1 Author's response to co-editor:

2	Thank you very much for your comment! You helped us improve this study significantly.
3	Comment: The referee asked why the (negative) deposition flux should INcrease when the SOC
4	increases (table 5 and 7). This question is based on the assumption that higher SOC provides more
5	substrate for CO formation in the soil, which would DEcrease the soil-atmosphere gradient and DEcrease
6	the deposition flux. There must be one process in your model that leads to an increase in the deposition
7	flux.
8	You argue now with time step arguments, which apparently play a role, but even with a short timestep
9	the sign of the flux change remains the same, even if the magnitude is reduced. I think it is still required
10	that you explain the physical basis of the effect. And then it would also be good if you can argue a bit
11	more quantitatively why the timestep has such a large influence (and that this then does not affect
12	other results of your model).
13	Response: Thank you for your suggestions to improve the sensitivity analysis presented in the paper.
13 14	Response: Thank you for your suggestions to improve the sensitivity analysis presented in the paper. In this revision, we removed Table 7 to avoid distraction from our main focus. We have also revised
13 14 15	Response: Thank you for your suggestions to improve the sensitivity analysis presented in the paper. In this revision, we removed Table 7 to avoid distraction from our main focus. We have also revised the sensitivity test (Table 5) by using SOC ±5% instead of ±30% (not a realistic variation), since during
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13 14 15 16 17	Response: Thank you for your suggestions to improve the sensitivity analysis presented in the paper. In this revision, we removed Table 7 to avoid distraction from our main focus. We have also revised the sensitivity test (Table 5) by using SOC ±5% instead of ±30% (not a realistic variation), since during our century-scale simulations, the SOC will not change beyond 4% within a century. This small variation of SOC did not affect CO consumption drastically, in contrast to the large effects due to
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13 14 15 16 17 18 19 20 21	Response: Thank you for your suggestions to improve the sensitivity analysis presented in the paper. In this revision, we removed Table 7 to avoid distraction from our main focus. We have also revised the sensitivity test (Table 5) by using SOC ±5% instead of ±30% (not a realistic variation), since during our century-scale simulations, the SOC will not change beyond 4% within a century. This small variation of SOC did not affect CO consumption drastically, in contrast to the large effects due to sudden high and unrealistic SOC changes in the original sensitivity test. Consequently, our other results of the global CO consumption simulations were not significantly affected by small variations of SOC (less than 4%). We followed your suggestion to mainly explain the physical basis of the effect in this revision. Please
13 14 15 16 17 18 19 20 21 22	Response: Thank you for your suggestions to improve the sensitivity analysis presented in the paper.In this revision, we removed Table 7 to avoid distraction from our main focus. We have also revisedthe sensitivity test (Table 5) by using SOC ±5% instead of ±30% (not a realistic variation), since duringour century-scale simulations, the SOC will not change beyond 4% within a century. This smallvariation of SOC did not affect CO consumption drastically, in contrast to the large effects due tosudden high and unrealistic SOC changes in the original sensitivity test. Consequently, our otherresults of the global CO consumption simulations were not significantly affected by small variations ofSOC (less than 4%).We followed your suggestion to mainly explain the physical basis of the effect in this revision. Pleasefind our changes for Table 5 and revisions on lines 126, 296, 364, 367, and 404-405 for sensitivity test

24 "Fourth, from sensitivity test (Table 5) we notice that SOC increasing (5%) resulted in a

25	net flux increase (2.57%). The SOC increase enhanced CO production (Equation 3),
26	CO concentrations (Equation 1), and CO oxidation (Equation 2). When the change of
27	total oxidation is larger than the difference between the change of total production and
28	the change of total soil CO concentration (Equation 1), the estimate of the net flux
29	change is negative (from atmosphere to soil) using a mass balance approach (Section
30	2.2), leading to a 2.57% increase in net flux in our SOC sensitive test. This is due to the
31	fact that CO production (Equation 3) is calculated independently from oxidation
32	calculation (Equation 2). This will not influence our other results since SOC varies
33	slightly during our simulation periods with only a 3% increase from 1900 to 2013 (Figure
34	4d) and up to a 4% increase from 2014 to 2100 (Figure 6g). This artifact problem in
35	SOC sensitivity test can be alleviated using a very fine time step (e.g., 1 second),
36	because CO concentrations will change slightly within the short time, allowing net flux
37	roughly equals the difference between production and oxidation within the short time
38	step. If the change of production is bigger than the change of oxidation, the change of
39	net flux will be positive, leading to a decrease of deposit to soil. The downside is that
40	running the model at one second time step will require significantly high computing time.

42 Global Soil Consumption of Atmospheric Carbon Monoxide:

43 An Analysis Using a Process-Based Biogeochemistry Model

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49 Abstract: Carbon monoxide (CO) plays an important role in controlling the 50 oxidizing capacity of the atmosphere by reacting with OH radicals that affect 51 atmospheric methane (CH₄) dynamics. We develop a process-based biogeochemistry model to quantify CO exchange between soils and the atmosphere with a 5-minute 52 53 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. 54 The model is then extrapolated to the global terrestrial ecosystems using monthly 55 climate forcing data. Global soil gross consumption, gross production, and net flux of 56 the atmospheric CO are estimated to be from -197 to -180, 34 to 36, and -163 to -145 57 Tg CO yr⁻¹ (1Tg = 10^{12} g), respectively, driven with satellite-based atmospheric CO 58 concentration data during 2000-2013. Tropical evergreen forest, savanna and 59 deciduous forest areas are the largest sinks at 123 Tg CO yr¹. Soil CO gross 60 consumption is sensitive to air temperature and atmospheric CO concentration while 61 gross production is sensitive to soil organic carbon (SOC) stock and air temperature. By 62 assuming that the spatially-distributed atmospheric CO concentrations (~128 ppby) are 63 not changing over time, global mean CO net deposition velocity is estimated to be 0.16-64 0.19 mm s⁻¹ during the 20th century. Under the future climate scenarios, the CO 65 deposition velocity will increase at 0.0002-0.0013 mm s⁻¹ yr⁻¹ during 2014-2100, 66 67 reaching 0.20-0.30 mm s⁻¹ by the end of the 21st century, primarily due to increasing temperature. Areas near the equator, Eastern US, Europe and eastern Asia will be the 68 largest sinks due to optimum soil moisture and high temperature. The annual global soil 69 net flux of atmospheric CO is primarily controlled by air temperature, soil temperature, 70 SOC and atmospheric CO concentrations, while its monthly variation is mainly 71 determined by air temperature, precipitation, soil temperature and soil moisture. 72

73 1. Introduction

Carbon monoxide (CO) plays an important role in controlling the oxidizing capacity of the atmosphere by reacting with OH radicals (Logan et al., 1981; Crutzen, 1987; Khalil & Rasmussen, 1990; Prather et al., 1995; Prather & Ehhalt, 2001). CO in the atmosphere can directly and indirectly influence the fate of critical greenhouse gases such as methane (CH₄) and ozone (O₃) (Tan and Zhuang, 2012). Although CO itself absorbs only a limited amount of infrared radiation from the Earth, the cumulative

indirect radiative forcing of CO may be even larger than that of the third powerful 80 greenhouse gas, nitrous oxide (N₂O, Myhre et al., 2013). Current estimates of global 81 82 CO emissions from both anthropogenic and natural sources range from 1550 to 2900 Tg CO yr¹, which are mainly from anthropogenic and natural direct emissions and from 83 the oxidation of methane and other Volatile Organic Compounds (VOC) (Prather et al., 84 1995; Khalil et al., 1999; Bergamaschi et al., 2000; Prather & Ehhalt, 2001, Stein et al., 85 2014). Chemical consumption of CO by atmospheric OH and the biological consumption 86 of CO by soil microbes are two major sinks of the atmospheric CO (Conrad, 1988; Lu & 87 Khalil, 1993; Yonemura et al., 2000; Whalen & Reeburgh, 2001). 88

Soils are globally considered as a major sink for CO due to microbial activities 89 (Whalen and Reeburgh, 2001; King and Weber, 2007). A diverse group of soil microbes 90 91 including carboxydotrophs, methanotrophs and nitrifiers are capable of oxidizing CO (King and Weber, 2007). Annually, 10-25% of total earth surface CO emissions were 92 consumed by soils (Sanhueza et al., 1998; King, 1999a; Chan & Steudler, 2006). Potter 93 et al. (1996) reported the global soil consumption to be from -50 to -16 Tg CO yr⁻¹ 94 (negative values represent the uptake from the atmosphere to soil), by using a single-95 box model over the upper 5 cm of soils. All existing estimates have large uncertainties 96 and range from -640 to -16 Tg CO yr⁻¹ (Sanhueza et al., 1998; King, 1999; Bergamaschi 97 98 et al., 2000). Similarly, the estimates of CO dry deposition velocities also have large uncertainties and range from 0 to 4.0mm s⁻¹ (here positive values are amount of 99 deposition to soils, King, 1999a; Castellanos et al., 2011). Soils also produce CO mainly 100 via abiotic processes such as thermal- and photo-degradation of organic matter or plant 101 materials (Conrad and Seiler, 1985b; Tarr et al., 1995; Schade et al., 1999; Derendorp 102 et al., 2011; Lee et al., 2012; van Asperen et al., 2015; Fraser et al., 2015, Pihlatie et al., 103 2016), except for a few cases of anaerobic formation. Photo-degradation is identified as 104 radiation-dependent degradation due to absorbing radiation (King et al., 2012). 105 Thermal-degradation is identified as the temperature-dependent degradation of carbon 106 in the absence of radiation and possibly oxygen (Derendorp et al., 2011; Lee et al., 107 2012; van Asperen et al., 2015; Pihlatie et al., 2016). These major soil CO production 108 processes, together with soil CO consumption processes, have not been adequately 109 modeled in global soil CO budget estimates. 110

To date, most top-down atmospheric models applied a dry deposition scheme 111 112 based on the resistance model of Wesely (1989). Such schemes provided a wide range of dry deposition velocities (Stevenson et al., 2006). Only a few models (MOZART-4, 113 Emmons et al., 2010; CAM-chem, Lamarque et al., 2012) have extended their dry 114 115 deposition schemes with a parameterization for CO and H₂ uptake through oxidation by soil microbes following the work of Sanderson et al. (2003), which itself was based on 116 extensive measurements from Yonemura et al. (2000). Potter et al. (1996) developed a 117 bottom-up model to simulate CO consumption and production at the global scale. This 118 model is a single box model, only considers top 5cm depth of soil and does not have 119 explicit microbial factors, which might have underestimated CO consumption (Potter et 120 al., 1996; King, 1999a). Current bottom-up CO modeling approaches are mostly based 121 122 on a limited number of CO in situ observations or laboratory studies to quantify regional and global soil consumption (Potter et al., 1996; Sanhueza et al., 1998; Khalil et al., 123 1999; King, 1999a; Bergamaschi et al., 2000; Prather & Ehhalt, 2001). To our 124 knowledge, no detailed process-based model of soil-atmospheric exchange of CO has 125 been published in the recent 15 years. One reason is that there is an incomplete 126 understanding of biological processes of uptake (King & Weber, 2007; Vreman et al., 127 2011; He and He, 2014; Pihlatie et al., 2016). Another reason is that there is lack of 128 129 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 130 laboratory experiments (e.g. Conrad and Seiler, 1985a; Funk et al., 1994; Tarr et al., 131 1995; Zepp et al., 1997; Kuhlbusch et al., 1998; Moxley and Smith, 1998; Schade et al., 132 1999; King and Crosby, 2002; Varella et al., 2004; Lee et al., 2012; Bruhn et al., 2013; 133 van Asperen et al., 2015). The first study to report long-term and continuous field 134 measurements of CO flux over grasslands using a micrometeorological eddy covariance 135 (EC) method is Pihlatie et al. (2016). 136

To improve the quantification of the global soil CO budget for the period 2000-2013 and CO deposition velocity for the 20th and 21st centuries, this study 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. The atmospheric CO concentration data from MOPITT (Gille,
2013) were used to drive model simulations from 2000 to 2013. A set of century-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, the effects of multiple forcings on the global CO consumption and
production, including the changes of climate and atmospheric CO concentrations at the
global scale were evaluated with the model.

149

150 **2. Method**

151 2.1 Overview

We first developed a daily soil CO dynamics module (CODM) that considers: (1) 152 soil-atmosphere CO exchange and diffusion process between soil layers, (2) 153 consumption by soil microbial oxidation, (3) production by soil chemical oxidation, and 154 (4) the effects of temperature, soil moisture, soil CO substrate and surface atmospheric 155 CO concentration on these processes. Second, we used the observed soil temperature 156 and moisture to evaluate TEM hydrology module and soil thermal module in order to 157 estimate soil physical variables. Then we used the data from laboratory experiments 158 159 and CO flux measurements to parameterize the model using the Shuffled Complex Evolution (SCE-UA) method (Duan et al., 1993). Finally, the model was extrapolated to 160 the globe at a 0.5° by 0.5° resolution. We conducted three sets of model experiments to 161 investigate the impact of climate and atmospheric CO concentrations on soil CO 162 dynamics: 1) simulations for 2000-2013 with MOPITT satellite atmospheric CO 163 concentration data; 2)simulations for 1901-2100 with constant atmospheric CO 164 concentrations estimated from an empirical function and the historical climate data 165 (1901-2013) and three future climate scenarios (2014-2100); and 3) Eight sensitivity 166 simulations by changing a) constant CO surface concentrations \pm 30%, b) SOC \pm 530%, 167 c) precipitation ±20% and d) air temperature ± 3°C for each pixel, respectively, while 168 holding other forcing data as they were, during 1999-2000. 169

170

171 2.2 Carbon Monoxide Dynamics Module (CODM)

172 Embedded in TEM (Figure 1), CODM is mainly driven by: (1) soil organic carbon 173 availability based on a carbon and nitrogen dynamics module (CNDM) (Zhuang et al., 174 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, 175 1996; Zhuang et al, 2004). Net exchange of CO between the atmosphere and soil is 176 determined by the mass balance approach (net flux = total production – total oxidation – 177 total soil CO concentration change). According to previous studies, we separated active 178 soils (top 30cm) for CO consumption and production into 1 cm thick layers (King, 1999a, 179 1999b; Whalen & Reeburgh, 2001; Chan & Steudler, 2006). Between the soil layers, the 180 changes of CO concentrations were calculated as: 181

182
$$\frac{\partial(C(t,i))}{\partial t} = \frac{\partial}{\partial z} \left(D(t,i) \frac{\partial(C(t,i))}{\partial z} \right) + P(t,i) - O(t,i) \quad (1)$$

Where C(t, i) is the CO concentration in layer i and at time t, units are mg m⁻³. z is the 183 depth of the soil, units are m. D(t, i) is the diffusion coefficient for layer i, units are m² s⁻ 184 ¹. P(t,i) is the CO production rate and O(t,i) is the CO consumption rate. The units of 185 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. 186 187 (1996), which are the functions of soil temperature, soil texture and soil moisture. The upper boundary condition is specified as the atmospheric CO concentration, which is 188 estimated by an empirical function of latitude (Potter et al., 1996) or directly measured 189 by the MOPITT satellite during 2000-2013. The lower boundary condition is assumed to 190 191 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. 192

193

194

CO consumption was modeled in unsaturated soil pores as:

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

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

201
$$f_1(C(t,i)) = \frac{C(t,i)}{C(t,i) + k_{CO}}$$
(2.1)

202
$$f_2(T(t,i)) = Q_{10}^{\frac{T(t,i) - T_{ref}}{10}}$$
(2.2)

203
$$f_3(M(t,i)) = \frac{(M(t,i) - M_{min})(M(t,i) - M_{max})}{(M(t,i) - M_{min})(M(t,i) - M_{max}) - (M(t,i) - M_{opt})^2}$$
(2.3)

Where $f_1(C(t,i))$ is a multiplier that enhances oxidation rate with increasing soil CO 204 concentrations using a Michaelis-Menten function with a half-saturation constant k_{CO} , 205 and their values were previous estimated ranging from 5 to 51 µl CO I⁻¹ for different 206 ecosystems (Whalen & Reeburgh, 2001); $f_2(T(t,i))$ is a multiplier that enhances CO 207 oxidation rates with increasing soil temperature using a Q10 function with Q_{10} 208 coefficients (Whalen & Reeburgh, 2001). T_{ref} is the reference temperature, units are °C 209 (Zhuang et al., 2004, 2013). $f_3(M(t,i))$ is a multiplier to estimate the biological limiting 210 effect that diminishes CO oxidation rates if the soil moisture is not at an optimum level 211 212 (Mopt). Mmin, Mmax and Mopt are the minimum, maximum and optimum volumetric soil moistures of oxidation reaction, respectively. Equation (2.2) will overestimate CO 213 214 consumption at higher temperature because in reality CO consumption will decrease at higher temperatures than optimum temperature, while f_2 will keep increasing with rising 215 temperature. However, the CO consumption is constrained by CO production, and 216 equation (1) is used to represent this constraint. 217

We modeled the CO production rate (P(t, i)) as a process of chemical oxidation constrained by soil organic carbon (SOC) decay (Conrad and Seiler,1985; Potter et al. 1996; Jobbagy & Jackson, 2000; van Asperen et al., 2015):

221
$$P(t,i) = P_r(t,i) \cdot E_{SOC} \cdot C_{SOC}(t) \cdot F_{SOC}$$
(3)

Where $P_r(t, i)$ is a reference soil CO production rate which has been normalized to rate at reference temperature (production rate at temperature (t, i) divided by production rate at reference temperature), which is affected by soil moisture and soil temperature (Conrad and Seiler,1985; van Asperen et al., 2015). E_{SOC} is an estimated nominal CO production factor of $3.5 \pm 0.9 \times 10^{-9}$ mg CO m⁻² s⁻¹ per g SOC m⁻² (to 30 cm surface soil depth) (Potter et al., 1996). $C_{SOC}(t)$ is a SOC content in mg m⁻², which is provided by CNDM module in TEM. F_{SOC} is a constant fraction of top 30cm SOC compared to total amount of SOC, which is 0.33 for shrubland areas, 0.42 for grassland areas and 0.50 for forest areas, respectively (Jobbagy & Jackson, 2000). $P_r(t, i)$ was calculated as:

231
$$P_r(t,i) = \exp\left(f_4(M(t,i)) \cdot Ea_{ref}/R \cdot \left(\frac{1}{273.15 + PT_{ref}} - \frac{1}{T(t,i) + 273.15}\right)\right)$$
(3.1)

232
$$f_4(M(t,i)) = \frac{PM_{ref}}{M(t,i) + PM_{ref}}$$
(3.2)

233 Where equation (3.1) is derived from Arrhenius equation for chemical reactions and normalized using the reference temperature PT_{ref} . Ea_{ref}/R is the reference activation 234 energy divided by gas constant R, units are K. $f_4(M(t,i))$ is the multiplier that reduces 235 activation energy using a regression approach based on laboratory experiment of 236 moisture influences on CO production (Conrad and Seiler, 1985). PM_{ref} is the reference 237 volumetric soil moisture, ranging from 0.01 to 0.5 volume/volume (v/v). We assumed 238 thermal-degradation as the main CO producing process due to lack of photo-239 degradation data and hard to distinguish photo-degradation from observations. In order 240 to reduce the bias from thermal-degradation to total abiotic degradation, the equation 241 (3.1) is parameterized by comparing with total production rate. For instance, $P_r(t,i)$ 242 calculation can perfectly fit the experiment results in Van Asperen et al., 2015 with 243 proper $PT_{ref}(18^{\circ}C)$, $Ea_{ref}/R(14000 \text{ K})$ and $PM_{ref}(0.5 \text{ v/v})$. 244

245 CO deposition velocity was modeled in the same way as equation (19.1) in 246 Seinfeld, et al. (1998):

 v_d

247

$$= -F_{net}/C_{CO,air} \tag{4}$$

Where the v_d is the CO deposition velocity, units are mm s⁻¹; F_{net} is the model estimated CO net flux rate, units are mg CO m⁻² day⁻¹; $C_{CO,air}$ is the CO surface concentration, units are ppbv. $C_{CO,air}$ can be MOPITT CO surface concentration data or derived CO surface concentrations using the same method as Potter et al. (1996). Positive values of v_d are soil uptake (deposition from air to soils) and negative values are soil emission.

254

255 2.3 Model Parameterization and Extrapolation

The model parameterization was conducted in two steps: 1) Thermal and 256 257 hydrology modules embedded in TEM were revised, calibrated and evaluated by running model with corresponding local meteorological or climatic data at four 258 representative sites, including boreal forest, temperate forest, tropical forest and 259 260 savanna (Table 1, site No.1 to 4, Figure 2) to minimize model data mismatch in terms of soil temperature and moisture. 2) CODM module was parameterized by running TEM 261 for observational periods with the corresponding local meteorological or climatic data at 262 each reference site (Table 1, Figure 3), and using the Shuffled Complex Evolution 263 Approach in R language (SCE-UA-R) (Duan et al., 1993) to minimize the difference 264 between simulated and observed net CO flux. Eleven parameters including k_{CO}, V_{max}, 265 T_{ref} , Q_{10} , M_{min} , M_{max} , M_{opt} , E_{SOC} , Ea_{ref}/R , PM_{ref} and PT_{ref} were optimized (Table 2). To 266 be noticed, F_{SOC} was not involved in the calibration process. Parameter priors were 267 decided based on previous studies (Conrad & Seiler, 1985; King, 1999b; Whalen & 268 Reeburgh, 2001; Zhuang et al., 2004). SCE-UA-R was used for site No. 6, 8, 10, 11 269 270 (Table 1). Each site has 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 271 the loops. For wetlands, the only available data is from site No.12. We used a trial-and-272 error method to make our simulated results in the range of observed flux rates, with a 273 10% tolerance. For tropical sites, since tropical savanna vegetation type is a 274 combination type of tropical forest and grassland in our model, we first used Site No. 13 275 276 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 277 evaluate our model results for temperate forest and grassland. Besides the observed 278 climatic and soil property data, we used ERA-Interim reanalysis data from The 279 European Centre for Medium-Range Weather Forecasts (ECMWF) (Dee et al., 2011), 280 AmeriFlux observed meteorology data (http://ameriflux.lbl.gov/) and reanalysis climatic 281 data from Climatic Research Unit (CRU, Harris et al., 2013) to fill the missing 282 283 environmental data. To sum up, parameters for various ecosystem types in Table 2 were the final results of our parameterization. Model parameterization was conducted 284 for ecosystem types including boreal forest, temperate coniferous forest, temperate 285 deciduous forest, and grassland using SCE-UA-R. Tropical forest and wet tundra used 286

a trial-and-error method to adjust parameters letting simulation results best fit the lab
data. Due to limited data availability, we assumed temperate evergreen broadleaf forest
having the same parameters as temperate deciduous forest.

290

291 2.4 Data Organization

To get spatially and temporally explicit estimates of CO consumption, production and net flux at the global scale, we used the data of land cover, soils, climate and leaf area index (LAI) from various sources at a spatial resolution of 0.5° latitude X 0.5° longitude to drive TEM. The land cover data include potential vegetation distribution (Melillo et al., 1993) and soil texture (Zhuang et al., 2003), which were used to assign vegetation- and texture-specific parameters to each grid cell.

For the simulation of the period 1901-2013, monthly air temperature, precipitation, 298 clouds fraction and vapor pressure data sets from CRU were used to estimate the soil 299 temperature, soil moisture and SOC with TEM (Figure 4). Monthly LAI data from TEM 300 were required to simulate soil moisture (Zhuang et al., 2004). During this period time, 301 we used an empirical function of latitude, which was derived from the observed 302 303 latitudinal distribution of tropospheric carbon monoxide (Badr and Probert, 1994) to calculate static CO surface concentration distribution (equation (7), Potter et al., 1996): 304 $C_{co,air} = 82.267856 + 0.8441503L + 1.55934 \times 10^{-2}L^2 + 2.37 \times 10^{-5}L^3 - 10^{-5}L^3 -$ 305

306

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

Where $C_{co,air}$ is the derived surface CO concentration (ppbv), L represents 307 latitude which is negative degrees for southern hemisphere and positive degrees for 308 northern hemisphere. We also used the atmospheric CO data from MOPITT satellite 309 310 during 2000-2013 (Figure 5). We averaged day-time and night-time monthly mean retrieved CO surface level 3 data (variables mapped on 0.5° latitude X 0.5° longitude 311 grid scales with monthly time step, Gille, 2013) to represent the CO surface 312 concentration level in each month. The missing pixels were fixed by the average of 313 314 pixels which had values and were inside 1.5 times of the distance between this missing pixel and the nearest pixel with values. These global mean values shown in Figure 5 do 315 316 not include ocean surfaces, thus there are differences between our surface CO

concentration results and Yoon and Pozzer's report in 2014, which is as low as 99.8ppb. 317 318 From 2014 to 2100, we used Intergovernmental Panel on Climate Change (IPCC) future 319 climate scenarios from Representative Concentration Pathways (RCPs) climate forcing data sets RCP2.6, RCP4.5 and RCP8.5 (Figure 6). RCP2.6, 4.5 and 8.5 datasets are 320 future climate projections with anthropogenic greenhouse gas emission radiative forcing 321 of 2.6 W m⁻², 4.5 W m⁻² and 8.5 W m⁻², respectively, by 2100. Since RCPs did not have 322 water vapor pressure data, we used the specific humidity and sea level air pressure 323 from the RCPs and elevation of surface to estimate the monthly surface vapor pressure 324 (Seinfeld & Pandis, 1998). 325

326

327 2.5 Model Experiment Design

We conducted two sets of core simulations and eight sensitivity test simulations 328 329 for a historical period. The two core sets of simulations were driven with MOPITT CO surface concentrations data for the period 2000-2013 (experiment E1) and with spatially 330 distributed CO surface concentrations assuming as constant over time estimated from 331 an empirical function of latitude for the period 1901-2100 (experiment E2), respectively. 332 333 Specifically, in experiment E2 we used the CRU climate forcing for the historical period 1901-2013 and the climate data of RCP2.6, RCP4.5 and RCP8.5 for different future 334 scenarios to examine the responses of CO flux to changing climates. Eight sensitivity 335 simulations were driven with varying different forcing variables while keeping others as 336 they were: 1) with constant CO surface concentrations \pm 30%, 2) SOC \pm 530%, 3) 337 precipitation ±20% and 4) air temperature ± 3°C for each pixel, respectively, during 338 1999-2000 (E3). 339

340

341 3. Results

342 3.1 Site Evaluation

Both the magnitude and variation of the simulated soil temperature and moisture from cold areas to warm areas compared well to the observations (Figure. 2). The magnitude of the simulated CO flux is comparable and correlated with the observations (r is about 0.5, p-value < 0.001, Figures 3, a2, b2, c2, d2). Estimated CO fluxes for different ecosystem types range from -28.4 to 1.7 mg CO m⁻² day⁻¹, and the root mean square error (RMSE) between simulation and observation at all sites is below 1.5 mg CO m⁻² day⁻¹. RMSE for site No. 7 is bigger than 2.0 mg CO m⁻² day⁻¹ when compared with transparent chamber observations. For boreal forest site, we only had 8 acceptable points in 1994 and 1996 (Figure 3c2).

352

353 3.2 Global Soil CO Dynamics During 2000-2013

Using the MOPITT CO surface concentration data during 2000-2013 (E1), the 354 estimated mean soil CO consumption, production and net flux (positive values indicate 355 CO emissions from soils to the atmosphere) are from -197 to -180, 34 to 36 and -163 to 356 357 -145 Tg CO yr¹, respectively (Figure 7a). Consumption is about 4 times larger than production. The annual consumption and net flux trends follow the atmospheric CO 358 concentration trends (Figure 5b, Figure 7a), with a small interannual variability (<10%). 359 The latitudinal distributions of consumption, production and net fluxes share the same 360 spatial pattern. Around 20°S-20°N and 20-60N° are the largest and second largest 361 areas for production and consumption, while the 45°S-45°N area accounts for nearly 90% 362 363 of the total consumption and production (Figure 7b, Table 3). The Southern and Northern Hemispheres have 41% and 59% of the total consumption, and 47% and 53% 364 of the total production, respectively (Table 3). The highest rates of consumption and 365 production are located in areas close to the equator, and consumption from areas such 366 as eastern US, Europe and eastern Asia also is high (>-1000 mg m⁻² yr⁻¹) (Figure 8a, b). 367 Global soils serve as an atmospheric CO sink (Figure 8c). Some areas, such as 368 western US and southern Australia, are CO sources, all of which are grasslands or 369 experiencing dry climate. Tropical evergreen forests are the largest sinks, consuming 86 370 Tg CO yr⁻¹, and tropical savanna and deciduous forest are second and third largest 371 sinks, consuming a total of 37 Tg CO yr⁻¹ (Table 4). These three ecosystems account 372 for 66% of the total consumption. Tropical evergreen forests are also the largest source 373 374 of soil CO production, producing 16 Tg CO yr¹, while tropical savanna has a considerable production of 6 Tg CO yr¹ (Table 4). Moreover, tropical areas, including 375 forested wetlands, forested floodplain and evergreen forests, are most efficient for CO 376 consumption, ranging from -18 to -13 mg CO m⁻² day⁻¹. They are also the most efficient 377

for CO production at over 2 mg CO m⁻² day⁻¹ (Table 4, calculated by fluxes divided by area).

380

381 3.3 Global Soil CO Dynamics During 1901-2100

Using the constant CO surface concentration, the estimated global mean CO 382 deposition velocities are 0.16-0.19 mm s⁻¹ for the period 1901-2013. For the period 383 2014-2100, deposition velocities are 0.18-0.21, 0.18-0.24 and 0.17-0.31 for RCP2.6, 4.5 384 and 8.5 scenarios, respectively (Figure 9). During 2014-2100, there are significant 385 trends of increasing deposition velocities for nearly all scenarios (Figure 9). The rates of 386 increasing are 0.0002, 0.0005 and 0.0013 mm s⁻¹ yr⁻¹, and will reach 0.20, 0.23 and 387 0.30 mm s⁻¹ by the end of the 21st century for the RCP2.6, RCP4.5 and RCP8.5 388 scenarios, respectively (Figure 9). These increasing trends are similar to air 389 temperature increasing trends (Figure 6a). Global distribution patterns of CO deposition 390 velocity are similar to net flux distribution for the period 2000-2013 but there are 391 significant differences among 1901-2013, RCP2.6, RCP4.5 and RCP8.5 scenarios 392 (Figure 10). Deposition velocities are increasing from RCP2.6 to RCP8.5 and larger 393 394 than in the historical periods in areas near the equator (Figure 10). Areas near the equator and eastern Asia become big sinks of atmospheric CO, while northeastern US 395 becomes a small source in the 21st century (Figure 10). Different vegetation types have 396 a large range of deposition velocity, from 0.008 to 1.154 mm s⁻¹ (Table 4). The tropical 397 forested wetland, tropical forested floodplain and tropical evergreen forest have top 398 three largest deposition velocity of 1.154, 1.117 and 0.879 mm s⁻¹, respectively, while 399 desert, short grasslands, and wet tundra have the smallest deposition velocity 0.008, 400 0.010 and 0.015 mm s⁻¹, respectively. 401

402

403 3.4 Sensitivity test

Eight sensitivity tests have been conducted for the 1999-2000 period, including changing atmospheric CO by $\pm 30\%$, SOC by $\pm 530\%$, precipitation by $\pm 30\%$ and air temperature by $\pm 3^{\circ}$ 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 (changing up to 36%) and SOC (530%). 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, a 30%
change in precipitation will not lead to large changes in CO flux (< 3%).

412

413 4. Discussion

414 4.1 Comparison with Other Studies

415 Previous studies estimated a large range of global CO consumption from -16 to -640 Tg CO yr⁻¹. Our estimates are from -197 to -180 Tg CO yr⁻¹ for for 2000-2013 using 416 MOPITT satellite CO surface concentration data. Previous studies also provided a large 417 range for CO production from 0 to 7.6 mg m⁻² day⁻¹ (reviewed in Pihlatie et al., 2016). 418 Our results showed averaged CO production ranging from 0.01 to 2.29 mg m⁻² day⁻¹. 419 Previously reported CO deposition velocities for different vegetation types range from 420 421 0.0 to 4.0 mm s⁻¹ while our results showed an averaged CO deposition velocity ranging from 0.006 to 1.154 mm s⁻¹ for different vegetation types. The large uncertainty of these 422 estimates is mainly due to a different consideration of the microbial activities, the depth 423 of the soil, and the parameters in the model. In contrast to the estimates of -57 to -16 Tg 424 CO yr⁻¹ which were based on top 5 cm soils (Potter et al., 1996), our estimates 425 426 considered 30cm soils, just as used in Whalen & Reeburgh (2001). In addition, we used a thinner layer division (1cm each layer) for diffusion process, and used the Crank-427 Nicolson method to solve partial differential equations to avoid time step influences. We 428 also included the microbial CO oxidation process to remove the CO from soils and the 429 effects of soil moisture, soil temperature, vegetation type and soil CO substrate on 430 microbial activities. Our soil thermal, soil hydrology and carbon and nitrogen dynamics 431 simulated in TEM provided carbon substrate spatially and temporally for estimating soil 432 CO dynamics. Overall, although a few previous studies have examined the long-term 433 impacts of climate, land use and nitrogen depositions on CO dynamics (Chan & 434

Steudler, 2006, Pihlatie et al., 2016), the global prediction of soil CO dynamics still hasa large uncertainty.

437

438 4.2 Major Controls to Soil CO Dynamics

Sensitivity tests indicate that consumption is normally much larger than CO 439 production so that the former will determine the dynamics of the net flux (Table 5). 440 Model being sensitive to air temperature explains the small increasing trends after the 441 1960s, the significant increasing trend in the 21st century and the large sinks over 442 tropical areas (Table 5, Figure 9). SOC did not directly influence CO consumption. For 443 instance, increasing SOC led to an increase in soil CO substrate, implying that more CO 444 in soils can be consumed. To be noticed, an extra 348 Tg CO yr⁻¹ was taken up from 445 the atmosphere to soils in sensitivity test when SOC increasing by 530% (Table 5), 446 which will be discussed in detail in Section 4.3. CO surface concentrations will only 447 influence the uptake rate and soil CO substrate concentrations, thus influencing the soil 448 CO consumption rate. 449

Annual CO consumption and net flux have a similar correlation coefficient with 450 451 forcing variables and both are significantly correlated with air temperature, soil temperature SOC and atmospheric CO concentration (R > 0.91 globally, Table 6). 452 Increasing temperature will increase microbial activities, while more SOC will increase 453 soil CO substrate level. Annual CO consumption and net flux have low correlations with 454 annual precipitation and soil moisture, especially at 45°N-45°S (R<0.54 Table 6). 455 Annual CO production is strongly correlated with annual mean SOC, air temperature 456 and soil temperature (R>0.91), while is less correlated with precipitation, soil moisture 457 and atmospheric CO concentration. Meanwhile, the monthly CO consumption, 458 production and net flux are well correlated with air temperature, soil temperature, 459 precipitation, and soil moisture (R>0.69 globally Table 6). The soil moisture is 460 significantly influenced by temperature at a monthly time step since increasing 461 temperature would induce higher evapotranspiration. Monthly CO consumption, 462 production and net flux have low correlations with SOC because it will not change 463 greatly within a month. 464

The R between annual soil CO consumption and atmospheric CO concentration is 0.91 at the global scale because the atmospheric CO concentration, air temperature, and soil temperature dominate the annual consumption rate. At monthly scale, this R is -0.48 because global atmospheric CO concentrations are high in winter and low in summer while the simulated soil CO consumption shows an opposite monthly variation (Table 6, Figure 11), suggesting that other factors such as precipitation, air temperature, and soil temperature are major controls for monthly CO fluxes.

472

473 **4.3. Model Uncertainties and Limitations**

There are a number of limitations, contributing to our simulation uncertainties. 474 475 First, due to lacking long-period observational data of CO flux and associated environmental factors, the model parameterization can only be conducted for 4 476 ecosystem types including boreal forest, temperate coniferous forest, temperate 477 deciduous forest and grassland. Tropical forest calibration is only conducted using a 478 very limited amount of lab experiment data, but tropical areas are hotspots for CO soil-479 atmosphere exchanges. Besides, tropical forest SOC for top 30cm can be really high 480 481 according to observations. TEM model may underestimate the top 30cm SOC, which will underestimate production rates, especially in tropical regions. Tropical regions 482 typically have high temperature during the whole year, which may result in 483 overestimation of CO consumption using equation (2.2). The large deviation for tropical 484 savanna (which is mosaic of tropical forest and grassland ecosystems) may be due to 485 486 using outside air temperature to represent inside air temperature of transparent chamber observations (Varella et al., 2004), and uncertain tropical forest 487 parameterization. Second, we used the conclusion from van Asperen et al. (2015) and 488 only considered the thermal-degradation process for CO production in this study. Photo-489 degradation process and biological formation process were not considered due to 490 lacking understanding of these processes. Third, the static CO surface concentration 491 derived from the empirical function is lower than MOPITT CO surface concentration, 492 which will lead to underestimation of CO deposition velocity during 1901-2100. Fourth, 493 from sensitivity test (Table 5) we notice that SOC increasing (5%) resulted in a net flux 494 increase (2.57%). The SOC increase enhanced CO production (Equation 3), CO 495

496 concentrations (Equation 1), and CO oxidation (Equation 2). When the change of total 497 oxidation is larger than the difference between the change of total production and the 498 change of total soil CO concentration (Equation 1), the estimate of the net flux change is negative (from atmosphere to soil) using a mass balance approach (Section 2.2), 499 leading to a 2.57% increase in net flux in our SOC sensitive test. This is due to the fact 500 that CO production (Equation 3) is calculated independently from oxidation calculation 501 (Equation 2). This will not influence our other results since SOC varies slightly during 502 503 our simulation periods with only a 3% increase from 1900 to 2013 (Figure 4d) and up to 504 a 4% increase from 2014 to 2100 (Figure 6g). This artifact problem in SOC sensitivity test can be alleviated using a very fine time step (e.g., 1 second), because CO 505 concentrations will change slightly within the short time, allowing net flux roughly equals 506 507 the difference between production and oxidation within the short time step. If the change of production is bigger than the change of oxidation, the change of net flux will be 508 509 positive, leading to a decrease of deposit to soil. The downside is that running the model at one second time step will require significantly high computing time. Fourth, 510 from sensitivity test (Table 5) and model test (Table 7), we notice that the diffusion and 511 consumption in the model is very sensitive to sudden 30% SOC changes with 5-minute 512 513 time step. In reality, diffusion and consumption shall only be slightly influenced by 514 indirect changes of soil CO concentration due to SOC changes. When we used 3minute or 1-minute time step, the model responses to SOC changes are reasonable 515 (Table 7). However, we believe 5-minute step is suitable in this study since SOC varies 516 slightly during the whole global simulation period with only 3% increasing from 1900 to 517 2013 (Figure 4d) and up to a 4% increase from 2014 to 2100 (Figure 6g). Our model 518 519 test showed there are small responses to these small amounts of SOC increasing (Table 7). Fifth, our model structure still has a large potential to improve. In this study 520 we divided the top 30cm soil into 30 layers (layer thickness dz=1cm), but finer division 521 will increase the accuracy (Figure 12). We chose dz=1cm because if dz>1cm, the model 522 vertical CO concentration profile will deviate from reality and diffusion process will be 523 influenced significantly. If dz<1cm, it will need much more computing time but don't 524 525 have much improvement compared to dz=1cm (Figure 12a-e). We notice that the 30layer division well represents soil CO concentration profile not only for the days with soil 526

527 CO net uptake, but also for the days with CO net emission (Figure 12c, f). Sixth, Michaelis-Menten function (equation 2.1) is used in this model and we notice that k_{CO} is 528 normally much larger than C(t, i) in those days of net soil uptake (over ten times larger, 529 Figure 12). However, we can't simplify equation (2.2) to $f_1(\mathcal{C}(t,i)) = \frac{\mathcal{C}(t,i)}{k_{CO}}$ since CO 530 concentrations in soils can be larger than in the atmosphere in the days of net 531 emissions and C(t, i) may be close to k_{CO} , which may lead to overestimation of CO 532 oxidation (Figure 12f). Finally, although we focused on natural ecosystems in this study, 533 land-use change, agriculture activity, and nitrogen deposition also affect the soil CO 534 consumption and production (King, 2002; Chan & Steudler, 2006). For instance, soil CO 535 consumption in agriculture ecosystems is 0 to 9 mg CO m⁻² day⁻¹ in Brazil (King & 536 Hungria, 2002). We used grass land or forest ecosystem to represent agriculture areas 537 in CODM module. Our future study shall include these processes and factors. 538

539

540 5. Conclusions

We analyzed the magnitude, spatial pattern, and the controlling factors of the 541 542 atmosphere-soil CO exchanges at the global scale for the 20th and 21st centuries using a process-based biogeochemistry model. Major processes include atmospheric CO 543 diffusion into soils, microbial oxidation removal of CO, and CO production through 544 chemical reaction. We found that air temperature and soil temperature play a dominant 545 role in determining annual soil CO consumption and production while precipitation, air 546 547 temperature, and soil temperature are the major controls for the monthly consumption and production. Atmospheric CO concentrations are important for annual CO 548 consumption. We estimated that the global annual CO consumption, production and net 549 fluxes for 2000-2013 are from -197 to -180, 34 to 36 and -163 to -145 Tg CO yr¹, 550 respectively, when using a MOPITT CO surface concentration data. Tropical evergreen 551 552 forest, savanna and deciduous forest areas are the largest sinks accounting for 66% of the total CO consumption, while the Northern Hemisphere consumes 59% of the global 553 total. During the 20th century, the estimated CO deposition velocity is 0.16-0.19 mm s⁻¹. 554 The predicted CO deposition velocity will reach 0.20-0.30 mm s⁻¹ in the 2090s, primarily 555 because of increasing air temperature. The areas near the equator, eastern Asia, 556

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 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.

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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)

819 **Table 2.** Ecosystem-specific parameters in the CODM module^a

	Ecosystem Type	k _{CO} (ul CO l ⁻¹)	$Vmax (ug CO g^{-1}h^{-1})$	<i>Т_{ref}</i> (°С)	Q10 (Unitless)	M_{min} $(\frac{v}{v})$	M_{max} $(\frac{v}{v})$	$\begin{array}{c} M_{opt} \\ (\frac{v}{v}) \end{array}$	E _{SOC}	F_{SOC} $(rac{g}{g})$	Ea _{ref} R (K)	$\frac{PM_{ref}}{(\frac{v}{v})}$	<i>РТ_{ref}</i> (°С)
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_{C0} 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;

	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 3. Regional soil CO consumption, net flux and production (Tg CO yr⁻¹) during 2000-2013

Table 4. Annual total soil CO consumption, net flux and production in different ecosystems during 2000-2013

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

Vegetation Type	Area (10 ⁶ km²)	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	

Table 5. Sensitivity of global CO consumption, net flux and production (Tg CO yr⁻¹) to changes in atmospheric

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

	Baselin	CO	CO	SOC	SOC	Prec	Prec	AT +3°C	AT -3°C
	е	+30%	-30%	+ <u>5</u> 30%	- <u>5</u> 30%	+30%	-30%		
Consumption	-147.65	-164.14	-131.12	<u>-152.27</u> -	-	-	-	-190.59	-114.83
				175.37	<u>143.03119.9</u>	150.72	143.50		
					θ				
Change (%)	0.00	11.17	-11.19	<u>3.13</u> 18.78	- <u>3.13</u> 18.79	2.08	-2.81	29.09	-22.23
Net flux	-113.65	-130.15	-97.12	<u>-116.58</u> -	- <u>110.73</u> 96.10	-	-	-144.23	-89.58
				131.18		116.97	109.32		
Change (%)	0.00	14.51	-14.54	<u>2.57</u> 15.42	- <u>2.57</u> 15.44	2.92	-3.81	26.90	-21.18
Production	33.99	33.99	33.99	<u>35.69</u> 44.1	<u>32.29</u> 23.80	33.74	34.17	46.36	25.25
				9					
Change (%)	0.00	0.00	0.00	<u>5.00</u> 30.00	- <u>5.00</u> 30.00	-0.75	0.53	36.39	-25.72

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Table 6. Correlation coefficients between forcing variables (precipitation (Prec), air temperature (Tair), soil organic carbon (SOC), soil temperature (Tsoil), soil moisture (Msoil) and atmospheric CO (CO air)) and absolute values of consumption, production and net flux for different regions and the globe

				Month	ıly				Annua	al	
		North-	45°N-	0°-	45°S-	Global	North-	45°N-	0°-	45°S-	Global
		45°N	0°	45°S	South		45°N	0°	45°S	South	
	Consumption	0.91	0.96	0.92	-0.34	0.87	0.65	0.21	0.26	0.13	0.52
Prec	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
	Consumption	0.97	0.98	0.91	0.96	0.95	0.92	0.93	0.88	0.84	0.91
Tair	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
	Consumption	-0.19	0.07	0.21	-0.01	0.15	0.68	0.90	0.92	0.47	0.92
SOC	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
	Consumption	0.97	0.98	0.92	0.96	0.95	0.94	0.93	0.88	0.85	0.95
Tsoil	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
	Consumption	0.85	0.96	0.92	0.19	0.76	0.03	0.22	0.14	0.26	0.22
Msoil	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
	Consumption	-0.66	-0.76	-0.29	0.14	-0.48	0.87	0.88	0.81	0.98	0.91
CO Air	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

85B

87 Table 7. Model test for site No.8 during 2002-2003. Time step is for solving equation (1). SOC increasing

871 represents the percentage of SOC increased in each test. Baseline is the simulation using original time step

872 and SOC input. Differences represent new simulation results minus baseline results.

Time Step	SOC Increasing	Units: mg m⁻² yr⁻¹	Consumption	Production	Diffusion
5min	0%	Baseline	-1611.5	410.0	-1201.5
5min	30%	Differences	-293.0	123.0	-170.0
3min	30%	Differences	-156.7	123.0	-33.7
1min	30%	Differences	-97.4	123.0	25.6
5min	10%	Differences	-97.7	41.0	-56.7
5min	1%	Differences	-9.8	4.1	-5.7



874

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 place.

- 880
- 881
- 882
- 883
- 884



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) probe output period using an empirical calibration function of Bourgeau-Chavez et al. (2012) for 5cm-30cm layer. Some of them resulted in calculations of values greater than 100% VSM in Nakai et al. (2013) study. Our 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)



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).

Figure 3. Contd. Parameter ensemble experiment results: Each parameter has 50 calibrated values generated from 991 running SCE-UA-R 50 times independently. Parameters are normalized to their largest potential values described in Table 992 2. (c1) and (c2) are boreal forest normalized parameter distribution boxplots and CO flux comparisons between model 993 994 simulations (solid line, using mean value of parameters) and observations (green diamond, red lines represent error bar, 995 site No. 12), respectively. For each box, line top, box top, horizontal line inside box, box bottom and line bottom represent 996 maximum, third quartile, median, first quartile and minimum of 50 parameter values. Red dot represents the mean value 997 of 50 parameter values. (d1) and (d2) are for grassland (site No.6). Grassland observation data is the sum of hourly observations so error bar represented the standard deviation. 998

.000

Figure 4. Historical global land surface (excluding Antarctic area and ocean area) mean climate, and simulated global mean soil moisture, soil temperature and SOC for the period 1901-2013.

.003

.004

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 Antarctic area and ocean area) mean climate from RCP2.6, RCP4.5 and RCP8.5 data sets and simulated 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.

Figure 7. Global mean soil CO consumption, production and net flux: (a) annual time series during 2000-2013and (b)
 latitudinal distribution during 2000-2013.

.091 .092

Figure 8. Global annual mean soil CO fluxes (mg CO m⁻² yr⁻¹) during 2000-2013 using MOPITT CO atmospheric surface concentration data (right side)

.093

Figure 9. Global mean annual time series of CO deposition velocity (mm s⁻¹) using constant in time, spatially distributed CO concentration data 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)

.097

Figure 10. Global annual mean CO deposition velocity using constant in time, spatially distributed CO .098 concentration data (mm s⁻¹) a) during 1901-2013 and b), c), d) under future climate scenarios RCP2.6, RCP4.5 and .099 RCP8.5 during 2014-2100, respectively,

Figure 11. Global mean monthly time series of MOPITT surface atmospheric CO concentration (ppbv) and soil CO consumption from model simulations E1 (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.