

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

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

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

Besides these, the authors should be careful with claiming that they quantify the soil budget for the 20th and 21st centuries.

This issue should be discussed thoroughly in Sect. 4.3.

Response: We highly appreciate your constructive suggestions and comments. We have chosen your second recommendation and changed historical simulation (1901-2013) and future simulation (2014-2100) results to be presented as deposition velocity by using the method in Seinfeld, et al. (1998). In this revision, what we have done include: 1) We only discussed absolute values of simulations during the period 2000-2013 when using MOPITT CO surface concentration data; 2) We removed all parts that involve absolute values of CO consumption, production and net fluxes; 3) We have plotted out new figures to present deposition velocity for 1901-2100; 4) We have re-organized the paper with a clearer experiment orders: E1: simulations during the period 2000-2013 using MOPITT CO surface concentration data; E2: simulations during the period 1901-2100 using constant in time, spatially distributed CO surface concentration data; E3: sensitivity tests; and 5) We added discussion of CO concentration influences on CO dynamics in Section 4.3, line 450 to line 452.

Other general comments:

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

Response: We have used ranges now throughout the paper.

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

Response: We have reorganized a number of paragraphs to get them smaller. In addition, we have also reduced many repeated references in Introduction.

4) - I find the Introduction up to line 60 difficult to read, because of the long list of references following each bit of information. Please consider rephrasing and reducing the references that keep being repeated.

Response: We have significantly revised the Introduction section and also removed repeated or non-necessary references. Please see line 33 to line 52.

5) - The summary of the experiments is given both in 2.1 and 2.5, but the two descriptions do not seem to match. In 2.1. it is mentioned that the purpose was to “investigate the impact of ...atmospheric CO concentrations “but the sensitivity tests that are meant for this are not mentioned here.

Response: We have corrected this and now they describe the same experiments. See Section 2.1 and Section 2.5.

6) - is Michaelis-Menten kinetics necessary? The net uptake means in principle that at least the uppermost layer of soil has lower concentration than the atmosphere, and this is much lower than k_{CO} . Are the CO concentrations in the lower soil layer much larger, and if yes, why? Related to this, please show the CO soil profiles for some typical situations.

Response: In this revision, we have re-considered the Michaelis-Menten kinetics and presented soil CO concentration profile in figure 12. We found that the Michaelis-Menten kinetics can't be replaced in our model since the soil CO concentration can be bigger than atmospheric CO surface concentration in the days of net emissions (Figure 12f). Replacing it with a simple version will overestimate the oxidation rate when soil CO concentration is comparable with atmospheric CO concentration. We have discussed this in Section 4.3. From line 460 to line 465

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

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

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

Response: We have moved sensitivity test results to Section 3.4 and we have reorganized 4.2 based on specific comments. See section 3.4 from line 361 to line 369, and section 4.2 from line 397 to line 408.

9) - Section 4.3 discussed the model uncertainties and limitations, but ignores two of the major issues: (1) the CO concentrations mismatch between satellite and constant function, and (2) the overestimation of soil consumption at high temperatures.

Response: We have added these two points in Section 4.3 in this revision. See line 450 to 452 and line 441 to line 443 for issue 1) and issue 2), respectively.

Specific comments

10) - line 16: “constant spatially distributed” – can be understood wrongly as constant in space, which is not; I suggest to change to “constant in time, spatially distributed”

Response: We have rephrased it following your suggestion. “By assuming that the spatially-distributed atmospheric CO concentrations (~128 ppbv) are not changing over time”. From line 21 to line 23.

11) - line 20: “the largest sinks at 93 Tg CO yr⁻¹” – is this from the 20th century or 2000-2013 simulation? Please specify. Same for the next phrase.

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

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

Response: Corrected. Now units for deposition velocity are mm s⁻¹ throughout the paper manuscript.

13) - lines 61-68: the phrase is unclear, consider breaking it into smaller pieces. Also, quite some information is given on the studies listed, but not enough to actually understand what they did and what the differences are. Please consider either giving more details, or shortening this part.

Response: We have shortened this part to increase readability and avoid misleading. See line 53 to line 57.

14) - lines 62 – 66: the text seems contradictory: the uptake flux when using a constant deposition velocity globally (115-230 Tg) is smaller than when using the same deposition velocity in general and some area set to zero (300 Tg) - please explain or reformulate. “With different approaches” is too unspecific and does not really say anything.

Response: We have removed this for shortening the paragraph and reducing confusing. See line 57 to line 59.

15) - line 67: “ using empirical approaches with higher probability for lower values” – unclear, higher probability of what? lower values for what?

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

16) - lines 68 – 70: which other substances? what are other deposition velocities? please give examples if you mention this.

Response: We have rewritten this sentence and now just presented the ranges for all vegetation types. Detailed deposition velocity for different vegetation types can be found in King (1999a) and Castellanos et al. (2011). See line 57 to line 59.

17) - lines 75 – 91: a lot of this discussion on the thermal and photo degradation is irrelevant for this paper, especially the part on photo degradation which is not included in the model. Please reduce the irrelevant parts.

Response: We have reduced the part related to photo-degradation. Now we only mentioned what is photo-degradation. See line 63 to line 64.

18) - lines 92 – 93: it is unclear to me what this phrase means. I think what the authors may intend to say is that little attention has been paid to CO (including soil consumption and production) in global (chemistry?) models. Please reformulate if true.

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

19) - lines 97-98: suggest replacing “oxidation from soil bacteria and microbes” by “oxidation by soil microbes”. Bacteria are microbes.

Response: Corrected. See line 74 to line 75.

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

Response: We have removed the “CO emission” from the sentence. “One reason is that there is an incomplete understanding of biological processes of uptake”. See line 85 to line 86.

21) - line 156: “determined by the mass balance” – unclear what this means: which mass balance, of what, between what? please clarify.

Response: We have added the detailed description: “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).”

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

Response: Removed “due to oxidation” from the sentence. See line 144.

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

Response: We have rewritten the sentence and removed “modeled as an anaerobic process” from description: “CO consumption is modeled in unsaturated soil pores”, see line 153

24) - lines 186- 188: phrase unclear - I think the authors mean that Eq 2.2 will overestimate CO consumption because in reality CO consumption decreases at high temperatures, while in Eq 2.2 CO keeps increasing with temperature. Please reformulate.

Response: We have reformulated the sentence. “Equation (2.2) will overestimate CO 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 temperature.” See line 171 to 174.

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

Response: We have added the detailed description: “production rate at temperature $T(t, i)$ divided by production rate at reference temperature”. $P_r(t, i)$ is also described in equation (3.1) and the paragraph right below. See line 181 to line 182 and line 191 to line 192.

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

Response: Corrected to “30 cm”. See line 186.

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

Response: We have added details of meaning of RCP2.6, 4.5 and 8.5. “RCP2.6, 4.5 and 8.5 datasets are future climate projections with anthropogenic greenhouse gas emission radiative forcing of 2.6 W m^{-2} , 4.5 W m^{-2} and 8.5 W m^{-2} , respectively, by 2100.” See line 278 to line 280.

28) line 297: is the reported correlation really r , or is it r^2 ? Also, a correlation coefficient r of 0.5 is usually not considered high correlation.

Response: Correlation coefficient is reported in R. We have rewritten the sentence to give proper description now. See section 3.1, line 303 to line 304.

29) - lines 299 - 300: please compare the RMSEs reported to the CO fluxes, in order to give an impression on the relative errors.

Response: We have presented CO net flux rates in order to compare with RMSEs. Please find in Section 3.1, line 305.

30) - lines 318 – 319: “consume 42% and 58% of the total consumption, and produce 41% and 59% of total production” – please reformulate

Response: We have changed Section 3.2 only for the period 2000-2013. We have also fixed this by reformulating. “The Southern and Northern Hemispheres have 41% and 59% of the total consumption, and 47% and 53% of the total production, respectively (Table 3)”. See line 321 to 323.

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

Response: We have changed section 3.2 only for the period 2000-2013. The table has been updated to only have CO flux values for the period 2000-2013 with MOPITT CO surface concentration data and CO deposition velocity for the period 1901-2013 with constant in time, spatially distributed CO surface concentration. We have added the description on how to get these numbers. “calculated by flux values divided by area”. See section 3.2, line 336 to line 337

32) - lines 329 – 343: I find the text in this paragraph somewhat misleading. The fluxes are presented as changing or increasing relative to the simulation for 20th century, which suggests a temporal evolution, but in fact they are different mainly because of a different model setup, i.e different atmospheric CO concentrations. Consider using “different” and “larger” instead of “increasing”.

Response: We have removed this part and only talked about simulation results during 2000-2013 in section 3.2. We moved 1901-2013 deposition velocity simulation results to Section 3.3.

33) - line 359: “the rate ranges of increasing of consumption...” – I think it should be something like “the ranges of the rates of increase in consumption...”; please explain what these ranges are, are they corresponding to the three scenarios?

Response: We have changed the results in Section 3.3 to present deposition velocity and added the results of 1901-2013. We have rewritten this sentence and added more information to explain what the ranges are. See section 3.3, line 341 to line 343.

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

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

35) - Sect 4.2: The information here belongs mostly to Results, please reorganize. Also, the section is hard to follow – there are many correlations mentioned without much coherence. Consider reformulating into a more focused way, with one paragraph per idea (e.g on annual time scales, the CO uptake is mostly correlated to X, Y, Z... and then comment more if needed on X). Try to separate the annual and monthly results.

Response: We have 1) moved the sensitivity results to new Section 3.4; 2) reorganized the rest part by totally separating the monthly and annual correlation test results. See Section 3.4, line 362 to line 369 and Section 4.2, line 409 to line 430.

36) - lines 398 – 400 and Table 5: The effect of the SOC on the gross uptake flux seems too large. In my understanding, the text tries to explain that SOC increases the gross production which makes more CO available, which in turn leads to an increase in the gross CO consumption. But, for an increase in SOC of 30%, the production increases by about 10Tg/year, and the consumption increases by 28 Tg/year! How can that be? Where are the extra 18 Tg/year coming from?

Response: We have added extra comments on this problem in discussion section 4.2. “To be noticed, the CO oxidation increasing due to CO substrate change is larger than production increasing due to SOC increasing, leading to an extra 18 Tg CO yr⁻¹ being taken up from the atmosphere to soils in sensitivity test when SOC increasing by 30% (Table 5).” See line 403 to 406.

37) line 406: “as CO flux” – do you mean “as does the CO flux”?

Response: This sentence has been removed now.

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

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

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

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

See section 4.3, line 434 to line 437.

40) - line 427: “with RMSE ... day⁻¹” – I think this info has no meaning here

Response: We have removed “with RMSE ... day⁻¹” now. See section 4.3, line 437.

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

Response: They are correlation coefficient (R) between forcing variables and model results. We have rewritten the description of Table 6 for better understanding. See table 6.

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

Response: We have changed to “take place”. See figure 1 caption, line 810.

43) - Fig. 2: is this really volumetric soil moisture? The values do not seem realistic; I think typical values for water holding capacity for most soils are around 50 % and that would give the saturation. Please check the units.

Response: Thank you for pointing this out. In this revision, we traced back to Nakai et al. (2013) and found that our units and values are the same as they presented. The reason why the values were so high is that the volumetric soil moisture (VSM) was 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. Although Bourgeau-Chavez et al. (2012) provided calibration functions for each soil horizon (i.e., dead moss, upper duff, lower duff, and mineral soil), some of them resulted in values greater than 100% VSM in Nakai et al. (2013) study. The 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. We added the discussion on Figure 2 caption in this revision. From line 482 to 485.

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

Response: We have corrected units for figure 2d2.

Text comments:

45) - line 60: should be "... consumption to be ..."

Response: Corrected. See line 53.

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

Response: Corrected. See line 63.

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

Response: Corrected. See line 95.

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

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

49) - line 266 and through the paper: "transient" is used wrongly. If what is meant is "variable" then please do use "variable". Transient does not mean variable, but something that disappears.

Response: We want to use "transient" with meaning of data having time variation. But we have removed all of them now in manuscript to avoid misleading.

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

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

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

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

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

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

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

53) - Fig. 8: typo in legend: "production"

- Fig. 9: typo in legend: "production"

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