

# 1 **Interpreting Space-Based Trends in Carbon Monoxide** 2 **with Multiple Models**

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## 14 **Abstract**

15 We use a series of chemical transport model and chemistry climate model simulations to  
16 investigate the observed negative trends in MOPITT CO over several regions of the  
17 world, and to examine the consistency of time-dependent emission inventories with  
18 observations. We find that simulations driven by the MACCity inventory, used for the  
19 Chemistry Climate Modeling Initiative (CCMI), reproduce the negative trends in the CO  
20 column observed by MOPITT for 2000-2010 over the eastern United States and Europe.  
21 However, the simulations have positive trends over eastern China, in contrast to the  
22 negative trends observed by MOPITT. The model bias in CO, after applying MOPITT  
23 averaging kernels, contributes to the model-observation discrepancy in the trend over  
24 eastern China. This demonstrates that biases in a model's average concentrations can  
25 influence the interpretation of the temporal trend compared to satellite observations. The  
26 total ozone column plays a role in determining the simulated tropospheric CO trends. A  
27 large positive anomaly in the simulated total ozone column in 2010 leads to a negative  
28 anomaly in OH and hence a positive anomaly in CO, contributing to the positive trend in  
29 simulated CO. These results demonstrate that accurately simulating variability in the  
30 ozone column is important for simulating and interpreting trends in CO.

## 31 **1. Introduction**

32

33 Carbon monoxide (CO) is an air pollutant that contributes to ozone formation and  
34 affects the oxidizing capacity of the troposphere (Thompson, 1992; Crutzen, 1973). Its  
35 primary loss is through reaction with OH, which leads to a lifetime of 1-2 months (Bey et  
36 al., 2001) and makes CO an excellent tracer of long-range transport. Both fossil fuel  
37 combustion and biomass burning are major sources of CO. The biomass burning source  
38 shows large interannual variability (van der Werf et al., 2010), while fossil fuel emissions  
39 typically change more gradually. The time-dependent MACCity inventory (Granier et  
40 al., 2011) shows decreases in CO emissions from the United States and Europe from  
41 2000 to 2010 due to increasing pollution controls, but increases in emissions from China.  
42 MACCity emissions for years after 2000 are based on the Representative Concentration  
43 Pathway (RCP) 8.5 (Riahi et al., 2007). The REAS (Kurokawa et al., 2013) and  
44 EDGAR4.2 (EC-JRC/PBL, 2011) inventories also show increasing CO emissions from  
45 China. The bottom-up inventory of Zhang et al. (2009) shows an 18% increase in CO  
46 emissions from China from 2001 to 2006, and Zhao et al. (2012) estimate a 6% increase  
47 between 2005 and 2009. However, there is considerable uncertainty in bottom-up  
48 inventories, and comparison of model hindcast simulations driven by bottom-up  
49 inventories with observations provides an important test of the time-dependent emission  
50 estimates.

51 Space-based observations of CO are now available for over a decade and show  
52 trends at both hemispheric and regional scales. Warner et al. (2013) found significant  
53 negative trends in both background CO and recently emitted CO at 500 hPa over southern  
54 hemisphere oceans and northern hemisphere land and ocean in Atmospheric Infrared  
55 Sounder (AIRS) data. Worden et al. (2013) calculated trends in the CO column from  
56 several thermal infrared (TIR) instruments including MOPITT and AIRS. They found  
57 statistically significant negative trends over Europe, the eastern United States, and China  
58 for 2002-2012. He et al. (2013) also report a negative trend in MOPITT near-surface CO  
59 over western Maryland.

60 Surface concentrations of CO show downward trends over the United States  
61 driven by emission reductions (EPA, 2011), consistent with the space-based trends.  
62 Decreases in the partial column of CO from FTIR stations in Europe also show decreases  
63 from 1996 to 2006, consistent with emissions decreases (Angelbratt et al., 2011). Yoon

64 and Pozzer (2014) found that a model simulation of 2001 to 2010 reproduced negative  
65 trends in surface CO over the eastern U.S. and western Europe, but showed a positive  
66 trend in surface CO over southern Asia.

67 The cause of the negative trend over China seen in MOPITT and AIRS data is  
68 uncertain. The trend is consistent with the results of Li and Liu (2011), who found  
69 decreases in surface CO measurements in Beijing, and with decreases in CO emissions in  
70 2008 inferred from the correlation of CO with CO<sub>2</sub> measured at Hateruma Island  
71 (Tohjima et al., 2014) and at a rural site in China (Wang et al., 2010). Yumimoto et al.  
72 (2014) used inverse modeling of MOPITT data to infer a decrease in CO emissions from  
73 China after 2007. The 2008 Olympic Games and the 2009 global economic slowdown  
74 led to reductions in CO (Li and Liu, 2011; Worden et al., 2012). However, the negative  
75 trend in MOPITT CO is inconsistent with the rising CO emissions of the MACCity and  
76 REAS inventories. Inverse modeling of MOPITT version 6 data yields a negative trend  
77 in CO emissions from China and a larger global decline in CO emissions than that found  
78 in the MACCity inventory (Yin et al., 2015).

79 This study examines whether global hindcast simulations can reproduce the trends  
80 and variability in carbon monoxide seen in the MOPITT record. We examine the role of  
81 averaging kernels and the contribution of trends at different altitudes to the trends  
82 observed by MOPITT. We then examine the impact of OH variability on the simulated  
83 trends in CO.

## 84 **2. Methods**

### 85 **2.1. MOPITT**

86 The MOPITT instrument onboard the Terra Satellite provides the longest satellite-  
87 based record of atmospheric CO, with observations available from March 2000 to  
88 present. It provides nearly global coverage every three days (Edwards et al., 2004). We  
89 use the monthly Level 3 daytime column data from the Version 5 TIR product, which has  
90 negligible drift in the bias over time (Deeter et al., 2013). The level 3 data is a gridded  
91 product and includes the a priori and averaging kernel for each grid box. Supplemental  
92 Figure S1 shows the MOPITT column averaging kernels averaged over four regions. The  
93 column averaging kernels depend on the observed scene, and vary year to year as well as

94 seasonally. The dependence of the column averaging kernels on the CO mixing ratio  
95 profile (Deeter, 2009) explains the high values in the lower troposphere over eastern  
96 China in winter.

97 We calculate trends and de-seasonalized anomalies for the Eastern U.S., Europe, and  
98 eastern China regions described by Worden et al. (2013). Trends that differ from zero by  
99 more than the two-sigma uncertainty on the trend are considered statistically significant.  
100 We account for autocorrelation of the data for a one-month lag when calculating the  
101 uncertainty on the trends. We calculate the annual cycle by fitting the data with a series  
102 of sines and cosines as well as the linear trend, and then remove the annual cycle to  
103 obtain the de-seasonalized anomalies. Months with no MOPITT data or only a few days  
104 of MOPITT data are excluded from the trend analysis. This includes May-August of  
105 2001 and August-September of 2009. We report the MOPITT trends for 2000-2010 for  
106 comparison with model simulations, and for 2000-2014 to give a longer-term view of the  
107 observed trends.

108

## 109 **2.2. Model Simulations**

110 We use a suite of chemistry climate model (CCM) and chemical transport model  
111 (CTM) simulations to interpret the observed trends. The Global Modeling Initiative  
112 (GMI) CTM includes both tropospheric (Duncan et al., 2007) and stratospheric (Strahan  
113 et al., 2007) chemistry, including over 400 reactions and 124 chemical species.  
114 Meteorology for the GMI simulations comes from the Modern-Era Retrospective  
115 Analysis for Research and Applications (MERRA) (Rienecker et al., 2011). The GEOS-  
116 5 Chemistry Climate Model (GEOSCCM)(Oman et al., 2011) incorporates the GMI  
117 chemical mechanism into the GEOS-5 atmospheric general circulation model (AGCM).  
118 The GEOSCCM simulations are forced by observed sea surface temperatures (SSTs)  
119 from (Reynolds et al., 2002).

120 The Community Earth System Model, CESM1 CAM4-chem, includes 191 chemical  
121 tracers and over 400 reactions for both troposphere and stratosphere (Tilmes et al., 2016).  
122 The model can be run fully coupled to a free-running ocean, with prescribed SSTs, or  
123 with nudged meteorology from GEOS-5 or MERRA analysis. CESM1 CAM4-chem is

124 further coupled to the land model, providing biogenic emissions from the Model of  
125 Emissions and Aerosols from Nature (MEGAN), version 2.1 (Guenther et al., 2012).

126 Several simulations were conducted as part of the Chemistry-Climate Model Initiative  
127 (CCMI) project (Eyring et al., 2013). These include the Ref-C1 simulation of the  
128 GEOSCCM and a Ref-C1 CESM1 CAM4-Chem simulation, hereafter called G-Ref-C1  
129 and C-Ref-C1, respectively, and the Ref-C1-SD simulation of the GMI CTM. Both the  
130 Ref-C1 and the Ref-C1-SD simulations use time-dependent anthropogenic and biomass  
131 burning emissions from the MACCity inventory (Granier et al., 2011), but the Ref-C1-  
132 SD simulations use specified meteorology while the Ref-C1 simulations run with  
133 prescribed SSTs. The MACCity inventory linearly interpolates the decadal  
134 anthropogenic emissions from the ACCMIP inventory (Lamarque et al., 2010) for 2000,  
135 and the RCP8.5 emissions for 2005 and 2010, to each year in between. The MACCity  
136 biomass burning emissions have year-to-year variability based on the GFED-v2 (van der  
137 Werf et al., 2006) inventory. From 2000 to 2010, CO emissions in the MACCity  
138 inventory decreased from 31 to 11 Tg yr<sup>-1</sup> over the eastern U.S., from 97 to 59 Tg yr<sup>-1</sup>  
139 over Europe, and increased from 56 Tg to 72 Tg yr<sup>-1</sup> over eastern China.

140 Given the uncertainty in CO emissions, we conduct a GMI CTM simulation using an  
141 alternative time-dependent emissions scenario, called AltEmis. This simulation is  
142 described in detail in (Strode et al., 2015b). Briefly, anthropogenic emissions include  
143 time-dependence based on EPA (<http://www.epa.gov/ttn/chief/trends/index.html>), the  
144 REAS inventory (Ohara et al., 2007), and EMEP  
145 ([http://www.ceip.at/ms/ceip\\_home1/ceip\\_home/webdab\\_emepdatabase/reported\\_emissiondata/](http://www.ceip.at/ms/ceip_home1/ceip_home/webdab_emepdatabase/reported_emissiondata/)),  
146 and annual scalings from van Donkelaar et al. (2008). Biomass burning  
147 emissions are based on the GFED3 inventory (van der Werf et al., 2010). While the  
148 regional emission trends in this simulation are of the same sign as in the Ref-C1 case, the  
149 magnitude of the negative trends over the U.S. and Europe are smaller and the positive  
150 trend over China is larger, leading to a positive global trend (Fig. 1). We also conduct a  
151 sensitivity study called EmFix with anthropogenic and biomass burning emissions held  
152 constant at year 2000 levels. Table 1 summarizes the simulations used in this study.

153 We regrid the model output to the MOPITT grid and convolve the simulated CO with  
154 the MOPITT averaging kernels and a priori in order to compare the simulated and

155 observed CO columns. The averaging kernels are space and time dependent. We use the  
156 following equation from Deeter et al. (2013):

$$157 \quad C_{\text{sim}} = C_0 + \mathbf{a}(\mathbf{x}_{\text{mod}} - \mathbf{x}_0) \quad (1)$$

158 where  $C_{\text{sim}}$  and  $C_0$  are the simulated and a priori CO total columns, respectively,  $\mathbf{a}$  is the  
159 total column averaging kernel, and  $\mathbf{x}_{\text{mod}}$  and  $\mathbf{x}_0$  are the modeled and a priori CO profiles,  
160 respectively. The column averaging kernel is calculated from the standard averaging  
161 kernel matrix, which is based on the log of the CO concentration profile, following the  
162 method of Deeter (2009):

$$163 \quad a_j = (K / \log_{10}e) \sum \Delta p_i v_{\text{rtv},i} A_{ij} \quad (2)$$

164 where  $\Delta p_i$  and  $v_{\text{rtv},i}$  are the pressure thickness and retrieved CO concentration,  
165 respectively, of level  $i$ ,  $\mathbf{A}$  is the standard averaging kernel matrix, and  $K = 2.12 * 10^{13}$   
166 molec cm<sup>-2</sup> hPa<sup>-1</sup> ppb<sup>-1</sup>.

167 We deseasonalize the simulated CO columns and calculate their linear trend  
168 following the same procedure that we applied to the MOPITT CO. Months that do not  
169 have MOPITT data (June-July 2001 and August-September 2009) are excluded from the  
170 analysis of the model trends as well.

171 The Ref-C1 and Ref-C1-SD simulations requested by CCMI extend until 2010.  
172 However, the MACCity biomass burning emissions extend only until 2008. CAM4-  
173 Chem therefore repeated the biomass burning emissions for 2008 for years 2009-2010.  
174 In contrast, the GEOSCCM Ref-C1 and GMI Ref-C1-SD simulations used emissions  
175 from GFED3 (van der Werf et al., 2010) for years after 2008. Some simulations were  
176 available through 2011, while others ended in 2010. We therefore report results for  
177 2000-2010, but note that extending the analysis through 2011 does not alter the  
178 conclusions.

### 179 **3. Results**

#### 180 **3.1. Trends over Europe, the United States, and the Northern Hemisphere**

181 The hindcast simulations driven by MACCity emissions (G-Ref-C1, Ref-C1-SD, and  
182 C-Ref-C1) show negative trends in CO over the U.S. and Europe that agree with the  
183 observed slope from MOPITT within the uncertainty (Fig. 2, Table 2). The MOPITT

184 trends for both regions are statistically significant for both regions, as shown by Worden  
185 et al. (2013). These results are consistent with the findings of Yin et al. (2015), whose  
186 inversion of MOPITT data showed a posteriori trends in CO emissions over the U.S. and  
187 western Europe that were consistent with but slightly larger than the a priori trends. The  
188 EmFix hindcast shows a positive, though non-significant, trend for both regions,  
189 indicating that the decrease in CO emissions is necessary for reproducing the downward  
190 trend in the CO column. The AltEmis simulation fails to produce the negative trends,  
191 despite including negative trends in regional emissions for both the U.S. and Europe.  
192 The impact of these negative regional trends is insufficient to overcome the positive  
193 global emission trend in the AltEmis scenario (Fig. 1), leading to positive trends in CO.

194 Figure 2 also reveals a negative bias in the simulated CO column between the models  
195 and MOPITT. A low bias in simulated CO at northern latitudes is often present in global  
196 models (Naik et al., 2013), and may indicate a high bias in northern hemisphere OH  
197 (Strode et al., 2015a) or CO dry deposition (Stein et al., 2014), as well as an  
198 underestimate of CO emissions.

199 The deseasonalized anomalies in the MOPITT and simulated CO columns are shown  
200 in Fig. 2b,d, and the correlation coefficient between the observed and simulated monthly  
201 anomalies are presented in Table 2b. The highest correlations are for the AltEmis and  
202 Ref-C1-SD simulations of the GMI CTM. This result is consistent with the use of year-  
203 specific meteorology, which we expect to better match the transport of particular years.  
204 The lowest correlations are for the EmFix simulation. This is expected since the EmFix  
205 simulation does not include inter-annual variability (IAV) in biomass burning. The IAV  
206 in biomass burning makes a large contribution to the IAV of CO (Voulgarakis et al.,  
207 2015).

208 The role of biomass burning in driving the CO variability is even more evident at the  
209 hemispheric scale. Figure 2g,h shows the anomalies in MOPITT and the simulations for  
210 the northern hemisphere (0-60N). The EmFix simulation shows almost no correlation,  
211 while the other simulations have correlation coefficients exceeding 0.6 (Table 2). The  
212 role of changing anthropogenic emissions is also evident, as the Ref-C1-SD simulation  
213 captures the 2008-2009 dip in the CO column while the EmFix simulation does not.  
214 Gratz et al. (2015) found decreasing CO concentrations at Mount Bachelor Observatory

215 in Oregon during spring for 2004-2013, which they attribute to reductions in emissions  
216 leading to a lower hemispheric background. We also note that Ref-C1-SD and G-Ref-  
217 C1 have similar correlations with the observed variability for the northern hemisphere  
218 (Table 2), indicating that transport differences are less important for variability at the  
219 hemispheric scale.

### 220 **3.2. Trend over China**

221 Observations from MOPITT show a negative trend in the CO column over eastern  
222 China for 2002-2012 (Worden et al., 2013). The negative trend for the years 2000-2014  
223 exceeds that for 2000-2010 (Table 2), showing that it is not driven solely by temporary  
224 emission reductions in 2008. Our simulations do not reproduce this trend, and instead  
225 show increases in the CO column (Fig. 2e), which is expected given that CO emissions  
226 from China increase in four of the five simulations. The anomalies (Fig. 2f) show that  
227 the discrepancy in the simulated versus observed trends is driven largely by the failure of  
228 the simulations to capture the 2008 dip in the CO column, leading to an overestimate that  
229 continues through 2010. This suggests emission reductions in China during this time  
230 period are not adequately captured by the emission inventories. However, the good  
231 agreement between the observed and simulated decreases in CO for the northern  
232 hemisphere as a whole (Fig. 2g,h) suggest that on a global scale, the emission time series  
233 is reasonable. Consequently, we examine several other factors that may contribute to the  
234 difference in sign between the MOPITT and simulated CO trends.

235 Regional trends in CO are expected to vary with altitude, with surface concentrations  
236 most heavily influenced by local emissions. MOPITT TIR retrievals have higher  
237 sensitivity to CO in the mid-troposphere than at the surface (Deeter et al., 2004), so the  
238 trend in the MOPITT CO column will be weighted towards the trends in free tropospheric  
239 CO rather than near-surface CO. We quantify this impact on our Ref-C1-SD CO column  
240 trends by comparing the trend in the pure-model CO column with that of the simulated  
241 column convolved with the MOPITT averaging kernels.

242 The simulated CO trend over eastern China for 2000-2010 is positive (but not  
243 significant) both with and without the averaging kernels, but application of the MOPITT  
244 kernels increases the positive trend from  $1.3 \times 10^{16}$  molec cm<sup>-2</sup> yr<sup>-1</sup> to  $1.4 \times 10^{16}$  molec cm<sup>-2</sup>  
245 yr<sup>-1</sup>. This result is initially surprising since we expect trends in the mid-troposphere to be



246 more strongly influenced by the decrease in the hemispheric CO background. Indeed, the  
247 trends in CO concentration over eastern China simulated in Ref-C1-SD switch from  
248 positive in the lower troposphere to negative in the middle and upper troposphere.  
249 However, the application of the kernels results in more positive (or less negative) trends  
250 in all regions.

251 Yoon et al. (2013) show that since the averaging kernels vary over time, a bias  
252 between the true atmosphere and the a priori assumed by MOPITT can lead to an  
253 artificial trend in the retrieved CO. Similarly, the bias between the average simulated CO  
254 concentrations and the MOPITT a priori, evident in Figure 2, can lead to an artifact in the  
255 simulated CO trend when the simulation is convolved with the MOPITT averaging  
256 kernels. This is due to the changing contribution of the a priori when the vertical  
257 sensitivity (averaging kernel) is varying in time. MOPITT vertical sensitivity varies with  
258 time due to instrument degradation as well as the change in CO abundance. The bias in  
259 CO varies with altitude, so if the vertical sensitivity described by the averaging kernel  
260 changes, this will change the value of the convolved CO column even if there were no  
261 changes in the CO profile. Furthermore, changes in the averaging kernel result in more  
262 or less weight placed on the a priori versus the CO simulated by the model. Thus, a  
263 difference between the a priori and the model means that placing more (or less) weight on  
264 the a priori will change the resulting value of  $C_{sim}$ . Since the a priori profiles and  
265 columns are constant in time, taking the time derivative of equation 1 yields:

$$266 \quad \partial C_{sim}/\partial t = \mathbf{a} (\partial \mathbf{x}_{mod}/\partial t) + \partial \mathbf{a}/\partial t (\mathbf{x}_{mod} - \mathbf{x}_0) \quad (3)$$

267 The second term on the right hand side shows that the larger the bias between the  
268 modeled CO and the a priori, the larger the impact of the changing averaging kernel.

269 We quantify this effect by convolving the simulated CO for each year with the  
270 MOPITT averaging kernels for the year 2008, thus removing the effect of the time-  
271 dependence of the averaging kernels. The resulting trend,  $0.56 \cdot 10^{16}$  molec  $\text{cm}^{-2}$   $\text{yr}^{-1}$ , is  
272 less positive than the pure model trend or the original simulated trend. Thus, accounting  
273 for the time-dependence of the averaging kernels convolved with model bias reduces but  
274 does not eliminate the discrepancy with the observed trend. Comparing the trend for the  
275 constant averaging kernel case with the original simulated trend for Ref-C1-SD ( $1.4 \cdot 10^{16}$   
276 molec  $\text{cm}^{-2}$   $\text{yr}^{-1}$ ) suggests that the changing averaging kernels combined with the model

277 bias contribute  $0.84 \times 10^{16}$  molec  $\text{cm}^{-2}$   $\text{yr}^{-1}$  to the simulated trend. Other regions also show  
278 a more negative trend when the same averaging kernel is applied to the model results for  
279 all years. The large bias in CO at middle and high northern latitudes commonly seen in  
280 modeling studies thus impacts the ability of models to reproduce and attribute observed  
281 trends in satellite data.

282 Figure 2 and Table 2 also show a positive trend in the GMI EmFix simulation for  
283 eastern China. This larger trend in the EmFix simulation than the Ref-C1-SD simulation  
284 indicates that the net decrease in emissions contributes to decreasing CO over eastern  
285 China, consistent with the observed negative trend, but other factors in the model cause  
286 an increase in CO over eastern China even when all emissions are constant. Subtracting  
287 the EmFix trend from the Ref-C1-SD trend shows that the changing emissions contribute  
288 a CO trend of  $-0.7$  molec  $\text{cm}^2$   $\text{yr}^{-1}$  over eastern China. The  $2.1$  molec  $\text{cm}^2$   $\text{yr}^{-1}$  trend in the  
289 EmFix simulation, which reflects the impacts of the simulated chemistry and transport,  
290 thus contributes to the erroneous sign of the trend in the GMI simulations. The trends in  
291 the EmFix simulation for the northern hemisphere average and the eastern U.S. and  
292 Europe are positive as well (Table 2). We examine their cause in the next section.

### 293 **3.3. Contribution of OH Interannual Variability**

294

295 Since the EmFix simulation shows a positive trend in the northern hemisphere, we  
296 next examine the variability in the CO sink, OH. We also examine variability in the total  
297 ozone column, since overhead ozone is a major driver of OH variability (Duncan and  
298 Logan, 2008). Figure 3 shows the variability in CO and OH in the EmFix simulation.  
299 The positive and negative anomalies in CO correspond with the negative and positive  
300 anomalies, respectively, in OH. The anomalies in OH are in turn inversely related to  
301 anomalies in the total ozone column. The correlation coefficient between OH and  
302 column ozone is  $-0.53$  for the  $15^{\circ}\text{S}$ - $15^{\circ}\text{N}$  average,  $-0.72$  for the  $15^{\circ}$ - $25^{\circ}\text{N}$  average, and  $-$   
303  $0.75$  for the  $30^{\circ}$ - $60^{\circ}\text{N}$  average. The large NH ozone anomaly in 2010, in particular, leads  
304 to a large anomaly in OH and thus CO. This OH anomaly extends from the northern  
305 tropics to the midlatitudes. The large CO anomaly near the end of the time series  
306 contributes to the apparent 11-year trend. We note that since the lifetime of CO is several

307 months, CO anomalies are not expected to have a one-to-one correspondence with the  
308 OH anomalies.

309 The large anomaly in the simulated total ozone column in 2010 is overestimated  
310 compared to observations. Figure 4 shows the time-dependence of the total ozone  
311 column from 30°-60°N in EmFix compared to SBUV data (Frith et al., 2014). While the  
312 observations show an anomaly in 2010, the magnitude is smaller than that produced by  
313 the simulation. Steinbrecht et al. (2011) attribute the 2010 anomaly in northern  
314 midlatitude ozone observations to a combination of an unusually strong negative Arctic  
315 Oscillation and North Atlantic Oscillation and the easterly phase of the quasi-biennial  
316 oscillation.

317 While the impact of OH interannual variability on the apparent trend in CO is clear in  
318 the EmFix simulation, this source of variability is partially masked by large interannual  
319 variability in CO emissions in the other simulations. We examine the correlation  
320 between the de-trended and deseasonalized CO anomalies from 10°S-10°N in the Ref-  
321 C1-SD simulation and the CO emissions as well as the simulated OH and column ozone.  
322 Since the CO emitted in a given month can influence concentrations for several  
323 subsequent months, we use a 3-month smoothing of the emission time series. We find a  
324 high correlation ( $r=0.88$ ) between the CO anomalies and the CO emissions. This  
325 correlation is also evident in the MOPITT data, as the MOPITT CO anomalies have a  
326 correlation of  $r=0.70$  with the emissions. Figure 5 shows the strong relationship between  
327 the simulated CO anomalies and the CO emissions. However, the colors in Fig. 5  
328 indicate that the scatter for a given level of emissions is often linked to the OH  
329 anomalies, with low/high OH anomalies leading to CO that is higher/lower than would be  
330 predicted just from the CO emissions. We find that the 10°S-10°N OH in the Ref-C1-SD  
331 simulation is anticorrelated with CO ( $r=-0.62$ ) and with the total ozone column ( $r=-0.68$ ).  
332 Consequently, the simulated ozone column plays a role in modulating tropical CO  
333 variability even when variable CO emissions are included, although the emissions still  
334 play the strongest role.

335 **4. Conclusions**

336 We conducted a series of multi-year simulations to analyze the causes of the negative  
337 trends in MOPITT CO reported by Worden et al. (2013). Both CTM and CCM  
338 simulations driven by the MACCity emissions reproduce the observed trends over the  
339 eastern U.S. and Europe, providing confidence in the regional emission trends.

340 None of the simulations reproduce the observed negative trend over eastern China.  
341 This negative trend persists even with the MOPITT data extended out to 2014. The  
342 MOPITT averaging kernels are weighted towards the free troposphere, where the relative  
343 importance of hemispheric versus local trends is greater. However, our simulations  
344 indicate that this effect is insufficient to explain the negative trends over China. Indeed,  
345 the negative trend in MOPITT CO over eastern China ( $-2.9 \times 10^{16}$  molec cm<sup>-2</sup> yr<sup>-1</sup>) is  
346 stronger than that of the northern hemisphere average ( $-1.4 \times 10^{16}$  molec cm<sup>-2</sup> yr<sup>-1</sup>),  
347 indicating that changes in hemispheric CO account for less than half of the trend over  
348 China. While the simulations' underestimate of the observed trend likely indicates a too  
349 positive emission trend for China, several other factors play a role in the model-  
350 observation mismatch. We find that the time-dependent MOPITT averaging kernels,  
351 combined with the low bias in simulated CO, provides a positive component to the  
352 simulated trends. Large anomalies in the simulated ozone column in the GMI CTM  
353 simulations also contribute a positive component to the northern hemisphere trends due to  
354 their impact on OH. For the Ref-C1-SD simulation, the trends due to the model bias  
355 combined with changing averaging kernels ( $0.84 \times 10^{16}$  molec cm<sup>-2</sup> yr<sup>-1</sup>) and to the  
356 simulated chemistry and transport ( $2.1 \times 10^{16}$  molec cm<sup>-2</sup> yr<sup>-1</sup>) can together account for  
357 almost 70% of the  $4.3 \times 10^{16}$  molec cm<sup>-2</sup> yr<sup>-1</sup> difference between the Ref-C1-SD and  
358 MOPITT trends over eastern China.

359 Variability in emissions is the primary driver of year-to-year variability in simulated  
360 CO, but OH variability also plays a role. The simulated OH is anti-correlated with both  
361 CO and the total ozone column, highlighting the importance of realistic overhead ozone  
362 columns for accurately simulating CO variability and trends. In addition, further work is  
363 needed to understand recent changes in CO emissions from China.

364

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**Table 1:** Description of Simulations

Simulation	Model	Meteorology	Anthropogenic Emissions	Biomass Burning Emissions
G-Ref-C1	GEOSCC M	internally derived	MACCity	MACCity, GFED3 (2009-2010)
C-Ref-C1	CAM4- Chem	internally derived	MACCity	MACCity, then repeat 2008
Ref-C1- SD	GMI	MERRA	MACCity	Same as GEOSCCM
EmFix	GMI	MERRA	Fixed at 2000	Fixed at 2000
AltEmis	GMI	MERRA	Strode et al [2015]	GFED3

**Table 2: Regional Trends and Correlations****a. Trends<sup>1,2</sup>**

	Years	E. USA	Europe	E. China	N. Hemisphere
G-Ref-C1 <sup>3</sup>	2000-2010	-2.2 (0.38)	-1.8 (0.42)	2.2 (1.1)	-0.76 (3.0)
C-Ref-C1 <sup>3</sup>	2000-2010	-3.4 (0.54)	-2.9 (0.50)	1.4 (1.4)	-0.90 (3.0)
Ref-C1-SD <sup>3</sup>	2000-2010	-2.4 (0.53)	-1.6 (0.59)	1.4 (1.1)	-0.76 (3.0)
EmFix <sup>3</sup>	2000-2010	1.3 (0.55)	1.5 (0.44)	2.1 (0.87)	0.96 (2.5)
AltEmis <sup>3</sup>	2000-2010	0.71 (0.73)	0.74 (0.66)	3.8 (1.4)	1.1 (3.4)
<i>MOPITT</i>	<i>2000-2010</i>	<i>-2.5 (0.64)</i>	<i>-1.8 (0.69)</i>	<i>-2.9 (1.8)</i>	<i>-1.4 (2.8)</i>
<i>MOPITT</i>	<i>2000-2014</i>	<i>-2.1 (0.41)</i>	<i>-1.7 (0.43)</i>	<i>-3.1 (1.1)</i>	<i>-1.4 (1.7)</i>

<sup>1</sup>10<sup>16</sup> molec cm<sup>-2</sup> yr<sup>-1</sup>

<sup>2</sup>1-sigma uncertainty given in parentheses

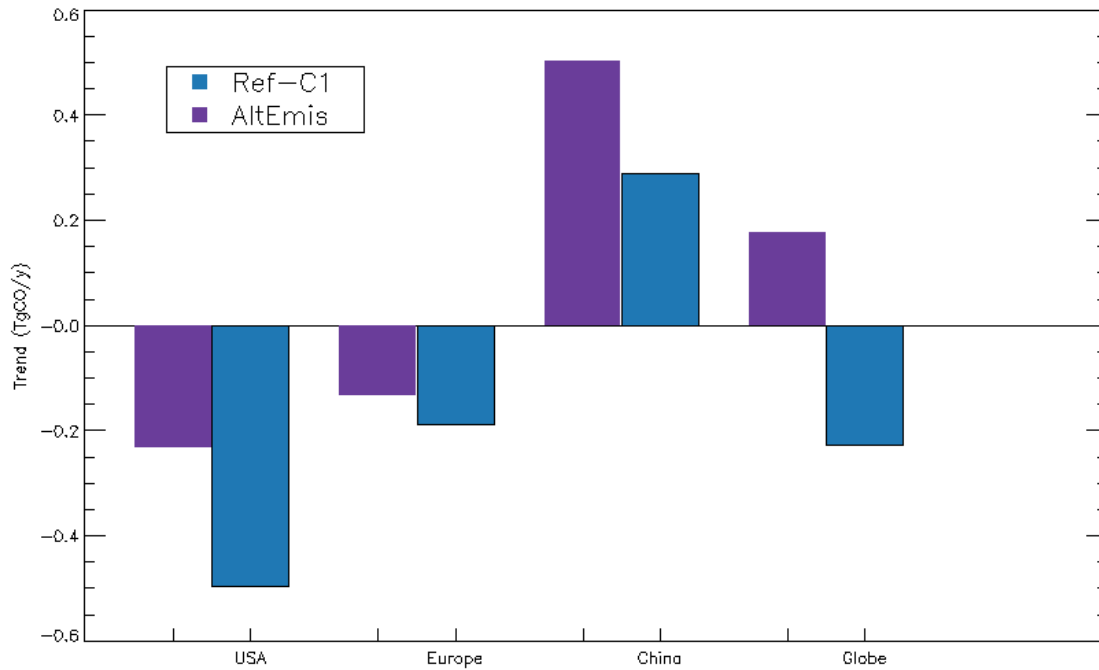
<sup>3</sup>Simulation results convolved with MOPITT averaging kernel and a priori

**b. Correlation coefficient (r) with monthly MOPITT anomalies<sup>1,2</sup>**

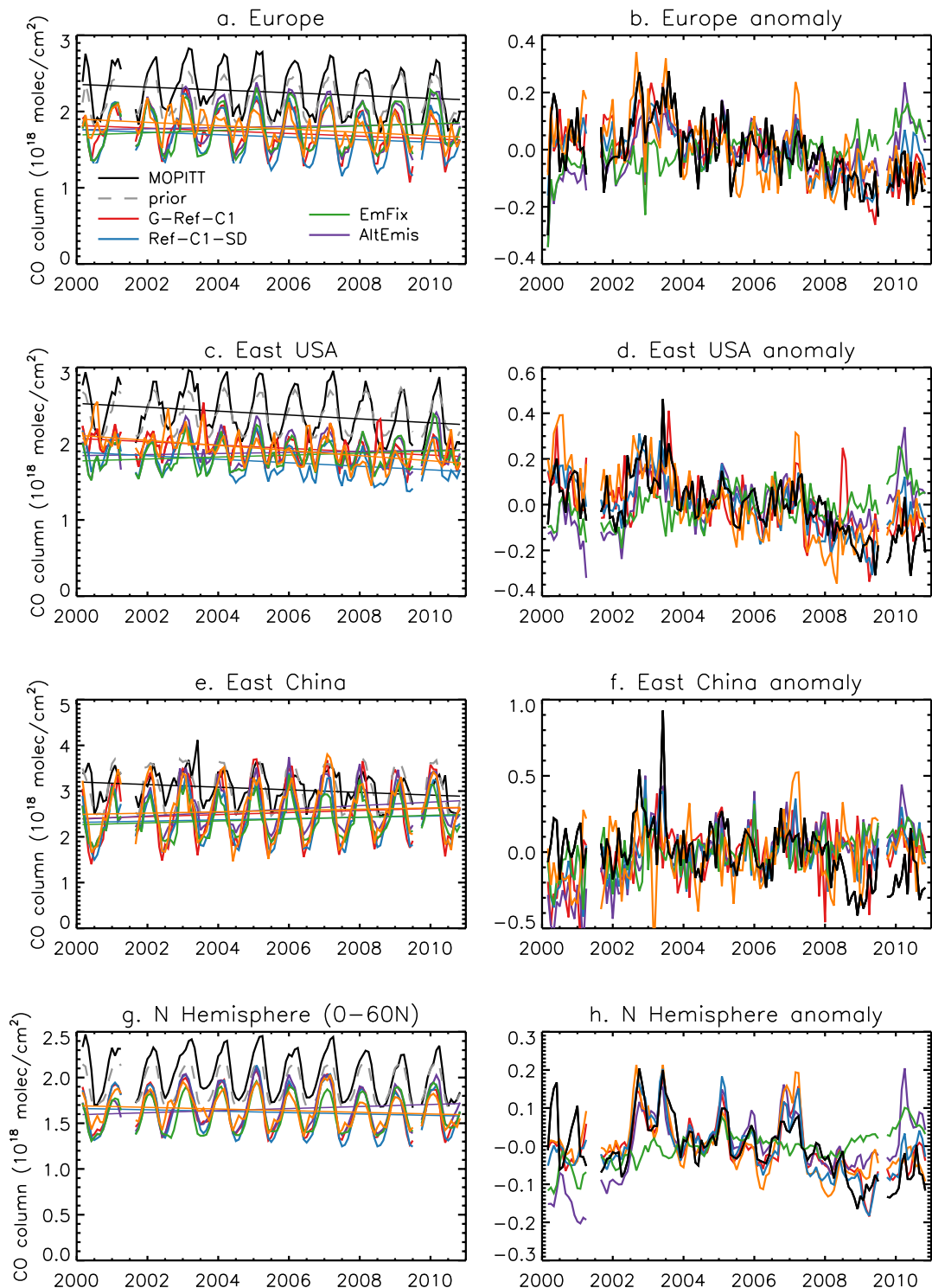
	Years	E. USA	Europe	E. China	N. Hemisphere
G-Ref-C1	2000-2010	<b>0.26</b>	<b>0.39</b>	0.061	<b>0.71</b>
C-Ref-C1	2000-2010	0.23	<b>0.36</b>	0.18	<b>0.62</b>
Ref-C1-SD	2000-2010	<b>0.43</b>	<b>0.51</b>	<b>0.39</b>	<b>0.73</b>
EmFix	2000-2010	0.10	0.21	0.071	0.059
AltEmis	2000-2010	<b>0.55</b>	<b>0.59</b>	<b>0.48</b>	<b>0.69</b>

<sup>1</sup>Correlations are calculated from the de-trended and de-seasonalized time series.

<sup>2</sup>Statistically significant correlations at the 95% confidence level are indicated in bold.

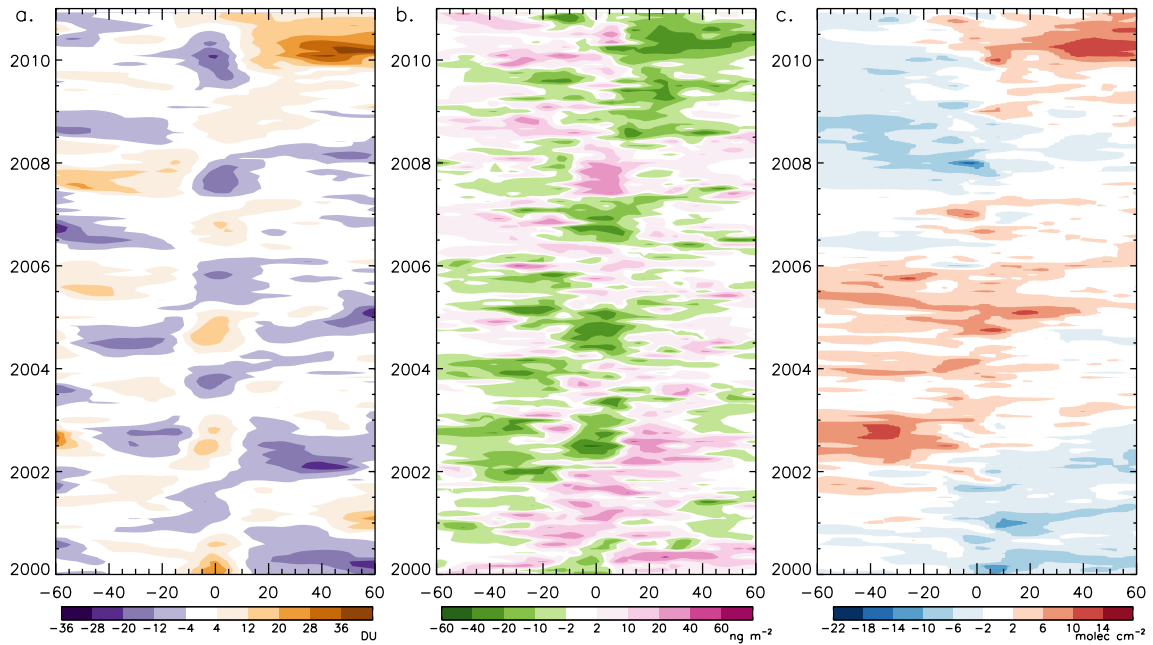


**Figure 1:** Trends in the CO emissions used in the Ref-C1 and Ref-C1-SD simulations (blue bars) and AltEmis simulation (purple bars) over 2000-2010 for the United States, Europe, China, and the world.

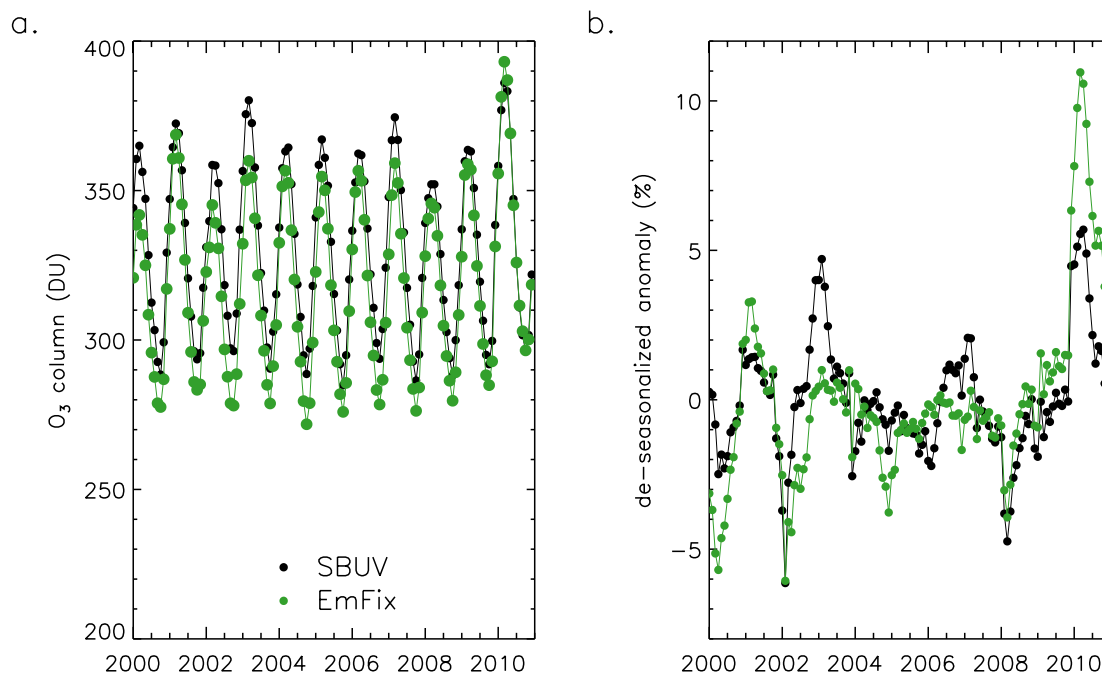


**Figure 2:** The time series and trends (left column) and de-seasonalized monthly anomalies (right column) of the CO column from MOPITT (black), the MOPITT a priori (gray), and simulated by G-Ref-C1 (red), Ref-C1-SD (blue), EmFix (green), C-Ref-C1 (orange), and AltEmis (purple) for 2000-2010. The regions shown are (a,b) Europe (0°-

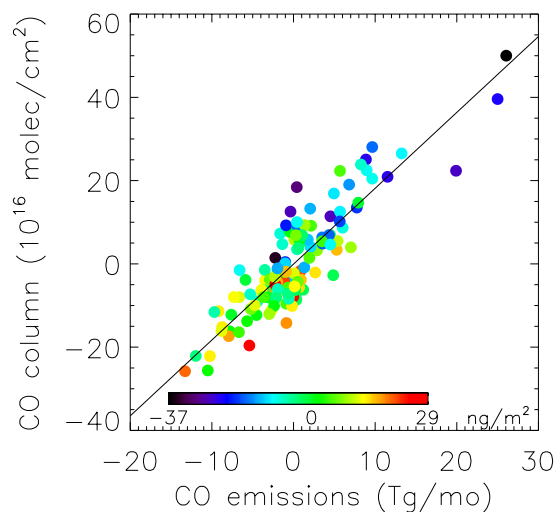
15°E, 45°-55°N), (c,d) eastern U.S.A. (95°-75°W, 35°-40°N), (e,f) eastern China (110°-123°E, 30°-40°N), and (g,h) the northern hemisphere (0°-60°N).



**Figure 3:** Deseasonalized monthly anomalies in the total ozone column (left), mean tropospheric OH (center), and CO column (right) from the EmFix simulation as a function of latitude and month.



**Figure 4:** Monthly ozone column (a) and de-seasonalized ozone column anomaly (b) in SBUV data (black) and the EmFix simulation (green) for 30°-60°N.



**Figure 5:** Monthly simulated CO column anomalies from the Ref-C1-SD simulation as a function of CO emissions for 10°S-10°N. Colors indicate the simulated OH column anomaly for the given month.