2013). Naik et al. (2013) found that ACCMIP models underestimate the methane and methyl chloroform lifetimes compared to the observation-based estimates of Prinn et al. (2005) and Prather et al. (2012) and produce a high bias in the NH to Southern Hemisphere (SH) ratio of OH, consis-
- tent with the underestimate of NH CO. A recent comparison of simulated and observed methyl chloroform levels also indicates that the NH/SH OH ratio is 0.97 ± 0.12 rather than the value of 1.28 calculated by the ACCMIP models (Patra et al., 2014). However, OH and CO are major losses for each other, complicating the determination of how much CO bias drives OH bias vs. OH bias driving CO bias.
- Previous modeling studies examined the consistency of CO emission estimates with surface and satellite observations. A modeling study by Duncan et al. (2007a) showed that their model compared well to observations in the NH extratropics, but they point out that a low bias in their model emissions may have been compensated by an assumption made in their simplified chemical scheme that non-methane hydrocarbons



(NMHCs) oxidize to CO immediately upon release; this assumption is valid during summer months for short-lived NMHCs (e.g., alkenes, isoprene), but is not valid in winter and spring for longer-lived NMHCS (e.g., alkanes). Their finding is consistent with inversions of MOPITT CO data that show that including greater winter emissions from

- the NH reduces the negative bias in springtime CO at northern latitudes (Petron et al., 2004). Kopacz et al. (2010) inverted CO observations from multiple satellites and concluded that northern mid-latitude CO sources were underestimated in winter, and that implementing large seasonal variations in emissions improved model agreement with observations. The inversion study of Fortems-Cheiney et al. (2011) also found that the
- ¹⁰ posterior CO emissions had large seasonality in the NH, with the maximum occurring in spring. However, the source strengths estimated by inversions are influenced by factors such as model transport (Arellano and Hess, 2006) and the concentrations of other species that interact with CO through OH chemistry (Pison et al., 2009; Muller and Stavrakou, 2005; Jones et al., 2009).
- ¹⁵ Uncertainty in the tropospheric burden and distribution of OH leads to further uncertainty in the CO budget. Hooghiemstra et al. (2011) found that higher NH OH concentrations led to higher anthropogenic CO emissions in their inversion study, while lower OH concentrations over tropical land masses and the SH led to lower biomass burning CO emissions and less CO from NMHCs. Duncan et al. (2007a) found that reducing
- OH by 20% globally improved the comparison of their simulated CO with surface observations in some locations, but degraded the comparison at other locations. Patra et al. (2014) suggest that top-down emission estimates from models with much higher OH in the NH than SH likely overestimate NH countries' emissions of CO and other reactive species. Mao et al. (2013) found that including conversion of HO₂ to H₂O on reactive species. Mao et al. (2013) found that including conversion of HO₂ to H₂O on
- ²⁵ aerosols reduced OH concentrations in a global model, correcting much of the negative model bias in the extratropical NH with the largest CO increase occurring in spring.

Understanding the causes and implications of CO bias in chemistry climate models is important for climate prediction as it may indicate or contribute to biases in methane and ozone and their respective radiative forcing contributions. The goal of this study



is to quantify the relationship of the extratropical NH CO bias seen in chemistry climate models with bias in oxidant concentrations and methane lifetime. We use the GEOS-5 Chemistry Climate Model (GEOSCCM) to compare the impact of increasing CO emissions vs. reducing OH concentrations on the CO distribution. We also quantify the contribution of model biases in other constituents such as ozone and water vapor to the simulated OH and CO distributions and methane lifetime.

2 Methods

2.1 Constituent observations and assimilated fields

Our primary constraint on the model CO distribution comes from surface observations of CO from the NOAA Global Modeling Division (GMD) network (Novelli and Masarie, 2014). We use the monthly mean data. The MOPITT instrument on the TERRA satellite provides additional constraints on the CO distribution. MOPITT provides almost global coverage of CO every three days from March 2000 to present (Edwards et al., 2004). We use the level 3 CO column data from the MOPITT version 5 thermal infrared (TIR) product (Deeter et al., 2011, 2013).

Observations of tropospheric ozone are important for constraining the source of OH. Ziemke et al. (2011) created a climatology of tropospheric column ozone (TCO) based on the difference between the stratospheric column ozone (SCO) from the Microwave Limb Sounder (MLS) and total ozone column data from the Ozone Monitoring Instru-²⁰ ment (OMI). We use the TCO product for 2004–2010 to constrain the tropospheric ozone column. Stratospheric ozone is also important for constraining the OH source due to its effect on photolysis (Rohrer and Berresheim, 2006). We use the Global Modeling and Assimilation Office (GMAO) ozone assimilation product for 2005–2010 to

constrain stratospheric ozone concentrations. The GMAO assimilated ozone product, described in Ziemke et al. (2014) and Wargan et al. (2015), is a gridded product that



ingests MLS ozone profiles and OMI total column ozone into the GEOS-5 assimilation system.

Water vapor is another important influence on OH concentrations. We use specific humidity from the Modern-Era Retrospective Analysis for Research and Applications (MERRA) (Rienecker et al., 2011).

2.2 Model and methodology

Our analysis uses several chemistry options within the GEOSCCM framework to assess possible causes and impacts of CO and OH bias. After spin-up, we conduct a series of time slice simulations of 1999–2009 with fixed emissions using observed sea surface temperatures (SSTs) to drive the CCM meteorology, and then average our results over all years of the time slice. All simulations use the Fortuna version of GEOS-5 (Molod et al., 2012) and have 2° latitude by 2.5° longitude horizontal resolution and 72 vertical levels. We describe each chemistry option and associated experiments below and in Table 1.

15 2.2.1 GMI chemistry option

GEOSCCM integrates the chemistry mechanism of the Global Modeling Initiative (GMI)
 CTM (Duncan et al., 2007b; Strahan et al., 2007) into the GEOS-5 AGCM. The GMI chemistry includes a comprehensive mechanism of tropospheric and stratospheric chemistry, including 117 species and over 400 reactions. The reference simulation for
 this study, hereafter called RefGMI, is the year 2000 timeslice simulation conducted for the ACCMIP intercomparison. The configuration of the ACCMIP simulations is described in Lamarque et al. (2013). The biases in CO and methane lifetime seen in the GEOSCCM simulation are similar to those seen in the ACCMIP multi-model mean (Naik et al., 2013). Consequently, our analyses of bias in the GEOSCCM are likely applicable to other chemistry climate models as well.



We conduct a sensitivity simulation, described in Sect. 3.2, that is identical to the ACCMIP simulation except for an increase in CO emissions. We refer to this simulation as GMI-HiEmis. Since trace gases in the GMI option of the GEOSCCM are radiatively coupled to the underlying GCM, altering emissions within this option produces feed-⁵ backs between CO, O_3 , CH_4 , and radiation and transport.

2.2.2 CO-only option

GEOSCCM includes a CO-only option to "tag" CO according to source type or location (Bian et al., 2010). This simplified chemistry option allows us to separate the contributions of different CO sources and to quantify the impact of a specific change in OH. In

- this chemistry option, the loss of CO is calculated based on prescribed OH fields, so changes in CO do not feed back onto OH. Our reference tagged-CO simulation, called RefCOonly, uses monthly OH fields archived from the GEOSCCM ACCMIP simulation, and the CO sources from methane and isoprene oxidation are calculated using monthly methane and isoprene fields archived from the ACCMIP simulation as well. Emissions
- and other forcings are chosen to parallel the ACCMIP simulation; however, the COonly 15 option includes an amplification factor for CO emissions to account for the absence of co-emitted NMHCs (Duncan et al., 2007a). Section 3.2 describes CO-only sensitivity studies.

The GEOSCCM includes an option to constrain the meteorology with MERRA or any GMAO assimilation product. The simulation in pulled towards the MERRA anal-20 vsis through application of an incremental analysis update (Bloom et al., 1996), calculated every six hours from comparison of the simulation with the analysis. We conduct a COonly simulation with specified dynamics from MERRA, which we refer to as COonlySD. The COonlySD simulation has the same emissions and OH field as Ref-

COonly, but the tracer transport will differ between the two simulations. 25



2.2.3 CO-OH option

A third chemistry option within the GEOSCCM is the CO-OH parameterization of Duncan et al. (2000, 2007a). This chemistry option is of intermediate complexity between the GMI and Tagged-CO options, and was designed to represent an older version of the

- ⁵ chemistry scheme used in the GMI chemistry option. This option is similar to the COonly option except that OH is calculated interactively, providing a feedback between CO and OH concentrations. In our reference simulation with this option, RefCO-OH, chemistry inputs to the OH parameterization such as ozone, NO_x, CH₄, and NMHC concentrations are prescribed from the archived monthly output of the ACCMIP sim-
- ulation. The temperature, water vapor, and irradiance-related variables input to the parameterization are calculated within the CCM. Since the parameterization was only designed to provide tropospheric OH values, we use the results of the parameterization from the surface to 300 hPa, and archived OH fields taken from the GMI-chemistry simulation above 300 hPa. The CO emission amplification factors are adjusted slightly
 downward compared to the COonly simulation to bring the RefCO-OH simulation into
- downward compared to the COonly simulation to better agreement with the RefGMI simulation.

We use the CO-OH option to examine how biases in a particular model field such as ozone affect OH and CO concentrations. Section 3.4 describes sensitivity studies conducted with this option.

20 2.2.4 Comparison of OH between chemistry options

The choice of chemistry option affects the CO and OH distributions produced by the GEOSCCM. Annually averaged OH is higher in the RefGMI simulation in both hemispheres (Table 2; Fig. 1). High OH values extend further down in the troposphere in RefGMI than in RefCO-OH. Global annual mean mass-weighted tropospheric OH is approximately 7% lower in RefCO-OH than in RefGMI, but the NH/SH OH ratio is

²⁵ approximately 7% lower in RefCO-OH than in RefGMI, but the NH/SH OH ratio is marginally higher: 1.22 instead of 1.19. Prather et al. (2012) report an observationbased estimate of methane lifetime of 9.1 ± 0.9 years, and a methane lifetime against



tropospheric OH of 11.2 ± 1.3 years. The lifetimes of methyl chloroform and methane against tropospheric OH in the RefGMI simulation are 5.9 and 9.6 years, respectively (Table 2). The lifetimes of methyl chloroform and methane against tropospheric OH in the RefCO-OH simulation are 6.4 and 10 years, respectively, within the uncertainty of the observation-based estimates. The seasonal cycles are similar in both simulations, but the difference in NH OH is larger in the first half of the year than the second half (Fig. 1d). Consequently, when using the CO-OH parameterization to investigate OH and CO, we present the changes due to a given factor rather than the absolute values of CO, OH, and methane lifetime for easier comparison with the other simulations.

10 3 Results

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We use the three chemistry options of the GEOSCCM to separate the contributions of emissions, chemistry, and transport to model bias in CO and methane lifetime. Section 3.1 compares the CO distribution simulated by the three options to observations and discusses the consistencies and differences between simulations. We analyze the impacts of increasing emissions, decreasing OH, and changing model transport on the CO distribution in Sect. 3.2, and examine how changing CO emissions affects ozone and OH in Sect. 3.3. Section 3.4 investigates the contribution of known model biases to the simulated OH and methane lifetime. Table 1 summarizes the key results of each of the sensitivity studies.

20 3.1 Comparison of CO in the reference simulations and observations

This section presents a comparison of CO and OH distributions from the RefGMI, RefCO-only, and RefCO-OH simulations to each other and to observations. Figure 2 compares the annual cycle of CO in simulations to observations from six GMD sites selected to represent a range of latitudes. The RefGMI simulation (purple) is biased low at the NH sites, with the bias most prominent in the first half of the year, especially



NH spring. The simulation shows less bias at the SH sites, but some negative bias is evident in SH spring. The RefCOonly simulation (green) shows similar results to RefGMI, including a large negative bias in the NH in spring. Consequently, diagnosing the cause of bias in the RefCOonly simulation can provide insight into the bias in the RefGMI simulation.

The RefCO-OH simulation (orange) shows less springtime bias compared to observations than the RefGMI and RefCOonly simulations. However, its annual cycle is shifted later, resulting in more negative biases in September–December at the higher northern latitudes. The difference between the RefCO-OH and the other two reference simulations is explained by the differences in the OH field calculated by the parameterization compared to that calculated by the GMI chemistry mechanism. In April, when the greater NH bias of the RefGMI simulation is most evident (Fig. 2), the RefGMI OH is markedly higher than the RefCO-OH OH (Fig. 1).

3.2 Sensitivity of simulated CO to sources, OH, and transport

3.2.1 Sensitivity of CO-only simulations to sources and OH

Adjusting the strength of mid-latitude CO sources can reduce the bias in the interhemispheric CO gradient. We use CO-only simulations to examine the impact of increasing specific sources of CO. Following the method of Strode and Pawson (2013), we estimate the impact of increasing a particular source by increasing the tagged-CO tracer

- for that source and then re-computing total CO. We impose increases of 10, 20, 50, 100, and 150 % for each tagged tracer. Figure 3 shows how the increased Asian an-thropogenic CO (COaa) or tropical biomass burning CO (COtrbb) alters the comparison between modeled CO and the GMD CO observations as a function of latitude taken as an average of March to August. Similar results are present for spring and summer individually. An increase in COaa of approximately 100 % removes the negative bias at birth latitude sites, but here little effect on the small present for spring and summer birth.
- at high latitude sites, but has little effect on the small negative bias at low latitudes (Fig. 3a). CO transported southward from Asia encounters higher OH than that trans-



ported northward, leading to a shorter lifetime (Duncan and Logan, 2008). Increasing COtrbb improves the model agreement with observations at low latitudes, but creates overestimates at some tropical sites while providing only a modest reduction in the high latitude bias (Fig. 3b).

- ⁵ We examine the impact of reducing OH concentrations globally or only in the NH in our COonly simulation and find that a large decrease in NH OH is effective in reducing the high latitude CO bias. Naik et al. (2013) found that the ACCMIP multi-model mean underestimated tropospheric OH by 5–10% globally. The NH/SH OH ratios in the ACCMIP multi-model mean and GEOSCCM ACCMIP simulation are 1.28 and
- 1.18, respectively (Naik et al., 2013), compared to the (Patra et al., 2014) estimate of 0.97 ± 0.12. Figure 3c shows that reducing OH by 5–10% leads to a small increase in CO across all latitudes, yielding a small reduction in the bias compared to GMD observations. Decreasing OH by 20% in the NH only leads to a large improvement in high latitude CO compared to observations, with only a small effect in the Southern
 15 Hemisphere.

3.2.2 Impact of sources and OH on global mean and IHG bias in CO

We find that achieving zero bias in both the IHG and the global mean would require changes in multiple emission sources. Figure 4 illustrates how changing the concentrations of several tagged CO tracers, as well as OH, impacts the global mean bias, inter-hemispheric gradient (IHG) bias, and correlation (r^2) of the simulated CO compared to the GMD observations for March through May. Increasing anthropogenic CO from Asia (COaa), Europe (COea), or North America (COnaa), or increasing Russian biomass burning (COrubb) reduces the bias in the IHG as well as the global mean. Increasing CO from tropical biomass burning (COtrbb) leads to a smaller reduction in

the IHG bias, while increasing CO from biogenic emissions (CObio) reduces the mean bias with little effect on the IHG (Fig. 4a). Consequently, increases in both groups of emissions are necessary to simultaneously remove the global mean bias and correct the IHG.



A similar analysis for June through August (Fig. 5) shows that increasing COaa, COea, COnaa, or COrubb can eliminate the majority of the bias in both the global mean and the IHG in summer. Increasing COrubb is more effective in summer than spring since boreal biomass burning emissions are larger in summer. In contrast to the spring

results, increasing COtrbb in summer leads to greater bias in the IHG. This difference occurs because tropical biomass burning occurs primarily north of the equator in March and April but shifts to the Southern Hemisphere in June, July, and August.

We exclude increases in COnaa and CObio in the rest of our study. While increases in COaa, COea, COnaa, and COrubb show similar slopes for IHG vs. global mean bias, increasing COnaa reduces the correlation with observations, where as increasing the

- other sources yields a slight improvement in correlation (Figs. 4b and 5b). Increases in both COtrbb and CObio reduce the correlation with observations in summer, but the effect is also present for CObio in spring.
- Figures 4 and 5 also show the impact of changing OH. The sensitivity of CO to thanges in OH is location dependent, with higher sensitivity in regions without strong local CO sources (Holloway et al., 2000). Reducing OH globally reduces both the IHG and global mean bias. However, reducing OH by 20% in the NH only yields a greater reduction in both biases as well as the greatest improvement in correlation. We refer to the simulation with the 20% decrease in NH OH as COonlyLowNHOH.

20 3.2.3 Impact of increased source strength in a GMI chemistry simulation

Based on the tagged tracer results (Figs. 4 and 5), we conduct a new simulation, called HiEmis, using the GMI chemistry with increased emissions from Asian and European anthropogenic sources as well as tropical and Russian biomass burning. All other aspects of the simulation are identical to the standard simulation. We adjust the emissions for each season based on the biases and tagged tracer sensitivities for the season. We choose the adjustment factors by solving for the linear combination of COaa + COea + COrubb and COtrbb that minimizes the error in both the IHG and the global mean bias. Table 2 shows the emissions adjustments for each season. Figure 6a



and b shows the simulated surface CO for April 2007 in the RefGMI and GMI-HiEmis simulations compared to GMD observations. The HiEmis simulation shows less negative bias at many of the high latitude sites, but shows a positive bias over Europe, one of the locations where the emissions were increased. This suggests that our adjust-⁵ ment of COea is too large, which is unsurprising given that European emissions are relatively well known.

3.2.4 Impact of adjusting sources vs. NH OH on comparison to observations

We next compare the impact of increasing emissions with that of reducing NH OH by 20%. The RefCOonly simulation shows a similar surface CO distribution to RefGMI
(Fig. 6a and c). The COonlyLowNHOH scenario (Fig. 6d) improves the agreement with the remote high latitude sites compared to the RefCOonly simulation, but like the HiEmis case, it leads to an overestimate of CO concentrations over Europe and eastern United States. Consequently, a combination such as reduced NH OH and reduced emissions over the eastern US is likely needed to reconcile the simulated CO with observations.

Comparison to MOPITT data suggests that the spring increase in Asian emissions used to reduce the NH bias in the GMI-HiEmis simulation is unrealistic (Fig. 7). Sparse surface data in this region makes it difficult to determine from comparison with GMD observations whether this higher CO seen in GMI-HiEmis is realistic (Fig. 6), so we also

- ²⁰ compare the four simulations shown in Fig. 6 to the CO column from MOPITT. Both the RefGMI and RefCOonly simulations show a large negative bias in NH CO compared to MOPITT (Fig. 7a and c). The GMI-HiEmis and COonlyLowNHOH simulations both reduce this negative bias (Fig. 7b and d), but the increased emissions of the GMI-HiEmis simulation lead to a greater overestimate of CO over East Asia and Indonesia.
- ²⁵ Furthermore, eliminating the increase in COea to reduce the high bias compared to surface observations over Europe and compensating with a larger adjustment in Asian CO would lead to an even greater bias over Asia compared to MOPITT.



3.2.5 CO sensitivity to transport

Transport, in addition to chemistry and emissions, plays a role in the IHG of CO. We examine the sensitivity of the CO bias to model transport by comparing RefCOonly, which is a free-running CCM simulation, with the COonlySD simulation, which has year-

- ⁵ specific meteorology from MERRA. The largest difference between the simulations occurs poleward of 30° N during July through October (Fig. 8a). The use of specified meteorology makes only a small difference in the global mean and IHG CO biases compared to surface observations in March–May (Fig. 4), but leads to a large reduction in bias as well as improved spatial correlation with the GMD observations in June–August
- (Fig. 5). The biases in global mean CO and the CO IHG are -3.5 and -6.6%, respectively, in COonlySD, compared to -11 and -26%, respectively, in RefCOonly. More CO from Northern Hemisphere anthropogenic and boreal biomass burning sources remains in the lower troposphere and reaches the high latitudes in COonlySD, where as more is transported to the upper troposphere in RefCOonly (Fig. 8b).

15 3.3 Impact of increased CO emissions on ozone, OH, and CH₄ lifetime

The increased CO sources considered in Sect. 3.2 will impact ozone production and the concentrations of OH and other constituents. We quantify these effects by comparing the GMI-HiEmis simulation to the RefGMI simulation.

Including larger CO emissions increases the loss of OH, reducing OH concentra-

- ²⁰ tions. The mass-weighted global mean tropospheric OH in the GMI-HiEmis simulation is 1.11×10^6 molec cm⁻³, a 3% decrease compared to the RefGMI simulation (Table 2). The methane lifetime against tropospheric OH also increases slightly, from 9.6 years in the RefGMI simulation to 9.9 years in the HiEmis simulation. This slightly improves the agreement with the observation-based estimate of 11.2 ± 1.3 years of
- ²⁵ Prather et al. (2012). Since the decrease in OH occurs primarily in the Northern Hemisphere, the NH/SH OH ratio shows a small reduction from 1.19 in the RefGMI simula-



tion to 1.16 in the GMI-HiEmis simulation, but the large hemispheric asymmetry in OH remains.

Both the RefGMI and GMI-HiEmis simulation show a large high bias in NH TCO compared to the OMI/MLS observations, as well as a low bias in the equatorial Pa-

⁵ cific and the extratropical SH (Fig. 9). Similar biases were present the ACCMIP multimodel mean (Young et al., 2013), indicating that these biases are a common feature in chemistry climate models. The increased CO emissions in the GMI-HiEmis simulation slightly increase the high bias in TCO in the northern midlatitudes, but the increase is small compared to the bias in the RefGMI simulation.

3.4 Sensitivity of OH to model biases

Given the sensitivity of the CO biases to OH (Figs. 3–5) and the relatively small changes in OH resulting from the increased CO emissions in the GMI-HiEmis simulation, we next examine how other biases in the RefGMI simulation may impact OH concentrations and consequently the CO distribution and methane lifetime. We conducted a series of sensitivity studies using the CO-OH parameterization option to isolate the

impact of several known model biases on OH and CO distributions.

The primary source of OH in the troposphere is ozone photolysis followed by reaction of O^1D with water vapor, and secondary production of OH occurs through reaction of HO₂ with NO or O₃ (Spivakovsky et al., 2000). Consequently, simulated OH con-

- ²⁰ centrations are sensitive to errors in NO_x and ozone concentrations, water vapor, and factors that influence photolysis such as overhead ozone column and clouds. Holmes et al. (2013) found that temperature, water vapor, stratospheric ozone, and emissions from biomass burning and lightning could together explain most of the interannual variability in methane lifetime against OH. Duncan and Logan (2008) found that changes
- in the ozone column were a major driver of OH variability over 1988–1997. Murray et al. (2013) found that lightning was more important for OH variability over 1998–2006, when there was less variability in the overhead ozone column. Murray et al. (2014) found that on glacial–interglacial timescales, OH concentrations were proportional to



the tropospheric ozone photolysis rate, specific humidity, and reactive nitrogen emissions, and inversely proportional to CO emissions. Here, we examine the sensitivity of OH to model biases in some of these factors.

3.4.1 Sensitivity to tropospheric ozone

⁵ Comparison of the CCMs that participated in the ACCMIP study to OMI/MLS TCO reveals positive biases in simulated tropospheric ozone over the NH midlatitudes, and negative biases over the tropical Pacific and SH (Young et al., 2013), which leads to an overestimate in NH OH production and an underestimate in SH OH production (Naik et al., 2013). The multi-model mean tropospheric ozone also shows a high bias in the NH and low bias in the SH compared to the Tropospheric Emission Spectrometer (TES) (Bowman et al., 2013). A multi-species assimilation study by Miyazaki et al. (2012) found that assimilating TES ozone increased OH concentrations in the SH.

We use the CO-OH parameterization to investigate the impact of removing the GEOSCCM's tropospheric ozone column bias relative to the OMI/MLS observations

- (Fig. 9). We scale the tropospheric ozone values input to the parameterization for each month between October 2004 and December 2010 from 60° S–60° N so that the tropospheric ozone column is unbiased compared to the OMI/MLS TCO for that month. We use the scaled ozone input in a sensitivity study for October 2004 to January 2010, called CO-OHSensTCO, which parallels the RefCO-OH simulation but uses
- the scaled tropospheric ozone values in the OH parameterization. CO-OHSensTCO shows a small (2%) decrease in global mean OH compared to RefCO-OH, with a 3% decrease in NH OH and 1% decrease in SH OH. The increased OH leads to a 2% increase in both methane and methyl chloroform lifetimes against OH. The NH/SH OH ratio decreases slightly to 1.19 (Table 4), suggesting that the inter-hemispheric gra-
- ²⁵ dient in the tropospheric ozone bias makes only a minor contribution to the model's interhemispheric OH asymmetry.



3.4.2 Sensitivity to stratospheric ozone

We next examine how biases in stratospheric ozone affect OH through their role in photolysis. Voulgarakis et al. (2013) found that changes in stratospheric ozone and tropospheric OH in the ACCMIP models both correlated strongly with $J(O^1D)$. We con-

- ⁵ duct a sensitivity study for 2005 through 2009, CO-OHSensStO3, which parallels the RefCO-OH simulation but replaces the simulated ozone with the GMAO ozone assimilation in the stratosphere for input into the parameterization. The change in global mean OH in CO-OHSensStO3 is nearly identical to that of CO-OHSensTCO, but CO-OHSensStO3 places more of the change in the SH, causing the NH/SH OH ratio
- to increase to 1.23 (Table 4). The change in OH due to stratospheric ozone bias is small in part because GEOSCCM has relatively small biases in tropical stratospheric ozone. However, the multi-model Chemistry Climate Model Validation-2 (CCMVal-2) study (Morgenstern et al., 2010) shows a large spread across models for column ozone in the tropics (Austin et al., 2010). Thus, biases in stratospheric ozone may play a larger role in tropospheric OH in models with larger stratospheric ozone biases.

3.4.3 Sensitivity to water vapor

We investigate the impact of model biases in water vapor on OH concentrations through its role in OH production. Inter-model differences in OH in the POLMIP study are correlated with inter-model differences in simulated water vapor (Monks et al., 2015). The

²⁰ GEOS-5 AGCM exhibits a high bias in specific humidity compared to the MERRA reanalysis in much of the troposphere (Molod et al., 2012), and a high bias in the midtroposphere is also seen throughout the year in GEOSCCM compared to AIRS data (Lamarque et al., 2013).

We quantify the impact of this bias by conducting a sensitivity study, CO-OHSensQ, that applies altitude and latitude-dependent zonal mean scaling factors for each month to the specific humidity provided to the OH parameterization. The scaling factors are based on comparison of the RefGMI simulation to MERRA and are applied in 100 hPa



intervals between 900 and 200 hPa for 60° S to 60° N. These scaling factors are designed to remove the mean bias in the simulated specific humidity compared to MERRA while allowing the simulated water vapor to vary in space and time in a manner consistent with the simulated meteorology. The water vapor scaling is applied only to the OH

⁵ parameterization, and does not impact the general circulation of the simulation. The scaling results in lower specific humidity in the middle and upper troposphere, with the largest percent reductions occurring between 300 and 500 hPa (Fig. 10).

Simulated OH is 6 % lower in the CO-OHSensQ simulation than the RefCO-OH simulation, and the methane and methyl chloroform lifetimes against OH are thus 6 % longer (Table 4). Consequently water vapor bias has a larger impact on OH concen-

¹⁰ longer (Table 4). Consequently, water vapor bias has a larger impact on OH concentrations than the stratospheric or tropospheric ozone biases in our simulations. The OH reduction is similar in both hemispheres in the annual mean, so the NH/SH OH gradient is unchanged compare to RefCO-OH.

3.4.4 Sensitivity to NO_x

- ¹⁵ Anthropogenic NO_x emissions, which contribute to secondary production of OH, are located primarily in the Northern Hemisphere. Consequently, an overestimate of these emissions could contribute to the NH/SH asymmetry in simulated OH. We quantify the sensitivity of simulated OH to NH NO_x with a sensitivity study, CO-OHSensNOx, which parallels RefCO-OH but includes 30% lower NO_x concentrations in the NH.
- ²⁰ This reduction in NH NO_x emissions leads to a 3% reduction in global OH (Table 4). SH and NH OH are reduced by 0.3 and 6%, respectively, reducing the NH/SH OH ratio to 1.15. While this shows that the hemispheric asymmetry in OH is affected by NO_x emissions, a much larger redistribution of NO_x emissions would be required to completely eliminate the asymmetry.



3.4.5 Summary of OH sensitivities

In the previous sections, we examined the sensitivity of OH and its interhemispheric gradient to model biases in tropospheric and stratospheric ozone, water vapor, and NH NO_x emissions. We find that water vapor has the largest impact on global mean OH, while NH NO_x emissions have the largest impact on the NH/SH ratio. However, none of these biases individually explains the 20 % reduction in NH OH that would remove the interhemispheric asymmetry and which Figs. 4 and 5 suggest are necessary to remove most of the CO bias. We note, however, that the OH parameterization simulations do not account for all the chemical feedbacks between ozone, methane, OH, and other species, and may underestimate the sensitivity of the full chemistry simulation to some of these biases.

Table 4 suggests that applying multiple bias corrections simultaneously would bring our simulation into good agreement with the observation-based estimates of global mean OH. We conduct a final sensitivity simulation, called CO-OHSensAll, incorporat-

- ¹⁵ ing the bias corrections for tropospheric and stratospheric ozone, as well as the specific humidity scaling and 30% reduction in NH NO_x emissions. This simulation results in a 13% reduction in global mean OH, with a 10% reduction in the SH and 16% reduction in the NH. The NH/SH OH ratio is reduced to 1.14. The lifetimes of methyl chloroform and methane in this simulation increase by 12 and 15%, respectively. Ap-
- ²⁰ plying this 15% increase in methane lifetime to the RefGMI methane lifetime against OH of 9.6 years would yield a lifetime of 11 years, consistent with the 11.2±1.3 year estimate of Prather et al. (2012). This shows that removing the main model biases related to OH production would bring simulated global mean OH into good agreement with observation-based estimates, although some NH/SH asymmetry would remain.
- The reduction in OH in the sensitivity studies compared to RefCO-OH leads to higher surface concentrations of CO. The bias in the global mean March through August surface CO changes from -4% in RefCO-OH to 3% in CO-OHSensAll, while the bias in the interhemispheric gradient changes from -11 to -4%. Figure 11 shows the latitudi-



nal distribution of the change in CO for each sensitivity simulation vs. RefCO-OH. The sum of the changes for CO-OHSensTCO, CO-OHSensStO3, CO-OHSensQ, and CO-OHSensNOx, shown in the dotted line, is similar but slightly smaller than the change for CO-OHSensAll, indicating small non-linearities in the system. Correcting water va-

⁵ por (CO-OHSensQ) makes a large contribution to the CO enhancement in both hemispheres, while adjusting NO_x emissions (CO-OHSensNOx) and to a lesser extent tropospheric ozone (CO-OHSensTCO) contributes to the larger increase in the Northern Hemisphere.

4 Conclusions

We examined possible causes of CO model bias, such as underestimated emissions or overestimated OH, in a global chemistry climate model. An underestimate of CO emissions can impact the chemistry–climate simulation through the interaction of CO with methane via OH and with ozone, but we find the effects to be small. In contrast, a CO bias due to excess OH would imply biases in methane lifetime, further influencing the simulated climate.

Either increasing emissions or decreasing Northern Hemisphere OH concentrations can remove the bias in the latitudinal gradient of CO compared to surface observations. However, we find that the large increases in Asian anthropogenic emissions needed to remove the negative CO bias at remote surface sites leads to overestimates of CO over

Asia compared to MOPITT. In contrast, reducing OH concentrations in the Northern Hemisphere improves the agreement between simulated and observed CO concentrations. This is consistent with the finding of Patra et al. (2014) that the ratio of Northern Hemisphere to Southern Hemisphere OH is close to one. We note that biases in OH, CO emissions, and transport are not mutually exclusive, and the model bias in CO is likely influenced by a combination of these factors.

The availability of satellite-based constraints on CO, O_3 , and water vapor enables us to assess model biases that affect the major sources and sinks of OH and hence



CO concentrations and methane lifetime. We used a CO-OH parameterization to explore the effect of model biases in ozone and water vapor on simulated OH and CO concentrations. Removing the high bias in Northern Hemisphere tropospheric ozone, a common feature of CCMs, had only a small effect on simulated OH and CH_4 lifetime.

- ⁵ A high bias in water vapor had a larger impact on global mean OH, but removing this bias did not remove the Northern vs. Southern Hemisphere asymmetry in OH. Removing both ozone and water vapor biases, as well as decreasing Northern Hemisphere NO_x , provided the desired increase in CH₄ lifetime, but was insufficient to remove the hemispheric asymmetry in OH. Thus, while a NH/SH OH ratio near one improves the simulation of CO, we cannot generate this ratio in our simulations by removing known
 - model biases in ozone and water vapor.

Our study suggests that the springtime low bias in CO at northern latitudes often seen in chemistry climate model simulations likely indicates a bias in CH_4 lifetime. Improving the model representation of water vapor and ozone, as well as reducing

- ¹⁵ uncertainty in NO_x emissions, could reduce these biases. However, additional research is needed to understand the causes of northern vs. southern asymmetry in simulated OH. Future field missions that provide data on the latitudinal distribution of CO and oxidant sources and losses will be valuable for understanding biases in simulated CO and OH.
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Table 1. Description of reference and sensitivity simulations used in this study with each chemistry option.

Simulation	Description	Key Result		
GMI chemistry simulations	· · · ·	-		
RefGMI*	ACCMIP timeslice simulation for 2000	CO biased low at high latitudes		
GMI-HiEmis	increased CO emissions compared to RefGMI	Regional CO biases compared to MOPITT		
CO-only simulations				
RefCOonly*	Tagged-CO version of RefGMI	Similar CO biases to RefGMI		
COonlyLowNHOH	NH OH uniformly decreased by 20 %	Improves agreement with GMD CO observations		
COonlySD	meteorology constrained by MERRA (specified dynamics)	Improves agreement with GMD CO observation in summer		
CO-OH simulations				
RefCO-OH*	Parameterized OH version of RefGMI	Lower OH than RefGMI		
CO-OHSensTCO	tropospheric ozone column adjusted to match OMI/MLS TCO	Small increase in CH_4 lifetime		
CO-OHSensStO3	stratospheric O_3 adjusted to match assimilated O_3	Small increase in CH ₄ lifetime		
CO-OHSensQ	tropospheric water vapor adjusted based on MERRA	Increase in CH ₄ lifetime		
CO-OHSensNOx	30 % decrease in NH NO _{x} concentrations	Small increase in CH ₄ lifetime; OH asymmetry reduced but not eliminated		
CO-OHSensAll	tropospheric and stratospheric O_3 , water vapor, and NH NO_x all adjusted	15 % increase in CH_4 lifetime; OH asymmetry reduced but not eliminated		

* Reference simulation.



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Table 2. Tropospheric OH concentrations and lifetimes against oxidation by tropospheric OH.

	RefGMI	GMI-HiEmis	RefCO-OH
Global Mean OH (10 ⁶ molec cm ⁻³)	1.14	1.11	1.051
SH OH (10 ⁶ molec cm ^{-3})	1.03	1.02	0.943
NH OH (10 ⁶ molec cm ^{-3})	1.22	1.18	1.15
NH/SH OH	1.19	1.16	1.22
CH ₄ lifetime (years)	9.65	9.89	10.5
CH_3CCI_3 lifetime (years)	5.91	6.06	6.39



Table 3. CO emission adjustment for the high emission simulation compared to the standard simulation.

Emission Increase (%)	Months			
	Jan–Feb	Mar–May	Jun–Aug	Sep-Dec
Asian and European anthropogenic	17.4	45.0	63.2	0.00
Russian BB	17.4	45.0	63.2	0.00
Tropical BB	26.4	60.6	28.2	0.00

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Table 4. NH/SH OH ratio and percent changes in tropospheric OH concentrations and lifetimes against tropospheric OH for the CO-OH sensitivity studies compared to the RefCO-OH simulation.

	SensTCO	SensStO3	SensQ	SensNOx	SensAll
NH/SH OH	1.19	1.23	1.22	1.15	1.14
	% change vs. RefCO-OH				
Global Mean OH	-2.1	-2.1	-6.2	-3.1	-13
SH OH	-1	-2.9	-6.3	-0.3	-10
NH OH	-3.1	-1.5	-6.1	-5.7	-16
CH₄ lifetime	2.5	2.4	5.7	3.5	15
CH ₃ CCl ₃ lifetime	2.4	2.3	5.9	3.5	15



Figure 1. Simulated annual zonal mean OH concentration for the RefGMI **(a)** and RefCO-OH simulations **(b)** averaged over 1999–2009. **(c)** The annual cycle of the pressure-weighted column mean OH by latitude from the RefGMI simulation. The column is averaged between the surface and 200 hPa. **(d)** The annual cycle of hemispheric mean OH from the RefGMI (solid lines) and RefCO-OH (dashed lines) simulations for the Northern Hemisphere (black lines) and Southern Hemisphere (gray lines).





Figure 2. Monthly mean GMD observations (black) and results from the RefGMI (purple), RefCOonly (green), and RefCO-OH (orange) simulations. Circles and error bars represent the mean and min-max range, respectively, for 1999–2009. The GMD sites are as follows: **(a)** Ny-Alesund, Svalbard (ZEP, 78.9° N, 11.9° E), **(b)** Barrow, Alaska (BRW, 71.3° N, 156.6° W), **(c)** Trinidad Head, California (THD, 41.0° N, 124.2° W), **(d)** Izana, Tenerife, Canary Islands (IZO, 28.3° N, 16.5° W), Tutuila, American Samoa (SMO, 14.2° S, 170.6° W), and **(e)** Cape Grim, Tasmania (CGO, 40.7° S, 144.7° E).





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Figure 3. Latitudinal distribution of CO observations from the GMD network (black circles) and simulated CO from the RefCOonly simulation (purple stars) averaged over March–August. Blue-green + signs indicate the effect of **(a)** increasing the Asian anthropogenic CO tracer, **(b)** increasing the tropical biomass burning CO tracer, and **(c)** decreasing OH globally by 5 or 10 %, or decreasing NH OH by 20 %.



Figure 4. The impact of increasing the tagged CO tracers for Asian anthropogenic (+), tropical biomass burning (star), Russian biomass burning (diamonds), European anthropogenic (triangles), biogenic (squares), and North American anthropogenic (×) sources on (a) bias in the inter-hemispheric gradient vs. global mean bias and (b) correlation vs. global mean bias compared to GMD observations form March-May. Large circles represent the RefGMI (red), Ref-COonly (black), and COonlySD (purple) simulations. Small circles show the impact of changing global or NH OH in the RefCOonly simulation.



Figure 5. As in Fig. 4, but for June through August.

Figure 6. CO for April 2007 from the GMD observations (circles) overplotted on the surface CO from (a) RefGMI, (b) GMI-HiEmis, (c) RefCOonly, and (d) COonlyLowNHOH simulations.

Figure 7. April 2007 difference between the simulated CO columns and MOPITT for the **(a)** RefGMI, **(b)** GMI-HiEmis, **(c)** RefCOonly, and **(d)** COonlyLowNHOH simulations. The simulated CO is convolved with the MOPITT averaging kernels and a priori.

Figure 8. (a) Annual cycle of surface CO in the RefCOonly simulation (circles) and COonlySD simulation (triangles) for $30-90^{\circ}$ N (black), 30° S -30° N (cyan), and $90-30^{\circ}$ S (magenta). **(b)** Annual zonal mean cross section of the difference in CO between the COonlySD and RefCOonly simulations.

Figure 9. Annual mean tropospheric column ozone (TCO) from the RefGMI (a) and GMI-HiEmis (b) simulations compared to the OMI/MLS TCO product (c) of Ziemke et al. (2011). (d) compares the zonal mean simulated and OMI/MLS values. The simulated values are averaged over the years of the timeslice (1999–2009).

experiment between 900 and 200 hPa, 60° S-60° N.

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Figure 11. Zonal mean surface CO difference compared to RefCO-OH for the CO-OHSensTCO (green), CO-OHSensStO3 (cyan), CO-OHSensQ (blue), CO-OHSensNOx (pink), and CO-OHSensAll (solid black) simulations averaged over March through August of 2005 to 2009. The sum of the differences for the CO-OHSensTCO, CO-OHSensStO3, CO-OHSensQ, and CO-OHSensNOx compared to RefCO-OH is shown in the black dotted line.

