1 **Author's response to referee 1:**

2 Thank you very much for your supportive and precious comments! You helped us significantly improve

3 this study.

4 1) Comments: Abstract The time step of the simulation in this study is monthly. Time step of the

5 calculation is very important. So, please explicit describe about the time step of the calculation even in

6 abstract.

7 **Response: We have added the information in Abstract: "We develop a process-based biogeochemistry**

8 **model to quantify CO exchange between soils and the atmosphere with a 5-minute internal time step**

9 **at the global scale. The model is parameterized using CO flux data from the field and laboratory**

10 **experiments for eleven representative ecosystem types. The model is then extrapolated to the global**

11 **terrestrial ecosystems using monthly climate forcing data." From line 10 to line 15.**

12 2) Comments: Page 2 line 31 I do not think that this sentence is necessary.

13 **Response: Deleted.**

14 3) Comments: Page 5 line 118 are in -> is

15 **Response: Corrected. "The first study to report long-term and continuous field measurements of CO**

16 **flux over grasslands using a micrometeorological eddy covariance (EC) method is Pihlatie et al. (2016)."**

17 **From line 93 to line 95.**

18 4) Comments: Page 5 line 127 withan-> with an

19 **Response: Corrected. "A set of century-long simulations of 1901-2100 were also conducted using the**

20 **atmospheric CO concentrations estimated with an empirical function (Badr & Probert, 1994; Potter et**

21 **al., 1996)". From line**

22 5) Comments: Page 6 line 139 Please spell out TEM at the first place.

23 **Response: We have mentioned at the first place of introduction. "To improve the quantification of the**

24 **global soil CO budget for the period 2000-2013 and CO deposition velocity for the 20th and 21st**

25 **centuries, this study developed a CO dynamics module (CODM) embedded in a process-based**

26 **biogeochemistry model, the Terrestrial Ecosystem Model (TEM) (Zhuang et al., 2003, 2004, 2007)."**

27 **From line 96 to line 99.**

28 6) Comments: Page 6 line 157 Vertical grid of 1cm can be used for simulation of CO_2 and CH_4 diffusion

29 processes in soil, but, I consider that 1cm is still not so finely-gridded for the simulation to model soil CO

30 consumption because of rapid CO consumption in soil (especially very active soil to consume CO). Some

31 soils are strong consumers of CO and these soils absorb CO within 2-3cm of top soil layer. Though this

32 comment does not deny robustness of the results of this study, I recommend that these technical

33 aspects should be mentioned in the Discussion section (as in line 376) to be kind for readers who may

34 study soil CO consumption. Furthermore, authors properly used implicit (Crank-Nicolson) method in

35 order to be independent from time-step which must be set as short as possible in case of explicit

36 method because the vertical grid must be finer for soil CO consumption for explicit method.

37 **Response: Thanks much for your suggestions. In this revision, we have tested the model using 3, 15,**

38 **30, 300, 3000 thin layers to examine the influence of layer thickness. It turned out that we have**

39 **chosen the proper layers division and more layers will need much more computing time, but not show**

40 **further improvement. We have summarized these tests in Figure 12 and Section 4.3, line 452 to 460.**

41 7) Comments: Page7 line 160 "i" and "t" should be italic.

42 **Response: Corrected. "Where $C(t, i)$ is the CO concentration in layer i and at time t, units are mg m⁻**

43 **³." Line 142.**

44 8) Comments: Page 12 line 297 Figure 3 (a2,b2,c2,d2) I cannot understand which observations in Table 1

45 were plotted in the sub-figures.

46 **Response: We added information to indicate the site being used in Figure 3 caption.**

47 9) Comments: Page 12 line 305- Page 13 Direction of consumption and production or net flux is

48 misleading. I felt that minus expression of values is difficult to see through.

49   **Response: Thanks for pointing this out. We have changed all values presented as ranges like " From -**

50   **180 to -197, 34 to 36 and -145 to -163 Tg CO yr^{-1}".**

51   10) Comments: Page 16 Model Uncertainties and Limitations I consider that CO concentrations at soil

52   surface (environmental CO concentration for soil) is a little different from CO data from MOPITT.

53   Boundary-layer processes are also complex. A comment about this point is necessary.

54   **Response: We have revised Section 4.3 to address your comments. ". Third, the derived CO surface**

55   **concentration is lower than MOPITT CO surface concentration, which will lead to overestimation of**

56   **CO deposition velocity during 1901-2100." From Line 450 to 452.**

57   11) Comments: Table2-6 Values were centered but should be formatted to be easily understood. For

58   example, Earea/R (K) in Table 2 In place of 8801 14165, 8801 14165 is better.

59   **Response: We have now centered all values and names of parameters.**

60   12) Comments: Figure2 Units of soil moisture are different among (a2), (b2) (c2) and (d2). My concern is

61   about the highness of the volumetric soil moisture. The soil moistures in (a2) (d2) are too high?

62   Volumetric soil moisture contents (a2) are too high, as high as 80%. Normal soils have no capacity to

63   hold such high moistures. The units of the soil moisture contents are all volumetric (m3/m3) ? Please

64   check and if the shown volumetric soil moistures are correct, please mention reasons.

65   **Response: Thank you for pointing this out. In this revision, we traced back to Nakai et al. (2013) and**

66   **found that our units and values are the same as they presented. The reason why the values were so**

67   **high is that the volumetric soil moisture (VSM) was converted from the water content reflectometry**

68   **(WCR) probe output period using an empirical calibration function of Bourgeau-Chavez et al. (2012)**

69   **for 5cm-30cm layer. Although Bourgeau-Chavez et al. (2012) provided calibration functions for each**

70   **soil horizon (i.e., dead moss, upper duff, lower duff, and mineral soil), some of them resulted in values**

71   **greater than 100% VSM in Nakai et al. (2013) study. The model estimated high VSM (close to 80%) is**

72 **due to top 10 cm moss in the model which has a saturation VSM of 0.8.   We added the discussion on**

73 **Figure 2 caption in this revision. From line 482 to 485.**

74 13) Comments: Figure 3 (c2) and (d2) Why authors showed over-scale (CO emission) graph? The y-axis of

75 (c2) should be -10 to 2

76 **Response: We have corrected the Y-axis's range to -10 to 2 in Figure 3 (c2).**

77 14) Comments: Figure 4 Please write clear the meaning of "global land surface". Global land surface

78 includes Antarctic area? Normal readers think that global average temperature is about 15C but the

79 shown temperature (a) is between 7.5-9C.

80 **Response: We have added extra information in caption of Figure 4 and Figure 6: "Global land surface**

81 **(excluding Antarctic area and ocean area)"**

82 15) Comments: Figure 6 Why SOC increases sharply before 2100?

83 **Response: In this revision, we have fixed this problem. The fixed values of SOC is showed in figure 6.**

84 **We also rerun the model to remove the influence of odd SOC to future prediction of CO dynamics.**

85

Author's response to referee 2:

Thank you for your constructive comments, which helped us improve this study. Here are our responses to each of your comments:

1) Main Concern: CO concentration and flux mismatch between MOPITT and constant function simulations The CO soil deposition is largest flux and is strongly dependent on the atmospheric CO mole fraction. The constant CO mole fractions used in past and future model runs are not realistic, at least for the period 2000-2013, as shown by the large mismatch in magnitude with the MOPITT satellite data for the overlapping period (Fig . 5b).

Correspondingly, the annual fluxes calculated over 1901-2013 using the constant CO distribution (determined by the function shown at lines 263) are very different from the fluxes calculated using the CO MOPITT data for the shorter period 2000-2013.

The authors report both sets of incompatible fluxes as results. Assuming that the satellite measured values are close to the truth, then the fluxes calculated using the constant function, for the 20th and 21st centuries, are obviously wrong. However, these are reported as main results and the authors claim to "quantify global soil budget for the 20th and 21st centuries".

In my opinion these fluxes should not be reported in the actual form. They can however be used to study the variability and the relative magnitude of the components, and the relative variation in time.

If the exercise is only for understanding the controls and the relative evolution (as the authors state in their reply to reviewer 2), this can be accepted as long as the limitations are made clear and the fluxes are not claimed to be solved.

Here are some ideas on what can be done:

- scale the CO distribution function in such a way that it matches the satellite observations for the overlapping period. This would need an assumption for the temporal evolution of CO during the 20th century

110   - report the results not in terms of fluxes, but in terms of net deposition velocities. This would require

111   changes through the paper.

112   - keep the fluxes as they are, but do not discuss the absolute values of the fluxes but only the relative

113   variations

114   Besides these, the authors should be careful with claiming that they quantify the soil budget for the 20th

115   and 21st centuries.

116   This issue should be discussed thoroughly in Sect. 4.3.

117   **Response: We highly appreciate your constructive suggestions and comments. We have chosen your**

118   **second recommendation and changed historical simulation (1901-2013) and future simulation (2014-**

119   **2100) results to be presented as deposition velocity by using the method in Seinfeld, et al. (1998). In**

120   **this revision, what we have done include: 1) We only discussed absolute values of simulations during**

121   **the period 2000-2013 when using MOPITT CO surface concentration data; 2) We removed all parts**

122   **that involve absolute values of CO consumption, production and net fluxes; 3) We have plotted out**

123   **new figures to present deposition velocity for 1901-2100; 4) We have re-organized the paper with a**

124   **clearer experiment orders: E1: simulations during the period 2000-2013 using MOPITT CO surface**

125   **concentration data; E2: simulations during the period 1901-2100 using constant in time, spatially**

126   **distributed CO surface concentration data; E3: sensitivity tests; and 5) We added discussion of CO**

127   **concentration influences on CO dynamics in Section 4.3, line 450 to line 452.**

128

129   **Other general comments:**

130   2) - Abstract: please report the same values given in the paper, either averages or ranges.

131   **Response: We have used ranges now throughout the paper.**

132   3) - please try to organize the paper in smaller paragraphs to improve readability

133 **Response: We have reorganized a number of paragraphs to get them smaller. In addition, we have**

134 **also reduced many repeated references in Introduction.**

135 4) - I find the Introduction up to line 60 difficult to read, because of the long list of references following

136 each bit of information. Please consider rephrasing and reducing the references that keep being

137 repeated.

138 **Response: We have significantly revised the Introduction section and also removed repeated or non-**

139 **necessary references. Please see line 33 to line 52.**

140 5) - The summary of the experiments is given both in 2.1 and 2.5, but the two descriptions do not seem

141 to match. In 2.1. it is mentioned that the purpose was to "investigate the impact of …atmospheric CO

142 concentrations "but the sensitivity tests that are meant for this are not mentioned here.

143 **Response: We have corrected this and now they describe the same experiments. See Section 2.1 and**

144 **Section 2.5.**

145 6) - is Michaelis-Menten kinetics necessary? The net uptake means in principle that at least the

146 uppermost layer of soil has lower concentration than the atmosphere, and this is much lower than kCO.

147 Are the CO concentrations in the lower soil layer much larger, and if yes, why? Related to this, please

148 show the CO soil profiles for some typical situations.

149 **Response: In this revision, we have re-considered the Michaelis-Menten kinetics and presented soil**

150 **CO concentration profile in figure 12. We found that the Michaelis-Menten kinetics can't be replaced**

151 **in our model since the soil CO concentration can be bigger than atmospheric CO surface concentration**

152 **in the days of net emissions (Figure 12f). Replacing it with a simple version will overestimate the**

153 **oxidation rate when soil CO concentration is comparable with atmospheric CO concentration. We**

154 **have discussed this in Section 4.3. From line 460 to line 465**

155 7) - please number the figures in the order they are discussed in text (e.g. Figs 7, 8) – or change the text

156 to mention the figures in the right order.

157   **Response: We have carefully checked the order of figures and their uses in main text.**

158   8) - Section 4.2 should be reorganized and at least partly moved to results – see specific comments

159   **Response: We have moved sensitivity test results to Section 3.4 and we have reorganized 4.2 based**

160   **on specific comments. See section 3.4 from line 361 to line 369, and section 4.2 from line 397 to line**

161   **408.**

162   9) - Section 4.3 discussed the model uncertainties and limitations, but ignores two of the major issues: (1)

163   the CO concentrations mismatch between satellite and constant function, and (2) the overestimation of

164   soil consumption at high temperatures.

165   **Response: We have added these two points in Section 4.3 in this revision. See line 450 to 452 and line**

166   **441 to line 443 for issue 1) and issue 2), respectively.**

167

168   **Specific comments**

169   10) - line 16: "constant spatially distributed" – can be understood wrongly as constant in space, which is

170   not; I suggest to change to " constant in time, spatially distributed"

171   **Response: We have rephrased it following your suggestion. "By assuming that the spatially-**

172   **distributed atmospheric CO concentrations (~128 ppbv) are not changing over time". From line 21 to**

173   **line 23.**

174   11) - line 20: "the largest sinks at 93 Tg CO yr-1" – is this from the 20th century or 2000-2013 simulation?

175   Please specify. Same for the next phrase.

176   **Response: This value is for 2000-2013 simulation. We corrected. See line 19.**

177   12) - lines 63 and 68: use the same units for the deposition velocity

178   **Response: Corrected. Now units for deposition velocity are mm s^{-1} throughout the paper manuscript.**

179 13) - lines 61-68: the phrase is unclear, consider breaking it into smaller pieces. Also, quite some

180 information is given on the studies listed, but not enough to actually understand what they did and what

181 the differences are. Please consider either giving more details, or shortening this part.

182 **Response: We have shortened this part to increase readability and avoid misleading. See line 53 to**

183 **line 57.**

184 14) - lines 62 – 66: the text seems contradictory: the uptake flux when using a constant deposition

185 velocity globally (115-230 Tg) is smaller than when using the same deposition velocity in general and

186 some area set to zero (300 Tg) - please explain or reformulate. "With different approaches" is too

187 unspecific and does not really say anything.

188 **Response: We have removed this for shortening the paragraph and reducing confusing. See line 57 to**

189 **line 59.**

190 15) - line 67: " using empirical approaches with higher probability for lower values" – unclear, higher

191 probability of what? lower values for what?

192 **Response: This part has been removed in this revision. See line 57 to line 59.**

193 16) - lines 68 – 70: which other substances? what are other deposition velocities? please give examples

194 if you mention this.

195 **Response: We have rewritten this sentence and now just presented the ranges for all vegetation types.**

196 **Detailed deposition velocity for different vegetation types can be found in King (1999a) and**

197 **Castellanos et al. (2011). See line 57 to line 59.**

198 17) - lines 75 – 91: a lot of this discussion on the thermal and photo degradation is irrelevant for this

199 paper, especially the part on photo degradation which is not included in the model. Please reduce the

200 irrelevant parts.

201 **Response: We have reduced the part related to photo-degradation. Now we only mentioned what is**

202 **photo-degradation. See line 63 to line 64.**

203 18) - lines 92 – 93: it is unclear to me what this phrase means. I think what the authors may intend to say

204 is that little attention has been paid to CO (including soil consumption and production) in global

205 (chemistry?) models. Please reformulate if true.

206 **Response: We have removed the sentence. See line 70.**

207 19) - lines 97-98: suggest replacing "oxidation from soil bacteria and microbes" by "oxidation by soil

208 microbes". Bacteria are microbes.

209 **Response: Corrected. See line 74 to line 75.**

210 20) - line 110: CO emission is abiotic, right?

211 **Response: We have removed the "CO emission" from the sentence. "One reason is that there is an**

212 **incomplete understanding of biological processes of uptake". See line 85 to line 86.**

213 21) - line 156: "determined by the mass balance" – unclear what this means: which mass balance, of

214 what, between what? please clarify.

215 **Response: We have added the detailed description: "Net exchange of CO between the atmosphere**

216 **and soil is determined by the mass balance approach (net flux = total production – total oxidation –**

217 **total soil CO concentration change)."**

218 22) - line 162: this term represents all the consumption; remove "due to oxidation"

219 **Response: Removed "due to oxidation" from the sentence. See line 144.**

220 23) - line 171: "modeled as an anaerobic process" there is nothing in Eq. 2 that makes it anaerobic

221 **Response: We have rewritten the sentence and removed "modeled as an anaerobic process" from**

222 **description: "CO consumption is modeled in unsaturated soil pores", see line 153**

223 24) - lines 186- 188: phrase unclear - I think the authors mean that Eq 2.2 will overestimate CO

224 consumption because in reality CO consumption decreases at high temperatures, while in Eq 2.2 CO

225 keeps increasing with temperature. Please reformulate.

10

226 **Response: We have reformulated the sentence. "Equation (2.2) will overestimate CO consumption at**

227 **higher temperature because in reality CO consumption will decrease at higher temperatures than**

228 **optimum temperature, while f_2 will keep increasing with rising temperature." See line 171 to 174.**

229 25) - line 193: i do not understand what Pr(t,i) is

230 **Response: We have added the detailed description: "production rate at temperature $T(t, i)$ divided**

231 **by production rate at reference temperature". $P_r(t, i)$ is also described in equation (3.1) and the**

232 **paragraph right below. See line 181 to line 182 and line 191 to line 192.**

233 26) - line 198: should it be 20 cm SOC?

234 **Response: Corrected to "30 cm". See line 186.**

235 27) - lines 275 – 277: give some details on the scenarios and datasets

236 **Response: We have added details of meaning of RCP2.6, 4.5 and 8.5. "RCP2.6, 4.5 and 8.5 datasets are**

237 **future climate projections with anthropogenic greenhouse gas emission radiative forcing of 2.6 W m^{-2},**

238 **4.5 W m^{-2} and 8.5 W m^{-2}, respectively, by 2100." See line 278 to line 280.**

239 28) line 297: is the reported correlation really *r*, or is it *r^2*? Also, a correlation coefficient *r* of 0.5 is

240 usually not considered high correlation.

241 **Response: Correlation coefficient is reported in R. We have rewritten the sentence to give proper**

242 **description now. See section 3.1, line 303 to line 304.**

243 29) - lines 299 - 300: please compare the RMSEs reported to the CO fluxes, in order to give an

244 impression on the relative errors.

245 **Response: We have presented CO net flux rates in order to compare with RMSEs. Please find in**

246 **Section 3.1, line 305.**

247 30) - lines 318 – 319: "consume 42% and 58% of the total consumption, and produce 41% and 59% of

248 total production" – please reformulate

11

249 **Response: We have changed Section 3.2 only for the period 2000-2013. We have also fixed this by**

250 **reformulating. "The Southern and Northern Hemispheres have 41% and 59% of the total consumption,**

251 **and 47% and 53% of the total production, respectively (Table 3)". See line 321 to 323.**

252 31) - line 328: Table 3 does not show the deposition flux in mg/m2 day

253 **Response: We have changed section 3.2 only for the period 2000-2013. The table has been updated to**

254 **only have CO flux values for the period 2000-2013 with MOPITT CO surface concentration data and CO**

255 **deposition velocity for the period 1901-2013 with constant in time, spatially distributed CO surface**

256 **concentration. We have added the description on how to get these numbers. "calculated by flux**

257 **values divided by area". See section3.2, line 336 to line337**

258 32) - lines 329 – 343: I find the text in this paragraph somewhat misleading. The fluxes are presented as

259 changing or increasing relative to the simulation for 20th century, which suggests a temporal evolution,

260 but in fact they are different mainly because of a different model setup, i.e different atmospheric CO

261 concentrations. Consider using "different" and "larger" instead of "increasing".

262 **Response: We have removed this part and only talked about simulation results during 2000-2013 in**

263 **section 3.2. We moved 1901-2013 deposition velocity simulation results to Section 3.3.**

264 33) - line 359: "the rate ranges of increasing of consumption…" – I think it should be something like "the

265 ranges of the rates of increase in consumption…"; please explain what these ranges are, are they

266 corresponding to the three scenarios?

267 **Response: We have changed the results in Section 3.3 to present deposition velocity and added the**

268 **results of 1901-2013. We have rewritten this sentence and added more information to explain what**

269 **the ranges are. See section 3.3, line 341 to line 343.**

270 34) - line 382: the references should be given in the method section, not here

271 **Response: Removed the references here. See section 4.1, line 390.**

12

272   35) - Sect 4.2: The information here belongs mostly to Results, please reorganize. Also, the section is

273   hard to follow – there are many correlations mentioned without much coherence. Consider

274   reformulating into a more focused way, with one paragraph per idea (e.g on annual time scales, the CO

275   uptake is mostly correlated to X, Y, Z… and then comment more if needed on X). Try to separate the

276   annual and monthly results.

277   **Response: We have 1) moved the sensitivity results to new Section 3.4; 2) reorganized the rest part by**

278   **totally separating the monthly and annual correlation test results. See Section 3.4, line 362 to line 369**

279   **and Section 4.2, line 409 to line 430.**

280   36) - lines 398 – 400 and Table 5: The effect of the SOC on the gross uptake flux seems too large. In my

281   understanding, the text tries to explain that SOC increases the gross production which makes more CO

282   available, which in turn leads to an increase in the gross CO consumption. But, for an increase in SOC of

283   30%, the production increases by about 10Tg/year, and the consumption increases by 28 Tg/year! How

284   can that be? Where are the extra 18 Tg/year coming from?

285   **Response: We have added extra comments on this problem in discussion section 4.2. "To be noticed,**

286   **the CO oxidation increasing due to CO substrate change is larger than production increasing due to**

287   **SOC increasing, leading to an extra 18 Tg CO yr^{-1} being taken up from the atmosphere to soils in**

288   **sensitivity test when SOC increasing by 30% (Table 5)." See line 403 to 406.**

289   37) line 406: "as CO flux" – do you mean "as does the CO flux"?

290   **Response: This sentence has been removed now.**

291   38) line 415: what is 0.91? Is it r or r^2, or something else? The same for line 418.

292   **Response: We have added description. They are R. See section 4.2, line 424 and line 426.**

293   39) - line 425: the same data limitation is when using any method, not only SCE-UA-R, correct? If yes,

294   remove "using SCE-UA-R method"

295 **Response: Yes. The data limitation is same to any method. We removed the "using SCE-UA-R method".**

296 **See section 4.3, line 434 to line 437.**

297 40) - line 427: "with RMSE … day-1" – I think this info has no meaning here

298 **Response: We have removed "with RMSE … day^{-1}" now. See section 4.3, line 437.**

299 41) - Table 6: what are the numbers? what are the units? These are not absolute values of fluxes.

300 **Response: They are correlation coefficient (R) between forcing variables and model results. We have**

301 **rewritten the description of Table 6 for better understanding. See table 6.**

302 42) - line 796: I suggest to replace "would happen inside" by "take place"

303 **Response: We have changed to "take place". See figure 1 caption, line 810.**

304 43) - Fig. 2: is this really volumetric soil moisture? The values do not seem realistic; I think typical values

305 for water holding capacity for most soils are around 50 % and that would give the saturation. Please

306 check the units.

307 **Response: Thank you for pointing this out. In this revision, we traced back to Nakai et al. (2013) and**

308 **found that our units and values are the same as they presented. The reason why the values were so**

309 **high is that the volumetric soil moisture (VSM) was converted from the water content reflectometry**

310 **(WCR) probe output period using an empirical calibration function of Bourgeau-Chavez et al. (2012)**

311 **for 5cm-30cm layer. Although Bourgeau-Chavez et al. (2012) provided calibration functions for each**

312 **soil horizon (i.e., dead moss, upper duff, lower duff, and mineral soil), some of them resulted in values**

313 **greater than 100% VSM in Nakai et al. (2013) study. The model estimated high VSM (close to 80%) is**

314 **due to top 10 cm moss in the model which has a saturation VSM of 0.8. We added the discussion on**

315 **Figure 2 caption in this revision. From line 482 to 485.**

316 44) Fig. 2-d2: please use the same units as in a2, b2 and c2 figures.

317 **Response: We have corrected units for figure 2d2.**

318

Text comments:

319 **Text comments:**

320 45) - line 60: should be "… consumption to be …"

321 **Response: Corrected. See line 53.**

322 46) - line 81: "formations" should be "formation"

323 **Response: Corrected. See line 63.**

324 47) - line 118: "are in Pihlatie" should be "is Pihlatie"

325 **Response: Corrected. See line 95.**

326 48) - line 219: I think "misfit" should be "mismatch"

327 **Response: We have changed to "mismatch". See line 218.**

328 49) - line 266 and through the paper: "transient" is used wrongly. If what is meant is "variable" then

329 please do use "variable". Transient does not mean variable, but something that disappears.

330 **Response: We want to use "transient" with meaning of data having time variation. But we have**

331 **removed all of them now in manuscript to avoid misleading.**

332 50) - Fig. 3: axes text too small; markers not visible in all figures (especially c2) , please consider using

333 color markers

334 **Response: We have enlarged the axes text and used green diamond markers. Please see figure 3.**

335 51) - Fig. 3 part 1, page 34: Remove from caption the explanation for c and d

336 - Fig. 3 part 2, page 35: Remove from caption the explanation for a and b

337 **Response: We have removed them. Now figure 3 should have right description. See figure 3**

338 52) - Fig. 6: x labels not visible; some of the y labels cut

339 **Response: We have remade the figure and now the x and y labels can all be seen. See figure 6**

340 53) - Fig. 8: typo in legend: "producion"
341 - Fig. 9: typo in legend: "producion"

342 **Response: We have remade the figure 8 and figure 9. Now they have "production" in the legend. See**

343 **figure 8 and 9.**

15

344 **Marked-up manuscript:**

345

346 # Global Soil Consumption of Atmospheric Carbon Monoxide:

347 # An Analysis Using a Process-Based Biogeochemistry Model

348 Licheng Liu[1], Qianlai Zhuang[1,2], Qing Zhu[1,3], Shaoqing Liu[1,4], Hella van Asperen[5], Mari

349 Pihlatie[6,7]

[1]Department of Earth, Atmospheric, Planetary Sciences, Purdue University, West Lafayette, IN 47907, USA

[2]Department of Agronomy, Purdue University, West Lafayette, IN 47907, USA

[3] Climate Sciences Department, Climate & Ecosystem Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA

[4]Department of Earth Sciences, University of Minnesota, Minneapolis, MN, 55455,USA

[5]Institute of Environmental Physics, University of Bremen, Otto-Hahn-Allee 1, Bremen, 28359, Germany

[6]Department of Physics, University of Helsinki, P.O. Box 48, 00014 University of Helsinki, Finland

[7]Department of Forest Sciences, P.O. Box27, 00014 University of Helsinki, Finland

350 *Correspondence to:* Qianlai Zhuang(qzhuang@purdue.edu)

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Abstract: Carbon monoxide (CO) plays an important role in controlling the oxidizing capacity of the atmosphere by reacting with OH radicals that affect atmospheric methane (CH_4) dynamics. We develop a process-based biogeochemistry model to quantify CO exchange between ~~the~~ soils and the atmosphere with a 5~~five~~-minute~~s~~ internal time step at the global scale. The model is parameterized using CO flux data from the field and laboratory experiments for eleven representative ecosystem types. The model is then extrapolated to the global terrestrial ecosystems using monthly climate forcing data. Global soil gross consumption, gross production, and net flux of the atmospheric CO are estimated to be from -180 to -197, 34 to 36, and -145 to -163 ~~132-154, 29-36 and 102-119~~ Tg CO yr^{-1} (1Tg = 10^{12} g), respectively, driven with~~assuming a constant spatially distributed atmospheric CO concentration (~128 ppbv) during the 20th century. When~~ using satellite-based atmospheric CO concentration data during 2000-2013. ~~are used, our estimates of the soil gross consumption are 180-197 Tg CO yr^{-1} in the period of 2000-2013.~~ Tropical evergreen forest, savanna and deciduous forest areas are the largest sinks at 123~~93~~ Tg CO yr^{-1}. Soil CO gross consumption is sensitive to air temperature and atmospheric CO concentration while gross production is sensitive to soil organic carbon (SOC) stock and air temperature. By assuming that the ~~a constant in time,~~ spatially~~-y~~ distributed atmospheric CO concentrations (~128 ppbv) are not changing over time~~during the 20th century and 21st century,~~ global mean CO deposition velocity is estimated to be 0.16-0.19 mm s^{-1} during the 20th century. ~~under historical climate scenarios; and UuU~~nder the future climate scenarios, the ~~soil gross consumption, gross production and net flux of~~ CO deposition velocity will increase at 0.0002-0.0013 mm s^{-1} year^{-1} ~~0.15-1.23, 0.04-0.3 and 0.12-0.94 Tg CO yr^{-2}~~ during 2014-2100, reaching 0.20-0.30 mm s^{-1} ~~162-194, 36-44, and 126-150 Tg CO yr^{-1}~~ by the end of the 21st century~~, respectively~~. Areas near the equator, Eastern US, Europe and eastern Asia will be the largest sinks due to optimum soil moisture and high temperature. The annual global soil net flux of atmospheric CO is primarily controlled by air temperature, soil temperature, SOC and atmospheric CO concentrations, while its monthly variation is mainly determined by air temperature, precipitation, soil temperature and soil moisture. ~~Our process-based soil CO dynamics~~

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383 ~~model and analysis shall benefit the modeling of the global climate and atmospheric~~
384 ~~chemistry.~~

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387 **1. Introduction**

388   Carbon monoxide (CO) plays an important role in controlling the oxidizing
389 capacity of the atmosphere by reacting with OH radicals (Logan et al., 1981; Crutzen,
390 1987; Khalil & Rasmussen, 1990; Prather et al., 1995; Prather & Ehhalt, 2001). CO in
391 the atmosphere can directly and indirectly influence the fate of critical greenhouse
392 gases such as methane (CH_4) and ozone (O_3) (~~Logan et al., 1981; Crutzen & Gidel,~~
393 ~~1983; Guthrie, 1989; Khalil & Rasmussen, 1990; Lu & Khalil, 1993; Daniel & Solomon,~~
394 ~~1998; Prather & Ehhalt, 2001;~~ Tan and Zhuang, 2012). Although CO itself absorbs only
395 a limited amount of infrared radiation from the Earth, the cumulative indirect radiative
396 forcing of CO may be even larger than that of the third powerful greenhouse gas, nitrous
397 oxide (N_2O, Myhre et al., 2013). Current estimates of global CO emissions from both
398 anthropogenic and natural sources range from 1550 to 2900 Tg CO yr^{-1}, which are
399 mainly from anthropogenic and natural direct emissions and from the oxidation of
400 methane and other Volatile Organic Compounds (VOC) (Prather et al., 1995; Khalil et
401 al., 1999; Bergamaschi et al., 2000; Prather & Ehhalt, 2001, Stein et al., 2014).
402 Chemical consumption of CO by atmospheric OH and the biological consumption of CO
403 by soil microbes are two major sinks of the atmospheric CO (Conrad, 1988; Lu & Khalil,
404 1993; ~~Prather et al., 1995; Prather & Ehhalt, 2001;~~ Yonemura et al., 2000; Whalen &
405 Reeburgh, 2001).
406   Soils are globally considered as a major sink for CO due to microbial activities
407 (~~Conrad and Seiler, 1982; Potter et al., 1996;~~ Whalen and Reeburgh, 2001; King and
408 Weber, 2007). A diverse group of soil microbes including carboxydotrophs,
409 methanotrophs and nitrifiers are capable of oxidizing CO (~~Ferenci et al., 1975; Jones~~
410 ~~and Morita, 1983; Bender and Conrad, 1994;~~ King and Weber, 2007). Annually, 10-25%
411 of <u>total earth surface</u> CO emissions were consumed by soils (Sanhueza et al., 1998;
412 ~~Khalil et al., 1999;~~ King, 1999a; ~~Bergamaschi et al., 2000; Prather & Ehhalt, 2001;~~ Chan
413 & Steudler, 2006). Potter et al. (1996) reported the global soil consumption <u>to</u> be <u>from</u> -

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16 ~~to~~ ~~-~~50 Tg CO yr[-1] (positive values represent ~~direction is~~ the emissions from soils to the atmosphere), by using a single~~-~~box model ~~approach~~ over the upper 5 cm of soils. All existing ~~Other~~ estimates have ~~after this still remained~~ large uncertainties ~~showed large ranges using simple assumptions,~~ ranging from -115 to -640 Tg CO yr[-1] (Sanhueza et al., 1998; King, 1999; Bergamaschi et al., 2000). Similarly, the estimates of ~~such as 115–230 Tg CO yr⁻¹ based on a constant dry deposition velocity (the uptake rate divided by the CO concentration) of 0.03 cm s⁻¹ (Sanhueza et al., 1998); 300 Tg CO yr⁻¹ using the same constant deposition velocity and zero deposition velocity value in deserts and areas with monthly mean temperatures below 0 °C with different approaches (Bergamaschi et al., 2000); 190-580 Tg CO yr⁻¹ using empirical approaches with a higher probability for lower values (King, 1999). Besides, reported~~ CO dry deposition velocities also have large uncertainties, ranging from ~~(~~0 to 4.0~~0.004~~mm s⁻¹~~)~~ ~~for vegetated surfaces based on measurements are relatively low compared with other substances~~ (here positive values are amount of deposition to soils ~~uptakes,~~ King, 1999a; Castellanos et al., 2011). Soils also produce CO mainly via abiotic processes such as thermal- and photo-degradation of organic matter or plant materials (Conrad and Seiler, 1985b; Tarr et al., 1995; Schade et al., 1999; Derendorp et al., 2011; Lee et al., 2012; van Asperen et al., 2015; Fraser et al., 2015, Pihlatie et al., 2016), except for a few cases of anaerobic formation. Photo-degradation is identified as radiation-dependent degradation due to absorbing radiation (King et al., 2012). Thermal-degradation is identified as the temperature-dependent degradation of carbon in the absence of radiation and possibly oxygen (Derendorp et al., 2011; Lee et al., 2012; van Asperen et al., 2015; Pihlatie et al., 2016). These major soil CO production processes, together with soil CO consumption processes, have not been adequately modeled in global soil CO budget estimates.

~~To date, there are still large uncertainties in estimating soil CO consumption, ranging from 15 to 640 Tg CO yr⁻¹. Although soil CO consumption and its environmental controls have been heavily studied, the impacts of long-term changes in climate and human activities on the atmosphere-biosphere CO exchange are still not clear (King & Weber, 2007; Vreman et al., 2011; He and He, 2014; Pihlatie et al., 2016). Moreover, production of CO has been widely found in soils, plant roots, living and degrading plant~~

~~materials and degrading organic matter (Pihlatie et al., 2016). CO production is dominantly due to abiotic processes such as thermal- and photo-degradation of organic matter or plant material (Conrad and Seiler, 1985b; Tarr et al., 1995; Schade et al., 1999; Derendorp et al., 2011; Lee et al., 2012; van Asperen et al., 2015; Fraser et al., 2015, Pihlatie et al., 2016), except for a few cases of anaerobic formations. Photo-degradation is identified as radiation-dependent degradation includes direct photo-degradation due to absorbing radiation by light-absorbing molecules and indirect photo-degradation due to radiation energy transferring to non-light-absorbing molecules (King et al., 2012). Thermal-degradation is identified as the temperature-dependent degradation of carbon in the absence of radiation and possibly oxygen (Derendorp et al., 2011; Lee et al., 2012; van Asperen et al., 2015; Pihlatie et al., 2016). Previous field and laboratory studies on the role of direct or indirect abiotic degradation showed very contrasting results, primarily due to the challenge of separation between CO formation through thermal-degradation and photo-degradation, because they can both occur simultaneously and the indirect photo-degradation may occur even without solar radiation if thermal energy is suitable (Lee et al., 2012).~~

Little ~~attention~~focus has been ~~paid~~placed so far ~~to estimate~~ on the role of net CO budget (including soil CO consumption and production) in global ~~chemistry~~ climate ~~modeling.~~ To date, m~~M~~ost top-down atmospheric models appl~~ied~~y a dry deposition scheme based on the resistance model of Wesely (1989). Such schemes provided ~~give~~ a wide range of dry deposition velocities (Stevenson et al., 2006). Only a few models (MOZART-4, Emmons et al., 2010; CAM-chem, Lamarque et al., 2012) have extended their dry deposition schemes with a parameterization for CO and H_2 uptake through ~~by~~ oxidation by ~~from~~ soil ~~bacteria and~~ microbes following the work of Sanderson et al. (2003), which itself was based on extensive measurements from Yonemura et al. (2000). Potter et al. (1996) developed a bottom-up model to simulate CO consumption and production at the global scale. This model is a single box model, only considers top 5cm depth of soil and does not have explicit microbial factors, which might have underestimated CO consumption (Potter et al., 1996; King, 1999a). Current bottom-up CO modeling approaches are mostly based on a limited number of CO *in situ* observations or laboratory studies to quantify regional and global soil consumption

Commented [LL2]: Reduce photo degradation

(Potter et al., 1996; Sanhueza et al., 1998; Khalil et al., 1999; King, 1999a; Bergamaschi et al., 2000; Prather & Ehhalt, 2001). To our knowledge, no detailed process-based model of soil-atmospheric exchange of CO has been published in the recent 15 years. One reason is that there is an incomplete understanding of biological processes of ~~CO emission and~~ uptake (King & Weber, 2007; Vreman et al., 2011; He and He, 2014; Pihlatie et al., 2016). Another reason is that there is ~~a~~ lack of long-term CO flux measurements for different ecosystem types to calibrate and evaluate the models. CO flux measurements are mostly from short-term field observations or laboratory experiments (e.g. Conrad and Seiler, 1985a; Funk et al., 1994; Tarr et al., 1995; Zepp et al., 1997; Kuhlbusch et al., 1998; Moxley and Smith, 1998; Schade et al., 1999; King and Crosby, 2002; Varella et al., 2004; Lee et al., 2012; Bruhn et al., 2013; van Asperen et al., 2015). The first study to report long-term and continuous field measurements of CO flux over grasslands using a micrometeorological eddy covariance (EC) method is ~~are in~~ Pihlatie et al. (2016).

To ~~Aiming to~~ improve ~~the understanding of processes associated with land-atmosphere CO exchange and t~~the quantification of the ~~o quantify~~ global soil CO budget for the period 2000-2013 and CO deposition velocity ~~for the 20th and 21st centuries,~~ this study ~~we~~ developed a CO dynamics module (CODM) embedded in a process-based biogeochemistry model, the Terrestrial Ecosystem Model (TEM) (Zhuang et al., 2003, 2004, 2007). CODM was then calibrated and evaluated using laboratory experiments and field measurements for different ecosystem types. ~~We then used T~~the atmospheric CO concentration data from MOPITT (Gille, 2013) were used to drive ~~our~~ model simulations from 2000 to 2013. A set of c~~We conducted c~~entury-long simulations of 1901-2100 were also conducted ~~,~~ using the atmospheric CO concentrations estimated with an empirical function (Badr & Probert, 1994; Potter et al., 1996). Finally, ~~We also evaluated~~ the effects of multiple forcings on the global CO consumption and production ~~estimates~~, including the changes of climate and atmospheric CO concentrations at the global scale were evaluated with the model.

2. Method

2.1 Overview

We first developed a daily soil CO dynamics module (CODM) that considers: (1) soil-atmosphere CO exchange and diffusion process between soil layers, (2) consumption by soil microbial oxidation, (3) production by soil chemical oxidation, and (4) the effects of temperature, soil moisture, soil CO substrate and surface atmospheric CO concentration on these processes. Second, we used the observed soil temperature and moisture to evaluate ~~(Terrestrial Ecosystem Model)~~ TEM hydrology module and soil thermal module in order to estimate soil physical variables ~~correctly~~. Then we used the data from ~~results of~~ laboratory experiments and CO flux measurements to parameterize the model ~~and calibrate the model~~ using the Shuffled Complex Evolution (SCE-UA) method (Duan et al., 1993). Finally, the model was extrapolated to the globe ~~al scale~~ at a 0.5° by 0.5° resolution. We conducted three sets of model experiments to investigate the impact of climate and atmospheric CO concentrations on soil CO dynamics: 1) simulations for 2000-2013 with MOPITT satellite atmospheric CO concentration data~~1901-2013 with constant atmospheric CO concentrations estimated from an empirical function~~; 2)simulations for ~~2000-2013 with MOPITT satellite atmospheric CO concentration data~~1901-2100 with constant atmospheric CO concentrations estimated from an empirical function and the ~~one~~ historical climate data ~~scenairos~~ (1901-2013) and three future climate scenarios (2014-2100); and 3) ~~Eight sensitivity simulations by changing with~~ a) constant CO surface concentrations ± 30%, b) SOC ±30%, c) precipitation ±20% and d) air temperature ± 3°C for each pixel, respectively, while holding other forcing data as they were, during 1999-2000~~2014-2100 with the same constant atmospheric CO concentrations as 1) and three future climate scenarios~~.

2.2 Carbon Monoxide Dynamics Module (CODM)

Embedded in TEM (Figure 1), CODM is mainly driven by: (1) soil organic carbon availability based on a carbon and nitrogen dynamics module (CNDM) (Zhuang et al., 2003); (2) soil temperature profile from a soil thermal module (STM) (Zhuang et al., 2001, 2003); and (3) soil moisture profile from a hydrological module (HM) (Bonan, 1996; Zhuang et al, 2004). Net exchange of CO between the atmosphere and soil is

determined by the mass balance approach (net flux = total production – total oxidation – total soil CO concentration change). According to previous studies, we separated active soils (top 30cm) for CO consumption and production into 1 cm thick layers (King, 1999a, 1999b; Whalen & Reeburgh, 2001; Chan & Steudler, 2006). Between the soil layers, the changes of CO concentrations are calculated as by:

$$\frac{\partial(C(t,i))}{\partial t} = \frac{\partial}{\partial z}\left(D(t,i)\frac{\partial(C(t,i))}{\partial z}\right) + P(t,i) - O(t,i) \quad (1)$$

Where $C(t,i)$ is the CO concentration in layer i i and at time t t, units are mg m^{-3}. z is the depth of the soil, units are m thickness of layer i. $D(t,i)$ is the diffusion coefficient for layer i, units are m^2 s^{-1}. $P(t,i)$ is the CO production rate and $O(t,i)$ is the CO consumption rate due to oxidation. The units of $P(t,i)$ and $O(t,i)$ are mg m^{-3} s^{-1}. $D(t,i)$ is calculated using the method from Potter et al. (1996), equations (2) to (4), which are is the functions of soil temperature, soil texture and soil moisture. The upper boundary condition is specified as the atmospheric CO concentration, which is estimated by an empirical function of latitude (Potter et al., 1996) or directly measured by the MOPITT satellite during 2000-2013. The lower boundary condition is assumed to have no diffusion exchange with the layer underneath. This partial differential equation (PDE) is solved using the Crank-Nicolson method for less time-step-sensitive solution.

CO consumption is modeled as an aerobic process occurring in unsaturated soil pores, which is estimated as:

$$O(t,i) = V_{max} \cdot f_1\big(C(t,i)\big) \cdot f_2\big(T(t,i)\big) \cdot f_3\big(M(t,i)\big) \quad (2)$$

Where V_{max} is the specific maximum oxidation rate, ranging from 0.3 to 11.1 µg CO g^{-1} h^{-1} (Whalen & Reeburgh, 2001). f_i represents the effects of soil are functions calculating CO concentration $C(t,i)$, temperature $T(t,i)$ and moisture $M(t,i)$ influences on CO soil consumption. Considering CO consumption as the result of microbial activities, we calculated $f_1\big(C(t,i)\big)$, $f_2\big(T(t,i)\big)$ and $f_3\big(M(t,i)\big)$ in a similar way as Zhuang et al. (2004):

$$f_1\big(C(t,i)\big) = \frac{C(t,i)}{C(t,i) + k_{CO}} \quad (2.1)$$

23

563
$$f_2\big(T(t,i)\big) = Q_{10}^{\frac{T(t,i)-T_{ref}}{10}} \qquad (2.2)$$

564
$$f_3\big(M(t,i)\big) = \frac{(M(t,i)-M_{min})(M(t,i)-M_{max})}{(M(t,i)-M_{min})(M(t,i)-M_{max})-(M(t,i)-M_{opt})^2} \qquad (2.3)$$

565 Where $f_1\big(C(t,i)\big)$ is a multiplier that enhances oxidation rate with increasing soil CO

566 concentrations using a Michaelis-Menten function with a half-saturation constant k_{CO},

567 ranging from 5 to 51 µl CO l^{-1} (Whalen & Reeburgh, 2001); $f_2\big(T(t,i)\big)$ is a multiplier that

568 enhances CO oxidation rates with increasing soil temperature using a Q10 function with

569 Q_{10} coefficients (Whalen & Reeburgh, 2001). T_{ref} is the reference temperature, units

570 are °C (Zhuang et al., 2004, 2013). $f_3\big(M(t,i)\big)$ is a multiplier to estimate the biological

571 limiting effect that diminishes CO oxidation rates if the soil moisture is not at an optimum

572 level (M_{opt}). M_{min}, M_{max} and M_{opt} are the minimum, maximum and optimum volumetric

573 soil moistures of oxidation reaction, respectively. Equation (2.2) will overestimate CO

574 consumption at higher temperature because in reality CO consumption ~~has an optimum~~

575 ~~temperature and it~~ will decrease at higher temperatures than optimum temperature,

576 while f$_2$ will ~~equation (2.2) CO consumption~~ keeps increasing with rising temperature.

577 However, the CO consumption is constrained by CO production, and equation (1) is

578 used to represent this constraint.

579   We modeled the CO production rate ($P(t,i)$) as a process of chemical oxidation

580 constrained by soil organic carbon (SOC) decay (Conrad and Seiler,1985; Potter et al.

581 1996; Jobbagy & Jackson, 2000; van Asperen et al., 2015):

582
$$P(t,i) = P_r(t,i) \cdot E_{SOC} \cdot C_{SOC}(t) \cdot F_{SOC} \qquad (3)$$

583 Where $P_r(t,i)$ is a reference soil CO production rate which has been normalized to rate

584 at reference temperature (production rate at temperature (t,i) divided by production

585 rate at reference temperature), which is affected by soil moisture and soil temperature

586 (Conrad and Seiler,1985; van Asperen et al., 2015). E_{SOC} is an estimated nominal CO

587 production factor of 3.5 ± 0.9 X 10^{-9} mg CO m^{-2} s^{-1} per g SOC m^{-2} (to 30 cm surface soil

588 depth) (Potter et al., 1996). $C_{SOC}(t)$ is a SOC content in mg m^{-2}, which is provided by

589 CNDM module in TEM. F_{SOC} is a constant fraction of top 320cm SOC compared to total

590 amount of SOC, which is 0.33 for shrubland areas, 0.42 for grassland areas and 0.50
591 for forest areas, respectively (Jobbagy & Jackson, 2000). $P_r(t,i)$ is calculated as:

592 $$P_r(t,i) = \exp\left(f_4\big(M(t,i)\big) \cdot Ea_{ref}/R \cdot \left(\frac{1}{273.15 + PT_{ref}} - \frac{1}{T(t,i) + 273.15}\right)\right) \quad (3.1)$$

593 $$f_4\big(M(t,i)\big) = \frac{PM_{ref}}{M(t,i) + PM_{ref}} \quad (3.2)$$

594 Where equation (3.1) is derived from Arrhenius equation for chemical reactions and
595 normalized using the reference temperature PPT_{ref}. Ea_{ref}/R is the reference activation
596 energy divided by gas constant R, units are K. $f_4\big(M(t,i)\big)$ is the multiplier that reduces
597 activation energy using an regression approach based on laboratory experiment of
598 moisture influences on CO production (Conrad and Seiler,1985). PM_{ref} is the reference
599 volumetric soil moisture, ranging from 0.01 to 0.5 volume/volume (v/v). We assumed
600 thermal-degradation as the main CO producing process due to since lack of photo-
601 degradation data and hard to distinguish photo-degradation from observations. In order
602 to reduce the bias from thermal-degradation to total abiotic degradation, the equation
603 (3.1) was is parameterized by comparing with total production rate. For instance, $P_r(t,i)$
604 calculation can perfectly fit the experiment results in Van Asperen et al., 2015 with
605 proper PT_{ref}(18°C), Ea_{ref}/R(14000 K) and PM_{ref}(0.5 v/v).
606   CO deposition velocity was is modeled in the same way as equation (19.1) in
607 Seinfeld, et al., (1998):

608 $$v_d = -F_{net}/C_{CO,air} \quad (4)$$

609 Where the v_d is the CO deposition velocity, units are mm s^{-1}; F_{net} is the model
610 estimated CO net flux rate, units are mg CO m^{-2} day^{-1}; $C_{CO,air}$ is the CO surface
611 concentration, units are ppbv. $C_{CO,air}$ can be MOPITT CO surface concentration data or
612 derived CO surface concentrations using the same method as Potter, et al. (, 11996).
613 Positive values of v_d are soil uptakes (depositions from air to soils) and negative values
614 are soil emissions.

615

616 **2.3 Model Parameterization and Extrapolation**

The model parameterization was conducted in two steps: 1) Thermal and
hydrology modules embedded in TEM were revised, calibrated and evaluated by
running model with corresponding local meteorological or climatic data at four ~~4~~
representative sites, including boreal forest, temperate forest, tropical forest and
savanna (Table 1, site No.1 to 4, Figure 2) to minimize model data mismatch~~fit~~ in terms
of soil temperature and moisture. 2) CODM module was parameterized by running TEM
for observational periods with the corresponding local meteorological or climatic data at
each reference site (Table 1, Figure 3), and using the Shuffled Complex Evolution
Approach in R language (SCE-UA-R) (Duan et al., 1993) to minimize the difference
between simulated and observed net CO flux. Eleven parameters including k_{CO}, V_{max},
T_{ref}, Q_{10}, M_{min}, M_{max}, M_{opt}, E_{SOC}, Ea_{ref}/R, PM_{ref} and PT_{ref} were ~~are~~ optimized (Table 2).
To be noticed, F_{SOC} was not involved in the calibration process. Parameter priors were
decided based on previous studies (Conrad & Seiler, 1985; King, 1999b; Whalen &
Reeburgh, 2001; Zhuang et al., 2004). SCE-UA-R was used for site No. 6, 8, 10, 11
(Table 1). Each site has~~d~~ been run 50 times using SCE-UA-R with 10000 maximum
loops for parameter ensemble, and all of them reached stable state before the end of
the loops. For wetlands, the only available data is from site No.12. We used a trial-and-
error method ~~instead~~ to ~~to~~ make our simulated results in the range of observed flux
rates, with a 10% tolerance. For tropical sites, since tropical savanna vegetation type is
a combination type of tropical forest and grassland in our model, we first used Site No.
13 to set priors to fit the experiment results with a 10% tolerance and then evaluated by
running our model comparing with site No.7 results. Site No. 9 and 5 were used to
evaluate our model results for temperate forest and grassland. Besides the observed
climatic and soil property data, we used ERA-Interim reanalysis data from The
European Centre for Medium-Range Weather Forecasts (ECMWF) (Dee et al., 2011),
AmeriFlux observed meteorology data (http://ameriflux.lbl.gov/) and reanalysis climatic
data from Climatic Research Unit (CRU, Harris et al., 2013) to fill the missing
environmental data. To sum up, parameters for various ecosystem types in T~~t~~able 2
were the final results of our parameterization. Model parameterization was conducted
for ecosystem types including boreal forest, temperate coniferous forest, temperate
deciduous forest, and grassland using SCE-UA-R. Tropical forest and wet tundra used

648 a trial-and-error method to adjust parameters letting simulation results best fit the lab
649 data. Due to limited data availability, we assumed temperate evergreen broadleaf forest
650 having the same parameters as temperate deciduous forest.
651
652 **2.4 Data Organization**
653 To get spatially and temporally explicit estimates of CO consumption, production
654 and net flux at the global scale, we used the data of land cover, soils, climate and leaf
655 area index (LAI) from various sources at a spatial resolution of 0.5° latitude X 0.5°
656 longitude to drive TEM. The land cover data include potential vegetation distribution
657 (Melillo et al., 1993) and soil texture (Zhuang et al., 2003), which were used to assign
658 vegetation- and texture-specific parameters to each grid cell.
659 For the simulation of the period 1901-2013, monthly air temperature, precipitation,
660 clouds fraction and vapor pressure data sets from CRU were used to estimate the soil
661 temperature, soil moisture and SOC with TEM (Figure 4). Monthly LAI data from TEM
662 were required to simulate soil moisture (Zhuang et al., 2004). During this period time,
663 we used an empirical function of latitude, which was derived from the observed
664 latitudinal distribution of tropospheric carbon monoxide (Badr and Probert, 1994) to
665 calculate CO surface concentrations (equation (7), Potter et al., 1996):

666 $$C_{CO,air} = 82.267856 + 0.8441503L + 1.55934 \times 10^{-2}L^2 + 2.37 \times 10^{-5}L^3 -$$

667 $$2.3 \times 10^{-6}L^4 \qquad (5)$$

668 Where $C_{CO,air}$ is the derived surface CO concentration (ppbv), L represents
669 latitude which is negative degrees for southern hemisphere and positive degrees for
670 northern hemisphere. We also used the ~~transient~~ atmospheric CO data from MOPITT
671 satellite during 2000-2013 (Figure 5). We averaged day-time and night-time monthly
672 mean retrieved CO surface level 3 data (variables mapped on 0.5° latitude X 0.5°
673 longitude grid scales with monthly time step, Gille, 2013) to represent the CO surface
674 concentration level in each month. The missing pixels were fixed by the average of
675 pixels which had values and were inside 1.5 times of the distance between this missing
676 pixel and the nearest pixel with values. These global mean values shown in Figure 5 do
677 not include ocean surfaces, thus there are differences between our surface CO

27

678 concentration results and Yoon and Pozzer's report in 2014, which is as low as 99.8ppb.

679 From 2014 to 2100, we used Intergovernmental Panel on Climate Change (IPCC) future

680 climate scenarios from Representative Concentration Pathways (RCPs) climate forcing

681 data sets RCP2.6, RCP4.5 and RCP8.5 (Figure 6). RCP2.6, 4.5 and 8.5 datasets are

682 future climate projections with anthropogenic greenhouse gas emission ~~and will drive~~

683 radiative ~~oactive~~ forcing ~~to~~ of 2.6 W m^{-2}, 4.5 W m^{-2} and 8.5 W m^{-2}~~-~~, respectively, by 2100.

684 Since RCPs did not have water vapor pressure data, we used the specific humidity and

685 sea level air pressure from the RCPs and elevation of surface to estimate the monthly

686 surface vapor pressure ~~data~~ (Seinfeld & Pandis,- 1998~~2006~~).

687

2.5 Model Experiment Design

689 We conducted two sets of core simulations and eight sensitivity test simulations

690 for a ~~in~~ historical period. The two core sets of simulations were driven with MOPITT CO

691 surface concentrations data ~~estimated from an empirical function of latitude (experiment~~

692 ~~E1)~~ for the period 2000~~1901~~-2013 (experiment E1) and with ~~constant in time,~~ spatially

693 distributed ~~transient~~ CO surface concentrations assuming as constant over time

694 estimated from an empirical function of latitude~~from MOPITT satellite data (experiment~~

695 ~~E2)~~ for the period 1901-2100 ~~2000-2013~~(experiment E2), respectively. Specifically, in

696 experiment E2 we used the CRU climate forcing for the historical period 1901-2013 and

697 the ~~~~climate data of RCP2.6, RCP4.5 and RCP8.5 for different future scenarios to

698 examine the responses of CO flux to changing climates. Eight sensitivity simulations

699 were driven with varying different forcing variables while keeping others as they were: 1)

700 with constant CO surface concentrations ± 30%, 2) SOC ±30%, 3) precipitation ±20%

701 and 4) air temperature ± 3°C for each pixel, respectively, during 1999-2000 (E3). ~~For~~

702 ~~the 21st century, we conducted simulations driven with climate data of RCP2.6, RCP4.5~~

703 ~~and RCP8.5 to examine the responses of CO flux to changing climates (E4).~~

704

3. Results

3.1 Site Evaluation

707   Both the magnitude and variation of the simulated soil temperature and moisture

708   from cold areas to warm areas compared well to the observations (Figure. 2). The

709   magnitude of the simulated CO flux is ~~highly~~ comparable and correlated with the

710   observations (r is about 0.5, p-value < 0.001, Figures 3, a2, b2 ,c2 ,d2). Estimated CO

711   fluxes ~~rates~~ for different ecosystem types ~~are~~ range ~~ing~~ from -28.4 to 1.7 mg CO m^{-2}

712   day^{-1}, and t~~with~~ he rRoot mean square error (RMSE) between simulation and

713   observation at ~~of~~ of ~~the simulated CO flux for~~ all sites ~~is~~ is below 1.5 mg CO m^{-2} day^{-1}.

714   RMSE for site No. 7 is bigger than 2.0 mg CO m^{-2} day^{-1} when compared with

715   transparent chamber observations. For boreal forest site, we only had~~ve~~ 8 acceptable

716   points in 1994 and 1996 (Figure 3c2).

717

718   **3.2 Global Soil CO Dynamics During 2000~~1901~~-2013 ~~(E1)~~**

719   Using the MOPITT ~~For the simulation with constant~~ CO surface concentrations

720   data (E1) during ~~1901~~2000-2013, the estimated mean soil CO consumption, production

721   and net flux (positive values ~~direction~~ indicate ~~is~~ CO emissions from soils to the

722   atmosphere) are from -180 to -197, 34 to 36 and -145 to -163~~141, 32 and -108~~ Tg CO

723   yr^{-1}, respectively (Figure 7a). ~~In the long-term simulations, annual soil CO fluxes vary~~

724   ~~slightly. The annual soil CO consumption, production and net flux vary within 10%~~

725   ~~during the period (Figure 8a).~~ Consumption is about 4 times larger than production. The

726   annual consumption and net flux trends follow the atmospheric CO concentration trends

727   (Figure 5b, Figure 7a), with a small interannual variability (<10%). The latitudinal

728   distributions of consumption, production and net flux ~~rates~~es share the same spatial

729   pattern. Around 20°S-20°N and 20-60N° are the largest and second largest areas for

730   production and consumption, while the 45°S-45°N area accounts for nearly 90% of the

731   total consumption and production (Figure 7b, Table 3). The Southern and Northern

732   Hemispheres have 41% and 59% of the total consumption, and 47% and 53% of the

733   total production, respectively (Table 3).   The highest rates of consumption and

734   production are located in areas close to the equator, and consumption from areas such

735   as eastern US, Europe and eastern Asia also is high ~~has large rates~~ (>-1000 mg m^{-2} yr^{-1})

736   (Figure 8~~7~~a, b). Global~~ly~~ soils serve as an atmospheric CO sink (Figure 8~~7~~c). Some

737   areas, such as western US and southern Australia, are CO sources, all of which are

738 grasslands or experiencing dry climate. ~~The latitudinal distributions of consumption,~~
739 ~~production and net flux rates share the same spatial pattern. Around 20°S-20°N and 20-~~
740 ~~60N° are the largest and second largest areas for production and consumption, while~~
741 ~~the 45°S-45°N area accounts for nearly 90% of total consumption and production~~
742 ~~(Figure 9a, Table 3). The Southern and Northern Hemispheres consume 42% and 58%~~
743 ~~of the total consumption, and produce 41% and 59% of total production, respectively~~
744 ~~(Table 3).~~ Tropical evergreen forests are the largest sinks, consuming 86~~66~~ Tg CO yr^{-1},
745 and tropical savanna and deciduous forest are second and third largest sinks,
746 consuming a total of 37~~27~~ Tg CO yr^{-1} (Table 4). These three ecosystems account for 66%
747 of the total consumption. Tropical evergreen forests are also the largest source of soil
748 CO production, producing 16~~5~~ Tg CO yr^{-1}, while tropical savanna ha~~sve~~ a considerable
749 production of 6 Tg CO yr^{-1} (Table 4). Moreover, tropical areas, including forested
750 wetlands, forested floodplain and evergreen forests, are most efficient for CO
751 consumption, ranging from -13~~0~~ to -18~~2~~ mg CO m^{-2} day^{-1}. They are also the most
752 efficient for CO production at over 2 mg CO m^{-2} day^{-1} (Table 4, calculated by fluxes
753 ~~values~~ divided by area~~3~~).
754 ~~For the simulation with transient atmospheric CO surface concentrations (E2)~~
755 ~~during 2000-2013, the mean annual global soil consumption increases to -187 Tg CO~~
756 ~~yr^{-1}, and areas near the equator become large sinks for atmospheric CO together with~~
757 ~~eastern US, Europe, and eastern Asia (Figure 7) due to the heavy atmospheric CO~~
758 ~~burden over these areas (Figure 5a). The annual consumption and net flux trends follow~~
759 ~~the atmospheric CO concentration trends (Figure 5b, Figure 8b), with a small~~
760 ~~interannual variability (<10%). The latitudinal distributions of soil CO fluxes for E1 and~~
761 ~~E2 are similar but E2's CO fluxes magnitudes are larger than E1's and around 30°N of~~
762 ~~E2's distribution shows another peak of CO consumption, due to the high atmospheric~~
763 ~~CO concentration over eastern Asia (Figure 5a, Figure 9b). The consumption between~~
764 ~~45°S-45°N increases by 35%, to -137 Tg CO yr^{-1}, which is 73% of the global total~~
765 ~~annual consumption. Consumption rates of high latitude areas (45°N North) do not~~
766 ~~change significantly (Figure 7, 9, Table 3), and the annual consumption only increases~~
767 ~~by 10%, thus the portion of soil CO sinks in northern high latitudes decreases from 12%~~
768 ~~to 10% of the global total.~~

3.3 Global Soil CO Dynamics During 1901~~2014~~-2100 ~~(E2)~~

Using the constant ~~atmospheric~~ CO surface concentration, the estimated global ~~annual~~ mean ~~soil~~ CO deposition velocities are 0.16-0.19 mm s^{-1} for the period 1901-2013. ~~consumptions~~ ~~F~~for the period 2014-2100, deposition velocities ~~–~~are 0.18-0.21, 0.18-0.24 and 0.17-0.31 ~~-162, -174 and -194 Tg CO yr^{-1} while estimated annual mean soil productions are 36, 39 and 44 Tg CO yr^{-1}~~ for RCP2.6, 4.5 and 8.5 scenarios, respectively (Figure 9). During 2014-2100, there are significant trends of increasing consumption, production and net flux for nearly all scenarios (Figure 9). The rates of increasing are 0.0002, 0.0005 and 0.0013 mm s^{-1} y~~ea~~r^{-1}, and will reach 0.20, 0.23 and 0.30 mm s^{-1} by the end of the 21st century for the RCP2.6, 4.5 and 8.5 scenarios, respectively (Figure 9). These increasing trends are similar to air temperature increasing trends (Figure 6a). ~~The net fluxes are -118.06, -117.31 and -115.13 Tg CO yr^{-1} at the beginning 10 years of the 21st century, and will reach -127.17, -144.99 and -187.25 Tg CO yr^{-1} at the end of the 21st century for RCP2.6, RCP4.5 and RCP8.5 scenarios, respectively (Figure 11).~~ Global distribution patterns of CO deposition velocity ~~consumption, production and net flux~~ are similar to net flux~~es~~ distribution for the period 2000-2013 ~~the 20th century~~ but there are significant differences among 1901-2013, RCP2.6, RCP4.5 and RCP8.5 scenarios (Figure 10) ~~on areas near the equator.~~ Deposition ~~flux~~ velocities ~~rates~~ are increasing from RCP2.6 to 8.5 and larger than in the historical periods i~~o~~n areas near the ~~equator~~ equator ~~and all three future scenarios' deposition velocity on these areas are larger than historical scenarios~~ (Figure 10). Areas near the equator and eastern Asia become big sinks of atmospheric CO, while northeastern US becomes a small source in the 21st century (Figure 10). Different vegetation types have a large range of deposition velocity, from 0.008 to 1.154 mm s^{-1} (Table 5). The tropical forested wetland, tropical forested floodplain and tropical evergreen forest have top three largest deposition velocity of 1.154, 1.117 and 0.879 mm s^{-1}, respectively, while desert, short grasslands, and wet tundra have the smallest deposition velocity 0.008, 0.010 and 0.015 mm s^{-1}, respectively. ~~The consumption has relatively fast growth rates during the 21st century (Figure 11). Furthermore, there are significant trends of increasing consumption, production and net flux for nearly all~~

3.4 Sensitivity test ~~(E3)~~

Eight sensitivity tests have been conducted for the 1999-2000 period, including changing atmospheric CO by ±30%, SOC by ±30%, precipitation by ±30% and air temperature by ±3°C for each pixel (Table 5). Soil CO consumption is most sensitive (changing 29%) to air temperature while production is most sensitive (changing up to 36%) to both air temperature and SOC (30%). The net CO fluxes have the similar sensitivities as consumption. Annual CO consumption, production and net flux follow the change of air temperature (Table 5). In addition~~Besides~~, a 30% change in precipitation will not lead to large changes in CO flux (< 3%).

4. Discussion

4.1 Comparison with Other Studies

Previous studies estimated a large range of global CO consumption from -16 to -6~~40~~36 Tg CO yr^{-1}. Our estimates are from -180 to -197 ~~-132 to -154~~ Tg CO yr^{-1} for ~~the 20th century and -180 to -197 Tg CO yr^{-1}~~ for 2000-2013 using MOPITT satellite CO surface concentration data. Previous studies also provided a large range for CO production from 0 to 7.6 mg m^{-2} day^{-1} (reviewed in Pihlatie et al., 2016). Our results showed averaged CO production ranging from 0.01 to 2.29 mg m^{-2} day^{-1}. Previously reported CO deposition velocities for different vegetation types range from 0.0 to 4.0 mm s^{-1} while o~~. O~~ur results showed an averaged CO deposition velocity ranging ~~e from~~ 0.006 to 1.154 mm s^{-1} for different vegetation types. The large uncertainty of these estimates is mainly due to a different consideration of the microbial activities, the depth

827 of the soil, and the parameters in the model. In contrast to the estimates of -16 to -57 Tg
828 CO yr^{-1} which were based on top 5 cm soils (Potter et al., 1996), our estimates
829 considered 30cm soils, just as used in Whalen & Reeburgh (2001). In addition, we
830 used a thinner layer division (1cm each layer) for diffusion process, and used the Crank-
831 Nicolson method to solve ing partial differential equations to avoid time step influences.
832 We also included the microbial CO oxidation process to remove the CO from soils and
833 the effects of soil moisture, soil temperature, vegetation type and soil CO substrate on
834 microbial activities. Besides, Oour soil thermal, soil hydrology and carbon and nitrogen
835 dynamics simulated in TEM provided carbon substrate spatially and temporally for
836 estimating soil CO dynamics (Bonan, 1996; Zhuang et al., 2001, 2003, 2004, 2007).
837 Overall, although a few previous studies have examined the long-term impacts of
838 climate, land use and nitrogen depositions on CO dynamics (Chan & Steudler, 2006,
839 Pihlatie et al., 2016), the global prediction of soil CO dynamics still has ve a large
840 uncertainty.
841
842 **4.2 Major Controls to Soil CO Dynamics**

843 Eight sensitivity tests have been conducted for the 1999-2000 period, including
844 changing atmospheric CO by ±30%, SOC by ±30%, precipitation by ±30% and air
845 temperature by ±3°C for each pixel (Table 5). Soil CO consumption is most sensitive
846 (changing 29%) to air temperature while production is most sensitive (changing up to
847 36%) to both air temperature and SOC (30%). The net CO fluxes have the similar
848 sensitivities to consumption, Sensitivity tests indicate that because consumption is
849 normally much larger than CO production so that the former it will determine the
850 dynamics of the net flux (Table 5). Annual CO consumption, production and net flux
851 follow the change of air temperature (Table 5), which Model being sensitive to air
852 temperature explains the small increasing trends after the 1960s, the significant
853 increasing trend in the 21st century and the large sinks over tropical areas (Table 5,
854 Figure 9). Besides, a 30% change in precipitation will not lead to large changes in CO
855 flux (< 3%). SOC did not directly influence CO consumption. For instance, iIncreasing
856 SOC led to an increase in soil CO substrate. so implying that more CO in soils can be
857 consumed. To be noticed, the CO oxidation increasing due to CO substrate change is

858 larger bigger than production rate increasing due to SOC increasing, which leading to
859 cause an extra 18 Tg CO yr^{-1} being up taken up from the atmosphere to soils in
860 sensitivity test when SOC increasing by 30% (Table 5) . CO concentrations will only
861 influence the uptake rate and soil CO substrate concentrations, thus influencing the soil
862 CO consumption rate.

863 Annual CO consumption and net flux have a similar correlation coefficient with to
864 forcing variables and both they are, production and net flux are significantly correlated
865 with air temperature, and soil temperature SOC and atmospheric CO concentration ,
866 due to increasing microbial activities (R > 0.91 globally, Table 6). Increasing
867 temperature will result in increase ing microbial activities, while more SOC increasing
868 will increase soil CO substrate level. Annual CO consumption and net flux have low
869 correlations with annual precipitation and soil moisture, especially at 45°N-45°S (R<0.54
870 Table 6). Specifically, annual Annual CO production is strongly correlated with annual
871 mean SOC, air temperature and soil temperature (R>0.91), while is less correlated with
872 precipitation, soil moisture and atmospheric CO concentration. The annual mean SOC
873 follows air temperature trends (Figure 4) as CO flux. Consumption has low correlations
874 with annual precipitation and soil moisture, especially at 45°N-45°S (Table 6). The soil
875 moisture is significantly influenced by temperature since increasing temperature would
876 result in higher evapotranspiration. In contrast, the monthly consumption and production
877 are correlated with the precipitation and soil moisture in the Northern Hemisphere
878 (R>0.85), which contains over 53% of the global soil CO consumption (Table 3).
879 Meanwhile, the monthly CO consumption, production and net flux flux are is still well
880 correlated with air temperature, and soil temperaturemoisture, and precipitation, and
881 soil moisture (R>0.69 globally Table 6). The soil moisture is significantly influenced by
882 temperature at a in monthly time step scale since increasing temperature would induce
883 result in higher evapotranspiration. Monthly CO consumption, production and net
884 fluxflux have as low correlations with SOC because the soil organic carbonit will not
885 change greatly within a month.

886 The Rcorrelation between annual soil CO consumption and atmospheric CO
887 concentration is 0.91 at the global scale because the atmospheric CO concentration, air
888 temperature, soil temperature dominate the annual consumption rate. At monthly

889 scalestep, this correlationR is -0.48 because global atmospheric CO concentrations are

890 high in winter and low in summer while the simulated soil CO consumption shows an

891 opposite monthly variation(Table 6, Figure 1211), suggesting that other factors such as

892 precipitation, air temperature, and soil temperature are major controls for monthly CO

893 fluxes.

894

4.3. Model Uncertainties and & Limitations

896   There are a number of limitations, contributing to our simulation uncertainties.

897 First, dDue to the lacking of long-period observational data of CO flux and associated

898 environmental factors, the model parameterization using SCE-UA-R method can only

899 be conducted done for 4 ecosystem types including boreal forest, temperate coniferous

900 forest, temperate deciduous forest and grassland, with RMSE ranging from 0.56 to 1.47

901 mg m⁻² day⁻¹. Tropical forest calibration is only conducted using a very limited amount of

902 lab experiment data, but tropical areas are hotspots for CO soil-atmosphere exchanges.

903 Besides, tropical forest SOC for top 30cm can be really high according to observations.

904 TEM model may underestimate the top 30cm SOC, which will underestimate production

905 rates, especially in tropical regions. Tropical regions typically have high temperature

906 during the whole year, which will increase the may result in overestimation ion of CO

907 consumption using due to equation (2.2). The large deviation for tropical savanna

908 (which is mosaic of tropical forest and grassland ecosystems) may be due to using

909 outside air temperature to represent inside air temperature of transparent chamber

910 observations (Varella et al., 2004), and uncertain tropical forest parameterization.

911 Second, wWe used the conclusion from van Asperen et al. (2015) and only considered

912 the thermal-degradation process for CO production in this study. Photo-degradation

913 process and biological formation process were not considered due to lacking

914 understanding of these processes. Third, Moreover, the derived CO surface

915 concentration is lower than MOPITT CO surface concentration, which will lead to

916 overestimation ed of CO deposition velocity during 1901-2100. Fourth, our mModel

917 structure still has a large potential to improve. In this study we have divided the top

918 30cm soil into 30 layers (layer thickness dz=1cm), but finer division will increase the

919 accuracy (Figure 12). We choose dz=1cm because if dz > 1cm, the model vertical CO

concentration profile will deviate from reality and diffusion process will be influenced significantly. ~~I; and~~ if dz < 1cm, it will need much more computing time ~~resources~~ but don't have much improvement compared to dz=1cm (Figure 12a-e). We ~~can~~ notice that the 30-~~-~~layer division well represents ~~s can describe the~~ soil CO concentration profile not only for the days with soil CO net uptake, but also for the days ~~for days~~ with CO net emission (Figure 12c,f). Fifth, Michaelis-Menten function (equation 2.1) is used in this model ~~and~~ ~~but~~ we notice ~~can find~~ that k_{CO} is normally much larger than $C(t,i)$ in those days of net soil uptake (over ten times ~~larger~~~~bigger~~, Figure 12). However, ~~But~~ we can't simplify equation (2.2) to ~~since the soil concentration can be larger than atmosphere in days of net emission and using simplified version like~~ $f_1(C(t,i)) = \frac{C(t,i)}{k_{CO}}$ since CO concentrations in soils can be larger than in the atmosphere in the days of net emissions and ~~will overestimate CO oxidation rate when~~ $C(t,i)$ may be close to ~~is comparable to~~ k_{CO}, which may lead to overestimation of CO oxidation (Figure 12f). Finally, ~~a~~Although we focused on natural ecosystems in this study, land-use change, agriculture activity, and nitrogen deposition also affect the soil CO consumption and production (King, 2002; Chan & Steudler, 2006). For instance, soil CO consumption in agriculture ecosystems is 0 to 9 mg CO m^{-2} day^{-1} in Brazil (King & Hungria, 2002). We used grass land or forest ecosystem to represent agriculture areas in CODM module. Our future study shall include these processes and factors.

5. Conclusions

We analyzed the magnitude, spatial pattern, and the controlling factors of the atmosphere-soil CO exchanges at the global scale for the 20th and 21st centuries using a ~~calibrated~~ process-based biogeochemistry model. Major processes include atmospheric CO diffusion into soils, microbial oxidation removal of CO, and CO production through chemical reaction. We found that air temperature and soil temperature play a dominant role in determining annual soil CO consumption and production while precipitation, air temperature, and soil temperature are the major controls for the monthly consumption and production. Atmospheric CO concentrations are ~~will be~~ important for annual CO consumption. We estimated that the global annual

CO consumption, production and net flux~~es~~ for 2000-2013 ~~the 20th century~~ are -180 to -197, 34 to 36 and -145 to -163~~132-154, 29-36 and 112-119~~ Tg CO yr^{-1}, respectively, when using a MOPITT ~~constant atmospheric~~ CO surface concentration data. ~~The CO consumption reaches 180-197 Tg CO yr^{-1} during 2000-2013 when using atmospheric CO concentrations observed by the MOPITT satellite.~~ Tropical evergreen forest, savanna and deciduous forest areas are the largest sinks accounting for 66% of the total CO consumption, while the Northern Hemisphere consumes 59~~60~~% of the global total. During the 20th century, the estimated CO deposition velocity is 0.16-0.19 mm s^{-1}. ~~During the 21st century,~~ T~~t~~he predicted CO deposition velocity ~~net CO flux~~ will reach 0.20-0.30 mm s^{-1}~~126-150 Tg CO yr^{-1}~~ in the 2090s, primarily because of increasing air temperature. The areas near the equator, eastern Asia, Europe and eastern US will become the sink hotspots because they have warm and moist soils. This study calls for long-period observations of CO flux for various ecosystem types and projection of atmospheric CO surface concentrations from 1901-2100 to improve future estimates ~~models~~of global soil CO consumption. The effects of land-use change, agriculture activities, nitrogen deposition, photo-degradation and biological formation shall also be considered to improve future quantification of soil CO fluxes.

37

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Table 1. Model parameterization sites for thermal and hydrology modules (site No. 1-4) and for CODM module

(site No. 5-13)

No.	Site Name	Location	Vegetation	Driving Climate	Observed Data	Source and Comments
1	Poker Flat Research Range Black Spruce Forest (US_PRR)	147°29'W/65°7'N	Boreal Evergreen Needle Leaf Forests	Site Observation & ERA Interim	Soil Temperature and Moisutre of 2011-2014	Suzuki (2016)
2	Morgan Monroe State Forest (US_MMS)	86°25W/39°19'N	Temperate Deciduous Broadleaf Forests	Site Observation & ERA Interim	Soil Temperature and Moisutre of 1999-2014	Philip and Novick (2016)
3	Santarem, Tapajos National Forest (STM_K83)	54°56'W/3°3'S	Tropical Moist Forest	Site Observation & ERA Interim	Soil Temperature and Moisutre of 2000-2004	SALESKA et al. (2013)
4	Bananal Island Site (TOC_BAN)	50°08'W/9°49'S	Tropical Forest-Savanna	Site Observation & ERA Interim	Soil Temperature and Moisutre of 2003-2006	SALESKA et al. (2013)
5	Eastern Finland (EF)	27°14E/63°9'N	Boreal Grassland	Site Observation & ERA Interim	CO flux of April-November,2011	Pihlatie et.al. (2016)
6	Viterbo, Italy (VI)	11°55'E/42°22'N	Mediterranean Grassland	Site Observation & ERA Interim	CO flux of August, 2013	van Asperen et al. (2015)
7	Brasilia, Brazil (BB)	47°51'W/15°56'S	Tropical Savanna	Site Observation & CRU	CO flux of October 1999 to July 2001	Varella et al. (2004)
8	Orange County, North Carolina (OC)	79°7'W/35°58'N	Temperate Coniferous Forest	AMF_US-Dk3 2002-2003	CO flux of March 2002 to March 2003	Fisher (2003)
9	Tsukuba Science City, Japan (TSC)	140°7'E/36°01'N	Temperate Mixed Forest	Site Observation & ERA Interim	CO flux of July 1996 to September 1997	Yonemura et. al. (2000)
10	Manitoba, Canada (CBS)	96°44'W/56°09'N	Boreal Pine Forest	Site Observation & AMF_CA-Man	CO flux of June-August, 1994	Kuhlbusch et. al (1998)
11	Scotland, U.K. (SUK)	3°12'W/55°51'N	Temperate Deciduous Forests	ERA Interim 1995	CO flux of 1995	Moxley and Smith (1998)
12	Alaska, USA (AUS)	147°41'W/64°52'N	Boreal wetland	CRU 1991	CO flux of Lab Experiment,1991	Funk et al. (1994)
13	Guayana Shield,Bolivar State,Venezuela (GBV)	62°57'W/7°51'N	Tropical Smideciduous Forest	CRU 1985	CO flux of Lab Experiment,1985	Scharffe et al. (1990)

Table 2. Ecosystem-specific parameters in the CODM module[a]

	Ecosystem Type	k_{CO} (ul CO l^{-1})	$Vmax$ (ug CO $g^{-1}h^{-1}$)	T_{ref} (°C)	$Q10$ (Unitless)	M_{min} ($\frac{v}{v}$)	M_{max} ($\frac{v}{v}$)	M_{opt} ($\frac{v}{v}$)	E_{SOC}	F_{SOC} ($\frac{g}{g}$)	$\frac{Ea_{ref}}{R}$ (K)	PM_{ref} ($\frac{v}{v}$)	PT_{ref} (°C)
1	Alpine Tundra & Polar Desert	36.00	0.78	4.00	1.80	0.10	1.00	0.55	3.00	0.33	7700	0.25	30.00
2	Wet Tundra	36.00	0.70	4.00	1.80	0.25	1.00	0.55	3.00	0.42	7700	0.25	30.00
3	Boreal Forest	27.34	1.18	9.81	1.60	0.15	0.64	0.53	2.98	0.50	8827	0.35	26.99
4	Temperate Coniferous Forest	42.64	2.15	6.90	1.87	0.02	0.96	0.53	2.86	0.50	8404	0.38	31.52
5	Temperate Deciduous Forest	40.16	2.43	8.54	1.51	0.17	0.81	0.51	2.45	0.50	8801	0.35	37.44
6	Grassland	42.41	0.49	11.27	1.65	0.16	0.82	0.51	3.09	0.42	14165	0.24	12.29
7	Xeric Shrublands	8.00	0.30	4.00	1.50	0.10	1.00	0.55	3.00	0.33	7700	0.25	30.00
8	Tropical Forest	45.00	2.00	4.00	1.50	0.10	1.00	0.55	3.80	0.50	14000	0.50	18.00
9	Xeric Woodland	8.00	0.30	4.00	1.50	0.10	1.00	0.55	3.00	0.50	7700	0.25	30.00
10	Temperate Evergreen Broadleaf Forest	40.16	2.43	8.54	1.51	0.17	0.81	0.51	2.45	0.50	8801	0.35	37.44
11	Mediterranean Shrubland	45.00	1.50	4.00	1.50	0.10	1.00	0.55	3.00	0.33	7700	0.25	30.00
**	Largest Potential Value	51.00	11.1	15.00	2.00	0.30	1.00	0.60	3.80	--	15000	0.60	40.00

[a] k_{CO} is the half-saturation constant for soil CO concentration; $Vmax$ is the specific maximum CO oxidation rate; T_{ref} is the reference temperature to account for soil temperature effects on CO consumption; $Q10$ is the an ecosystem-specific Q10 coefficient to account for soil temperature effects on CO consumption; M_{min}, M_{max}, M_{opt} are the minimum, optimum, and maximum volumetric soil moistures of oxidation reaction to account for soil moisture effects on CO consumption; E_{SOC} is an estimated nominal CO production factor, similar as Potter et al. (1996) (10^{-4} mg CO m^{-2} d^{-1} per g SOC m^{-2}); F_{SOC} is a constant fraction of top 20cm SOC compared to total amount of SOC to account for SOC effects on CO production; Ea_{ref}/R is the is the ecosystem-specific activation energy divided by gas constant to account for the reaction rate of production; PM_{ref} is the reference moisture to account for soil temperature effects on CO production; PT_{ref} is the reference temperature to account for soil temperature effects on CO production

Table 3. Regional soil CO consumption, net flux and production (Tg CO yr^{-1}) ~~during 1901- 2013 (E1) and~~

during 2000-2013 ~~with MOPITT data transient CO surface concentration~~

	South-45S	45S-0	0-45N	45N-North	Global
Consumption	0.22	75.77	91.66	18.90	186.55
Net flux	0.13	59.34	77.17	14.63	151.27
Production	0.09	16.43	14.49	4.27	35.28

Table 4. ~~Global~~ Annual total soil CO consumption, net flux and production in different ecosystems during

2000~~1901~~-2013 (E1) and mean CO deposition velocity ~~ ~~ in different ecosystems during 1901-2013 (E2)

Vegetation Type	Area (10^6 km^2)	Pixels	Consumption (Tg CO yr^{-1})	Net flux (Tg CO yr^{-1})	Production (Tg CO yr^{-1})	Deposition velocity (mm s^{-1})
Alpine Tundra & Polar Desert	5.28	3580	-0.92	-0.69	0.23	0.023
Wet Tundra	5.24	4212	-1.00	-0.42	0.58	0.015
Boreal Forest	12.47	7578	-7.76	-6.01	1.75	0.070
Forested Boreal Wetland	0.23	130	-0.14	-0.09	0.04	0.109
Boreal Woodland	6.48	4545	-2.48	-1.54	0.94	0.036
Non-Forested Boreal Wetland	0.83	623	-0.35	-0.18	0.17	0.029
Mixed Temperate Forest	5.25	2320	-10.49	-9.98	0.51	0.204
Temperate Coniferous Forest	2.49	1127	-3.51	-3.21	0.30	0.185
Temperate Deciduous Forests	3.65	1666	-5.07	-4.83	0.25	0.151
Temperate Forested Wetland	0.15	60	-0.35	-0.35	0.01	0.281
Tall Grassland	3.63	1567	-1.66	-0.65	1.01	0.021
Short Grassland	4.71	2072	-1.05	-0.27	0.78	0.010
Tropical Savanna	13.85	4666	-21.86	-15.88	5.98	0.234
Xeric Shrubland	14.71	5784	-1.95	-1.64	0.31	0.021
Tropical Evergreen Forest	17.77	5855	-85.90	-69.66	16.24	0.879
Tropical Forested Wetland	0.55	178	-3.59	-3.09	0.50	1.154
Tropical Deciduous Forest	4.69	1606	-14.81	-11.78	3.03	0.532
Xeric Woodland	6.85	2387	-8.48	-7.44	1.04	0.246
Tropical Forested Floodplain	0.15	50	-0.89	-0.77	0.12	1.117
Desert	11.61	4170	-0.62	-0.57	0.05	0.008
Tropical Non-forested Wetland	0.06	19	-0.03	-0.02	0.01	0.067
Tropical Non-forested Floodplain	0.36	120	-0.35	-0.24	0.10	0.083
Temperate Non-Forested Weland	0.34	120	-0.33	-0.20	0.14	0.089
Temperate Forested Floodplain	0.10	48	-0.13	-0.12	0.00	0.197
Temperate Non-forested Floodplain	0.10	45	-0.05	-0.03	0.02	0.050
Wet Savanna	0.16	59	-0.39	-0.32	0.07	0.434
Salt Marsh	0.09	35	-0.05	-0.03	0.03	0.035
Mangroves	0.12	38	-0.49	-0.41	0.08	0.809
Temperate Savannas	6.83	2921	-3.83	-3.22	0.61	0.076
Temperate Evergreen Broadleaf	3.33	1268	-7.17	-6.95	0.22	0.252
Mediterranean Shrubland	1.47	575	-0.86	-0.71	0.16	0.100
Total	133.56	59424	-186.55	-151.27	35.28	--

1240 **Table 5.** Sensitivity of global CO consumption, net flux and production (~~units are~~ Tg CO yr^{-1}) to changes in

1241 atmospheric CO, soil organic carbon (SOC), precipitation (Prec) and air temperature (AT)

	Baseline	CO +30%	CO -30%	SOC +30%	SOC -30%	Prec +30%	Prec -30%	AT +3°C	AT -3°C
Consumption	-147.65	-164.14	-131.12	-175.37	-119.90	-150.72	-143.50	-190.59	-114.83
Change (%)	0.00	-11.17	11.19	-18.78	18.79	-2.08	2.81	-29.09	22.23
Net flux	-113.65	-130.15	-97.12	-131.18	-96.10	-116.97	-109.32	-144.23	-89.58
Change (%)	0.00	-14.51	14.54	-15.42	15.44	-2.92	3.81	-26.90	21.18
Production	33.99	33.99	33.99	44.19	23.80	33.74	34.17	46.36	25.25
Change (%)	0.00	0.00	0.00	30.00	-30.00	-0.75	0.53	36.39	-25.72

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Table 6. <u>Correlation coefficient</u>s between ~~Effects of annual and monthly climate~~ <u>forcing variables</u> (precipitation (Prec), air temperature (Tair), soil organic carbon (SOC), soil temperature (Tsoil), soil moisture (Msoil) and atmospheric CO (CO air)<u> and</u> ~~on~~ absolute values of consumption, production and net flux for different regions and the globe ~~during the 20th Century~~

|		Monthly					Annual				
		North-45°N	45°N-0°	0°-45°S	45°S-South	Global	North-45°N	45°N-0°	0°-45°S	45°S-South	Global
Prec	Consumption	0.91	0.96	0.92	-0.34	0.87	0.65	0.21	0.26	0.13	0.52
	Production	0.91	0.70	0.45	-0.34	0.82	0.63	0.10	0.15	-0.11	0.47
	Net flux	0.91	0.97	0.94	-0.33	0.87	0.65	0.25	0.31	0.32	0.54
Tair	Consumption	0.97	0.98	0.91	0.96	0.95	0.92	0.93	0.88	0.84	0.91
	Production	0.96	0.83	0.72	0.98	0.94	0.92	0.92	0.91	0.95	0.91
	Net Flux	0.97	0.97	0.88	0.90	0.95	0.91	0.92	0.85	0.62	0.91
SOC	Consumption	-0.19	0.07	0.21	-0.01	0.15	0.68	0.90	0.92	0.47	0.92
	Production	-0.19	0.31	0.47	-0.02	0.24	0.72	0.92	0.92	0.50	0.93
	Net Flux	-0.19	0.03	0.14	0.00	0.13	0.67	0.88	0.91	0.38	0.91
Tsoil	Consumption	0.97	0.98	0.92	0.96	0.95	0.94	0.93	0.88	0.85	0.95
	Production	0.97	0.83	0.72	0.98	0.94	0.94	0.92	0.91	0.96	0.95
	Net Flux	0.98	0.97	0.88	0.90	0.95	0.93	0.93	0.86	0.63	0.95
Msoil	Consumption	0.85	0.96	0.92	0.19	0.76	0.03	0.22	0.14	0.26	0.22
	Production	0.85	0.75	0.44	0.14	0.69	-0.02	0.12	0.02	0.05	0.17
	Net Flux	0.84	0.96	0.95	0.25	0.77	0.04	0.26	0.19	0.40	0.24
CO Air	Consumption	-0.66	-0.76	-0.29	0.14	-0.48	0.87	0.88	0.81	0.98	0.91
	Production	-0.70	-0.66	0.08	-0.40	-0.66	-0.36	-0.48	-0.54	-0.44	-0.57
	Net Flux	-0.64	-0.73	-0.35	0.55	-0.41	0.92	0.91	0.88	0.99	0.94

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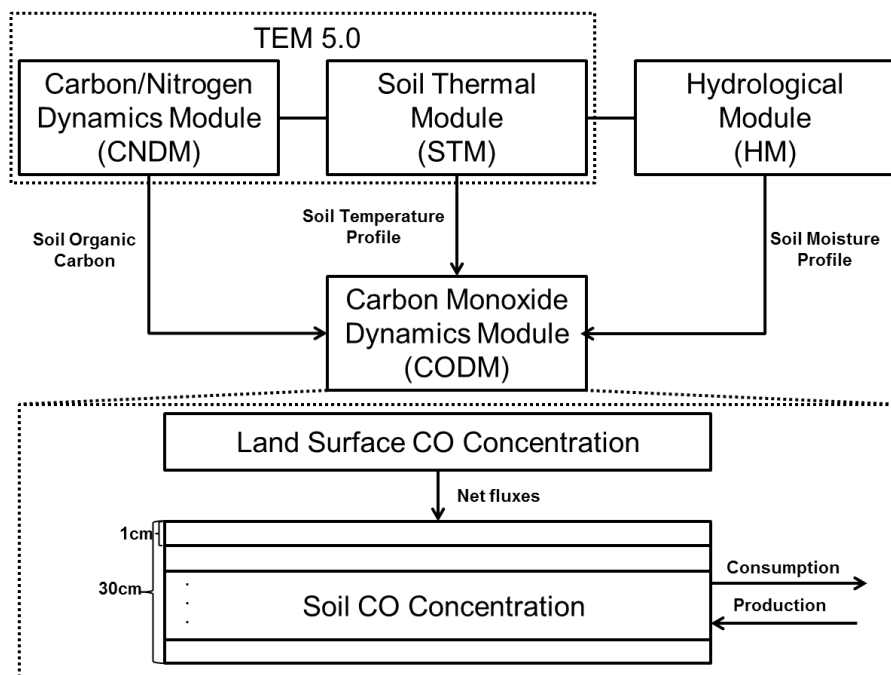
Figure 1. The model framework includes a carbon and nitrogen dynamics module (CNDM), a soil thermal module (STM) from Terrestrial Ecosystem Model (TEM) 5.0 (Zhuang et al., 2001, 2003), a hydrological module (HM) based on a Land Surface Module (Bonan, 1996; Zhuang et al., 2004), and a carbon monoxide dynamics module (CODM). The detailed structure of CODM includes land surface CO concentration as top boundary and thirty 1 cm thick layers (totally 30 cm) where consumption and production take placewould happen inside.

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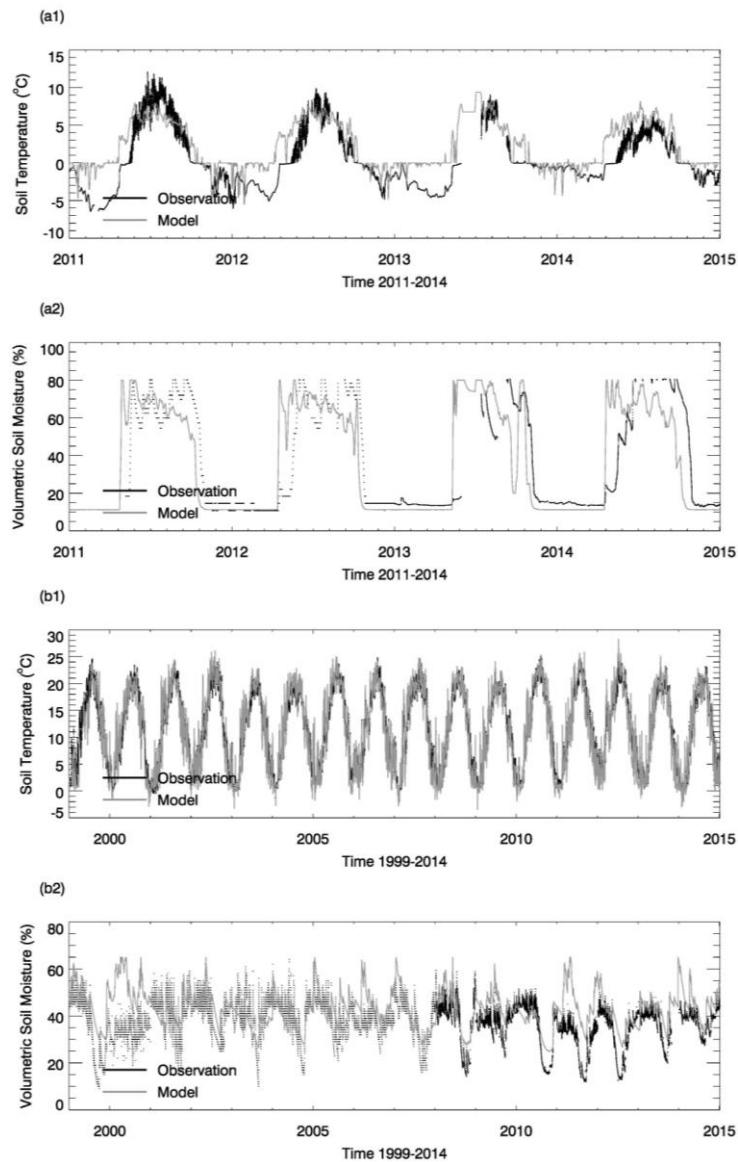
Figure 2. Evaluation of thermal and hydrology module at four sites: (a) Boreal Evergreen Needle Leaf Forests, (b) Temperate Deciduous Broadleaf Forests. (1) shows the soil temperature comparison between model simulations (gray line) and observations (black line) and (2) shows the soil moisture comparison between model simulations (gray line) and observations (black line). Specifically, the volumetric soil moisture is converted from the water content reflectometry (WCR)

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318 probe output period using an empirical calibration function of Bourgeau-Chavez et al. (2012) for 5cm-30cm layer. (talk in

319 results 3.1 part) Some of them resulted in calculations of values greater than 100% VSM in Nakai et al. (2013) study. Our

320 model estimated high VSM (close to 80%) is due to top 10 cm moss in the model which has a saturation VSM of 0.8
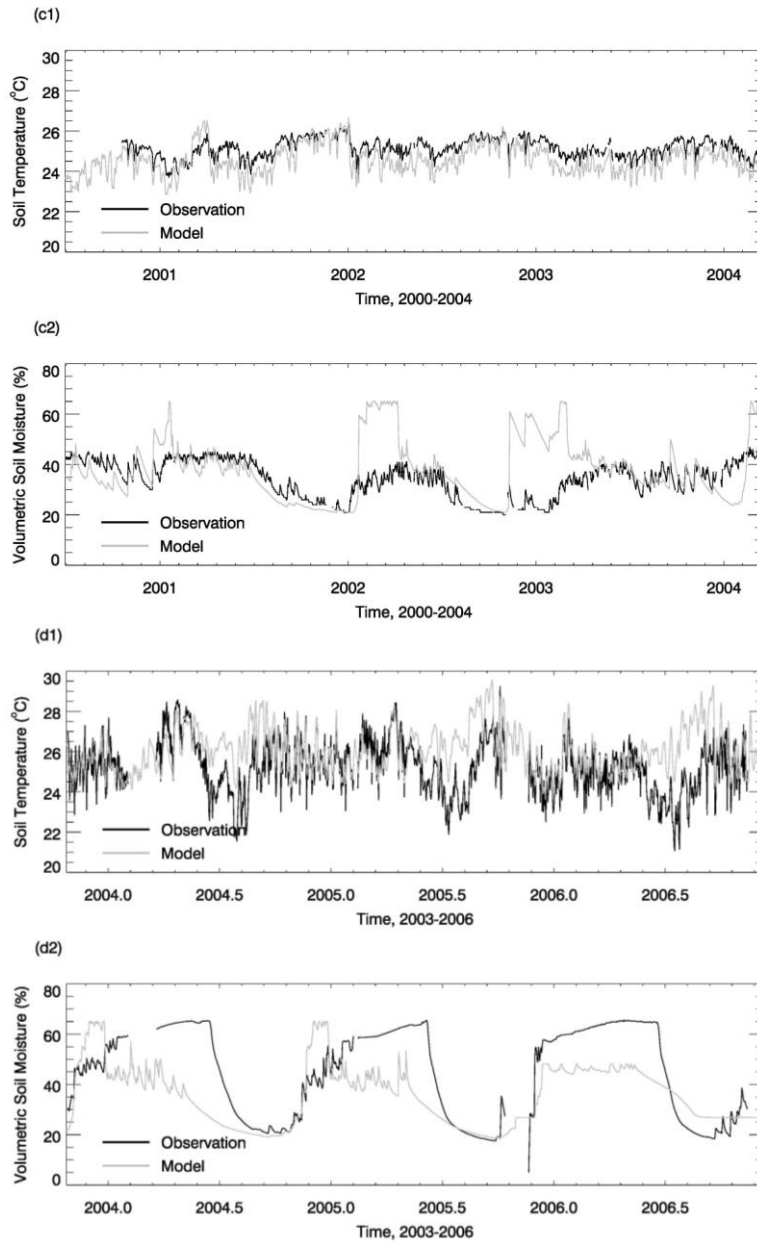
Figure 2. Contd. Evaluation of thermal and hydrology module at four sites: (c) Tropical Moist Forest, (d) Tropical Forest-Savanna. (1) shows the soil temperature comparison between model simulations (gray line) and observations (black line) and (2) shows the soil moisture comparison between model simulations (gray line) and observations (black line)

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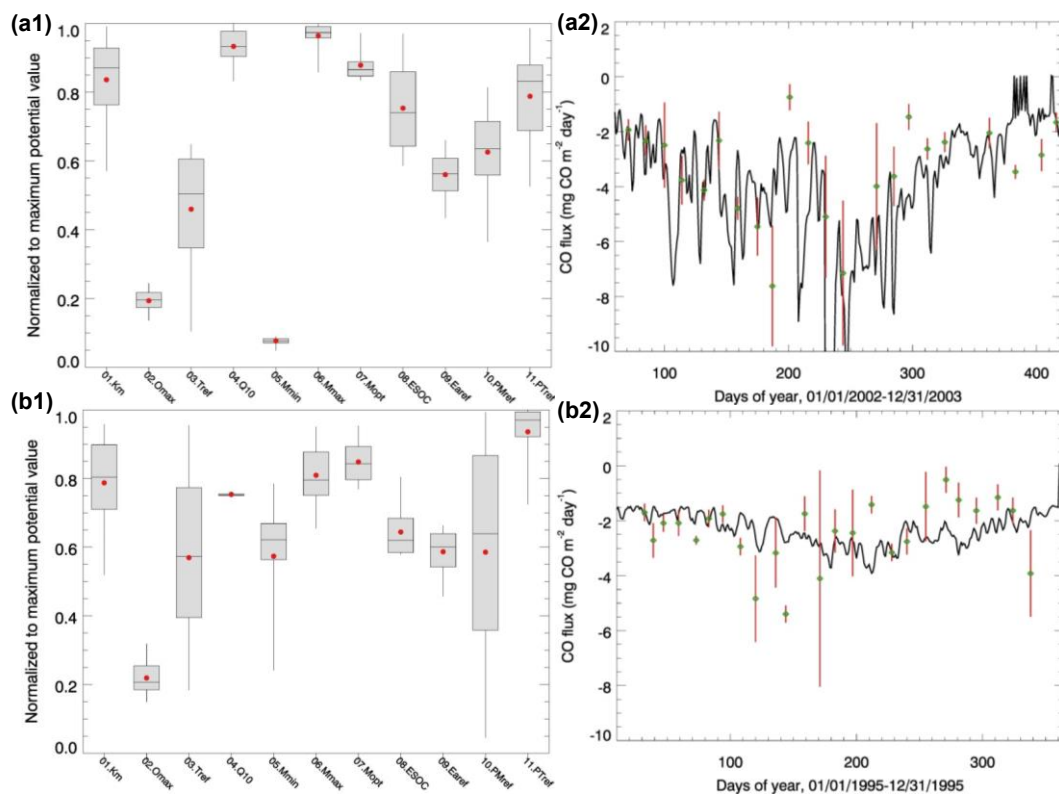
Figure 3. Parameter ensemble experiment results: Each parameter has 50 calibrated values generated from running SCE-UA-R 50 times independently. Parameters are normalized to their largest potential values described in Table 2. (a1) and (a2) are temperate coniferous forest normalized parameter distribution boxplots and CO flux comparisons between model simulations (solid line, using mean value of parameters) and observations (green diamond"+", red lines represent error bar, site No.8), respectively. For each box, line top, box top, horizontal line inside box, box bottom and line bottom represent maximum, third quartile, median, first quartile and minimum of 50 parameter values. Red dot represents the mean value of 50 parameter values. (b1) and (b2) are plots for temperate deciduous forest (site No.11); (c1) and (c2) are for boreal forest; (d1) and (d2) are for grassland. Grassland observation data is the sum of hourly observations so there is no error bar presented.

(c1) **(c2)** **(d1)** **(d2)**
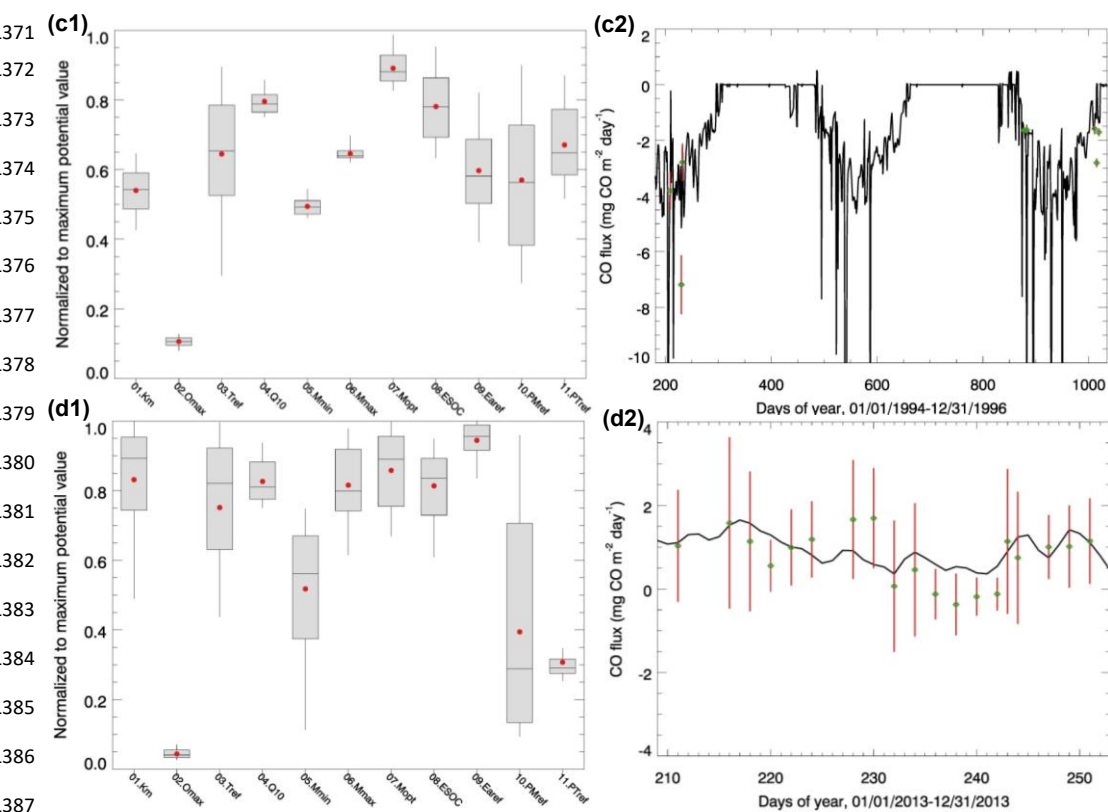
Figure 3. Contd. Parameter ensemble experiment results: Each parameter has 50 calibrated values generated from running SCE-UA-R 50 times independently. Parameters are normalized to their largest potential values described in Table 2. (ca1) and (ca2) are borealtemperate coniferous forest normalized parameter distribution boxplots and CO flux comparisons between model simulations (solid line, using mean value of parameters) and observations (green diamond"+", red lines represent error bar, site No. 12), respectively. For each box, line top, box top, horizontal line inside box, box bottom and line bottom represent maximum, third quartile, median, first quartile and minimum of 50 parameter values. Red dot represents the mean value of 50 parameter values. (b1) and (b2) are plots for temperate deciduous forest; (c1) and (c2) are for boreal forest; (d1) and (d2) are for grassland (site No.6). Grassland observation data is the sum of hourly observations so error bar represented the standard deviationthere is no error bar presented.
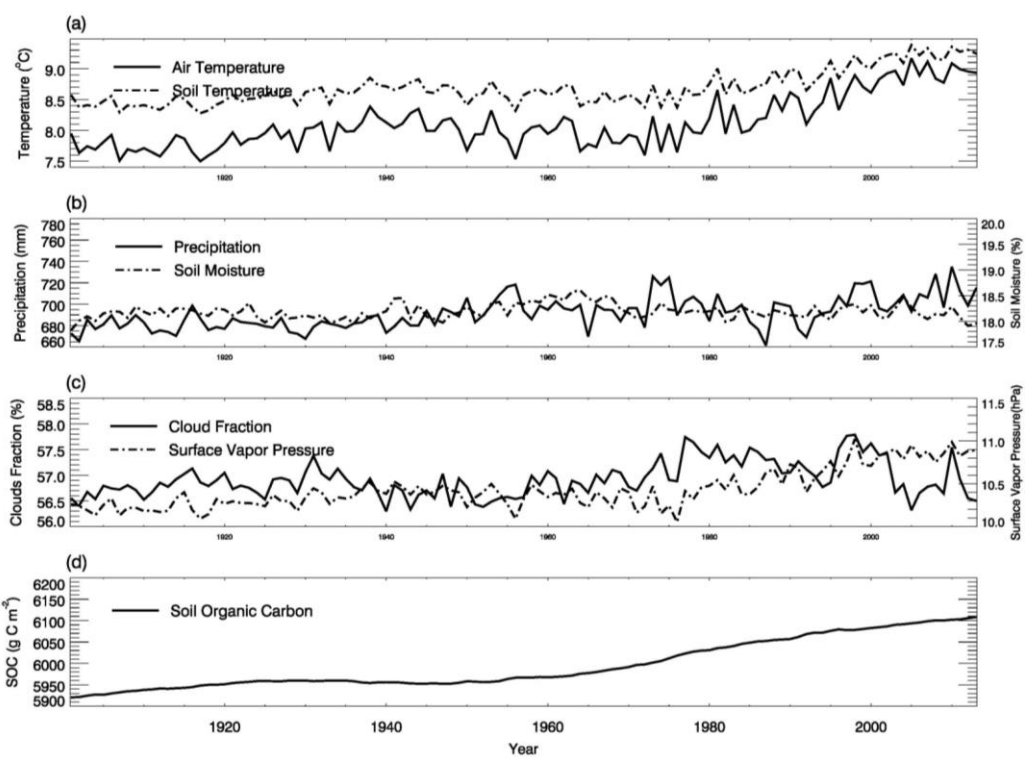
Figure 4. Historical global land surface (excluding e Antarctic area and ocean area) mean climate, and simulated global mean soil moisture, soil temperature and SOC for the period 1901-2013.
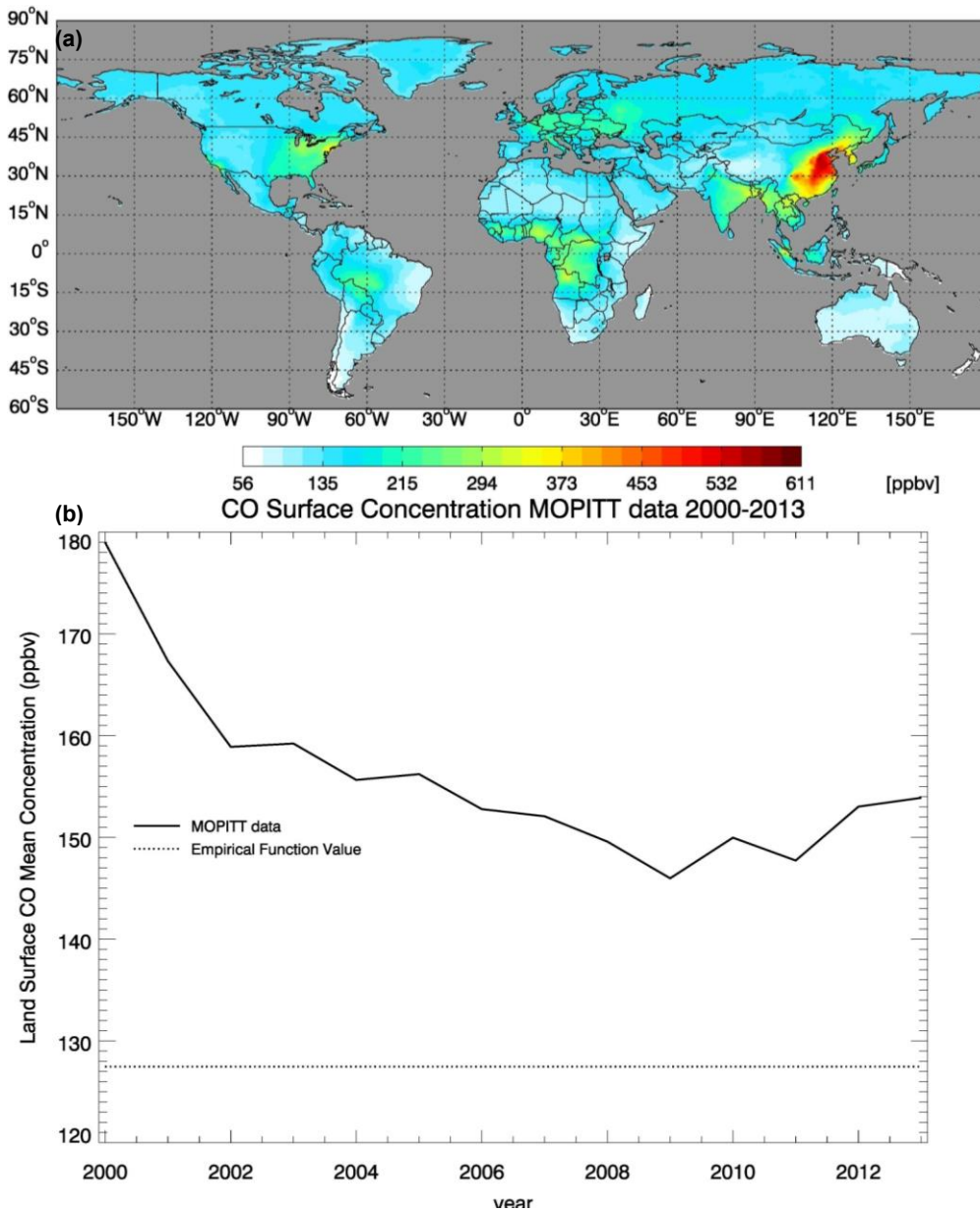
Figure 5. CO surface concentration data from MOPITT satellite (ppbv): (a) global mean CO surface concentrations from MOPITT during 2000-2013; (b) the CO annual surface concentrations from both MOPITT and empirical functions (Potter et al., 1996).
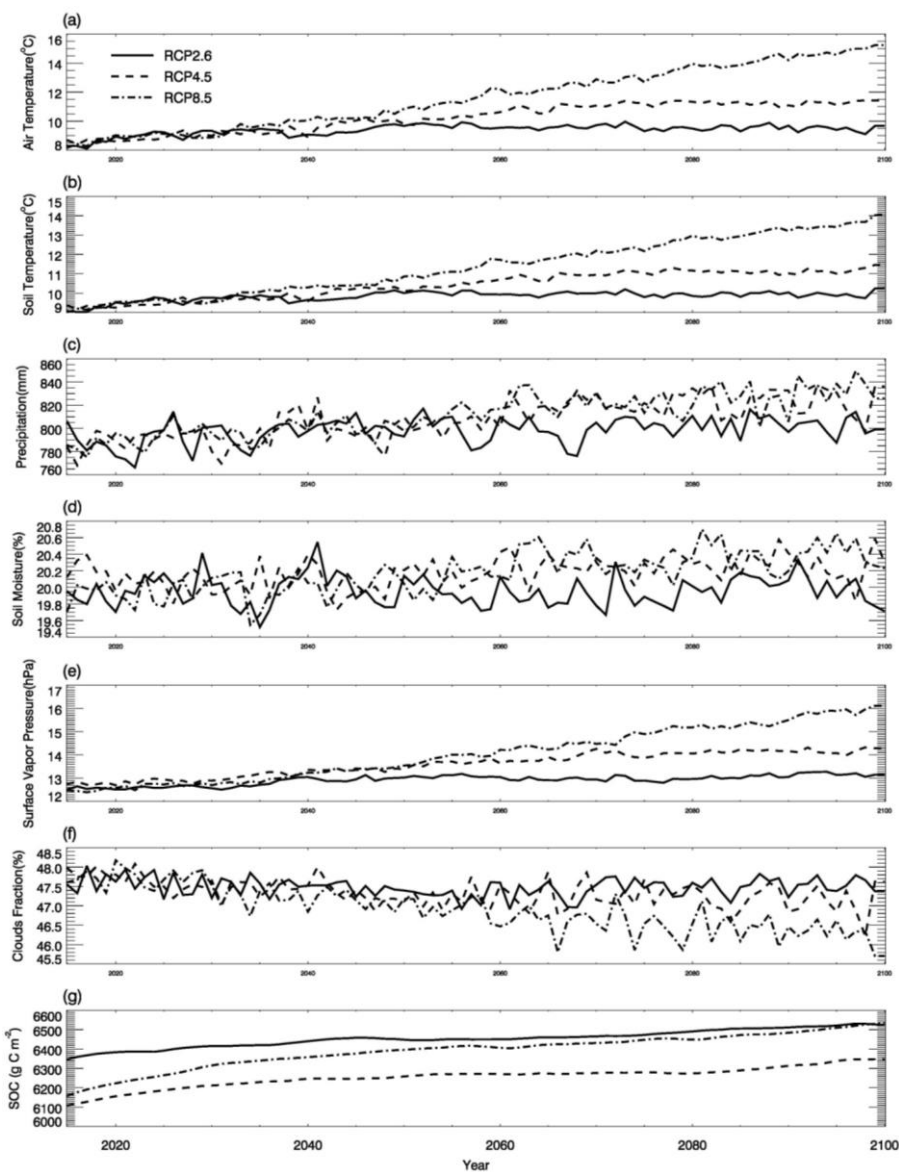
Figure 6. Global land surface (excluding e Antarctic area and ocean area) mean climate from RCP2.6, RCP4.5 and RCP8.5 data sets and simulated global mean soil temperature, moisture and SOC: (a)-(g) are land surface air temperature (°C), soil temperature (°C), precipitation (mm), soil moisture (%), surface water vapor pressure (hpa), cloud fraction (%), and SOC (mg m^{-2}), respectively.

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(a)

(b)
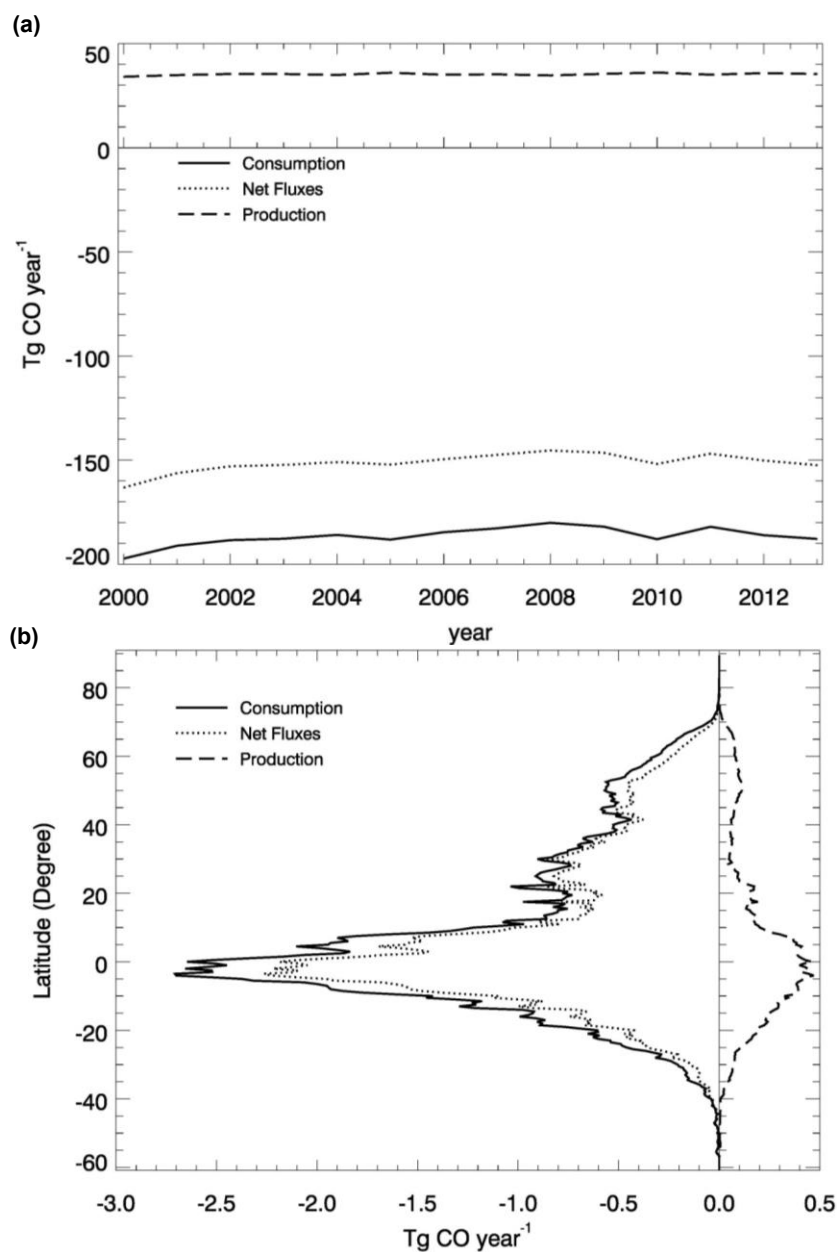
Figure 79. Global ~~annual~~ mean ~~latitudinal distributions of~~ soil CO consumption, production and net flux: (a) annual time series during 2000-2013~~during 1901-2013 (Tg CO yr⁻¹) estimated with constant CO surface concentration data~~ and (b) latitudinal ~~e~~distribution during 2000-2013~~during 2000-2013 estimated with MOPITT CO surface concentration data~~.

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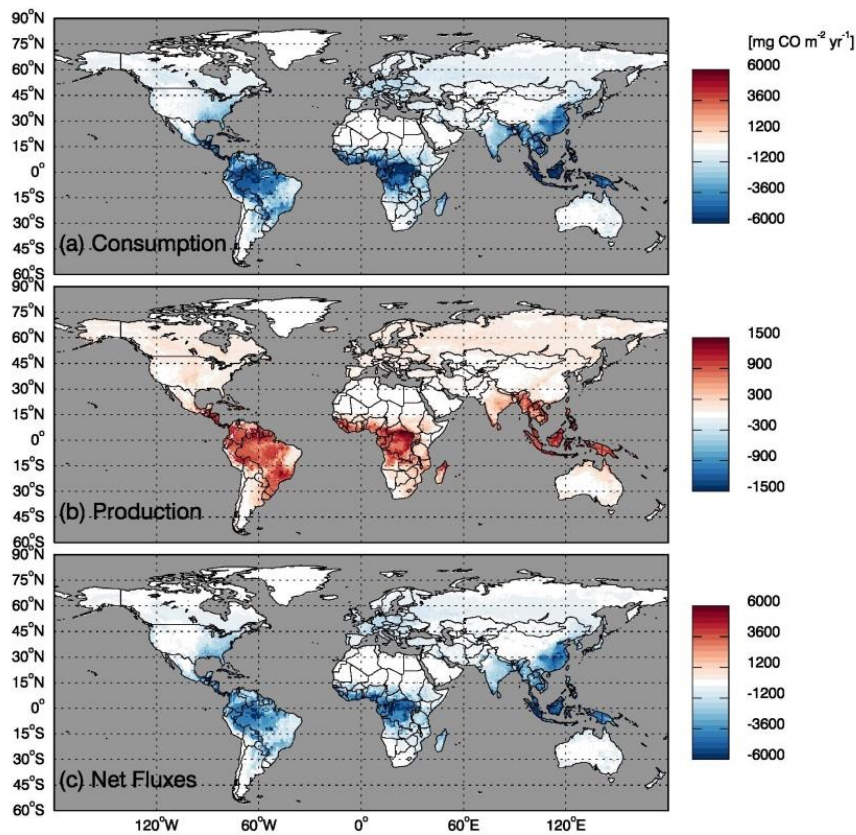
Figure 87. Global annual mean soil CO fluxes (mg CO m^{-2} yr^{-1}) ~~during 1901-2013, estimated using constant CO concentration data (left side) and mean annual global soil CO fluxes~~ during 2000-2013 using
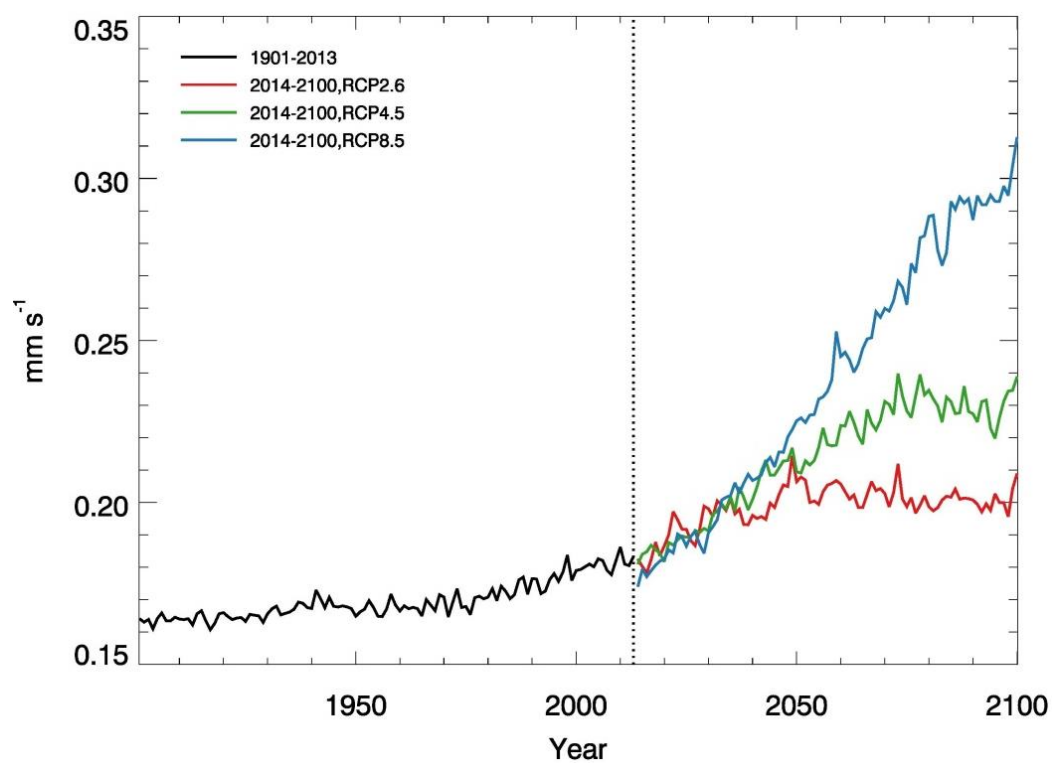
Figure 911. Future Global mean annual time series of CO deposition velocity soil CO consumption, net flux and production (mm s^{-1}Tg CO yr^{-1}) using constant in time, spatially distributed CO concentration data input under historical climate scenarios during 1901-2013 (left side of dot line) and under future climate scenarios RCP2.6, RCP4.5 and RCP8.5 during 2014-2100 (right side of dot line)
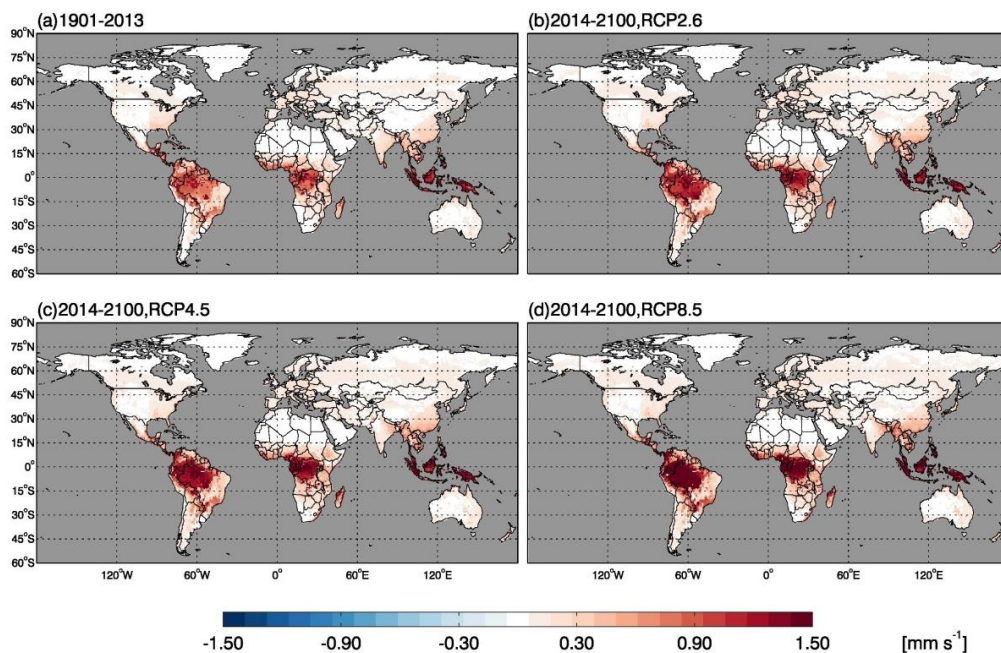
Figure 10. Global annual mean CO deposition velocity using constant in time, spatially distributed CO concentration data input consumption, production and net flux (mg CO m⁻² yr⁻¹ mm s⁻¹) a) under historical climate scenarios during 1901-2013 and b), c), d) under future climate scenarios RCP2.6, RCP4.5 and RCP8.5 during 2014-2100, respectively,
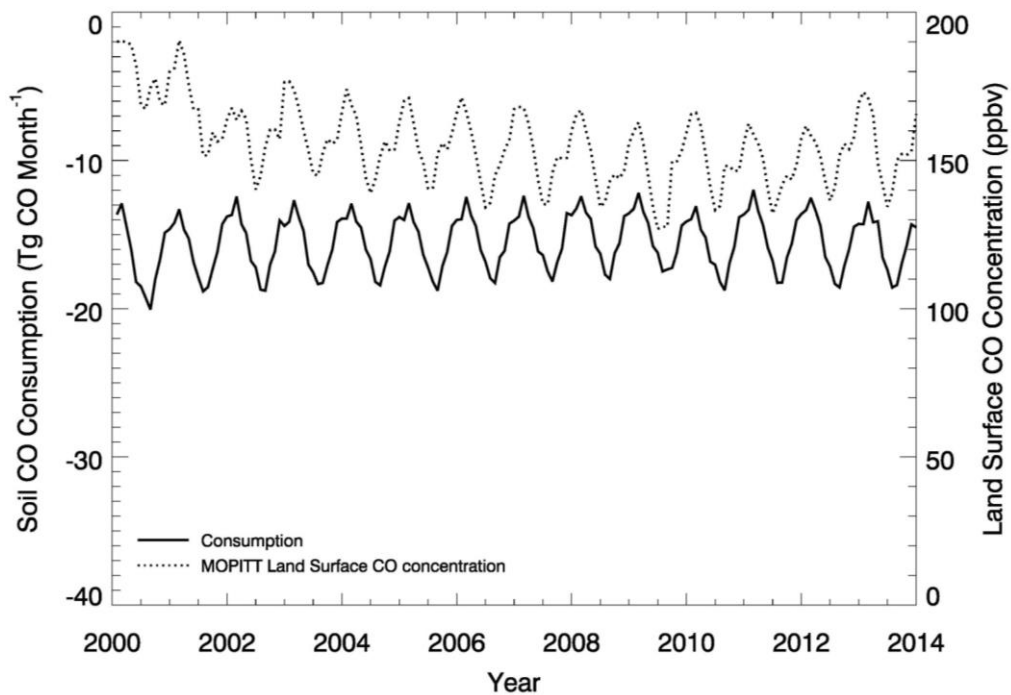
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Figure 1112. Global mean mMonthly time series of MOPITT surface atmospheric CO concentration (ppbv) and soil CO consumption from model simulations E1 and E2 (Tg CO mon^{-1})
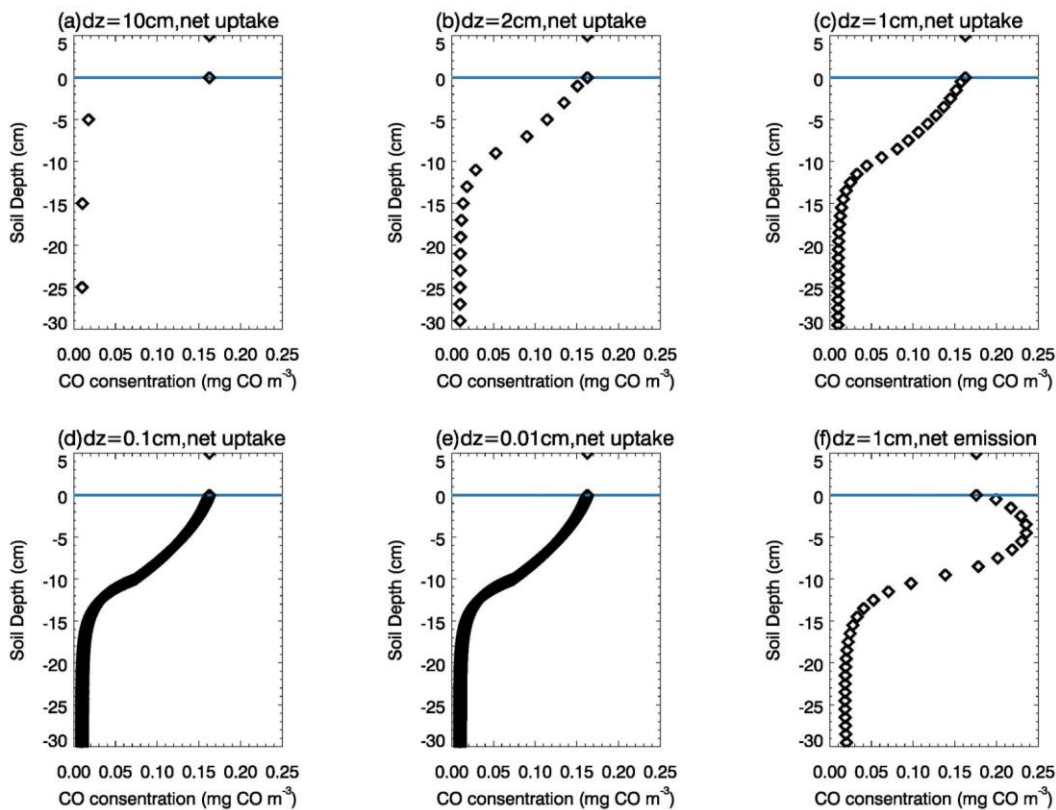
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Figure 12. Daily mean vertical soil CO concentration profiles of top 30cm. In soils (depth < 0cm), black diamonds represent the soil CO concentration (mg CO m^{-3}). Above the surface (depth>=0cm), black diamonds represent atmospheric CO concentration. a), b), c), d) and e) are the results from the same day when soil is a net sink of CO but with different layer thickness (dz=10cm, 2cm, 1cm, 0.1cm and 0.01cm respectively); f) is the result from the day when soil is a net source of CO, with dz=1cm.