

Answer to reviewer number 1

Thank you for the time and effort spent into this review. Most of your comments have been directly addressed. They allowed us to improve the clarity of the paper, especially the overall structure and to provide a more intuitive understanding of the study.

Specific comments:

- 1) First and foremost, I would say that the paper is not very well organized. Due to its very descriptive nature, one loses the purpose of the paper. I would, hence suggest moving some of the techniques used in the paper to the appendix. Towards the end of section 2.3, the authors can add the first paragraph of section 2.4.1 accordingly, making necessary changes. Section 2.4.3 can be added as the last paragraph of section 2.3.

We agree with the reviewer that a reader would easily lose the purpose of the paper during the long description of the method. Following some of your suggestions, the paper has thus been substantially reorganized. The order of paragraphs has changed and portions of the text have been simplified. However, we did not merge some of the “optimization” sections (2.4.1 and 2.4.3) into the section 2.3 as suggested but rather keep them separated and reduced to the core points. The main changes are thus:

- **A drastic reduction of the method section with a decrease of the text by nearly 50% in order to keep only the main points that constitutes the backbone of the paper.**
 - **We have thus created an Appendix to gather all methodological details that are needed to fully understand the different derivations.**
 - **As the method section refers to CO₂ and OCS concentration measurements early in the section, we moved the corresponding paragraph 2.3 (measurements’ description) at the beginning of the method section.**
 - **As mentioned above, we did not merge section 2.4.3 (optimization setup) into section 2.3 but kept the description/choice of the errors associated to the parameters that we optimize (flux model error and transport error) separate from the description of the optimization scheme itself.**
- 2) Give a brief description of the three DGVMs selected, at least in the appendix and 3) Define “S2” reference simulation.

Brief descriptions of the 3 DGVMs that were used and of the protocol used for the so-called “S2” simulations of the TRENDY inter-comparison have been added to the article: only the main points are described in the main text (short and concise) and a more detailed description was added in the appendix.

- 3) Use a common terminology –MSE or RMSE:

The exact denomination should be “MSE” and this has been corrected throughout the entire article.

4) The labeling on Figs 9 & 11 are tiny:

Figures 9 and 11 have been reworked (this was also a concern of reviewer #2), and the labeling uses now larger fonts.

5) There are too many subsections and subsubsections under each section. I would recommend to avoid this. All the sections should be made more concise. Suggested plan:

2 The different surface OCS fluxes and their representation in models

2.1 Sea-air OCS emissions

2.2 uptake of OCS and CO₂ during photosynthesis

2.3 Soil-atmosphere OCS emissions

2.4 Other sources and sinks of OCS

3 Models used in this study

3.1 Terrestrial biosphere models (2.1.2)

3.2 Atmospheric transport model

4 Atmospheric OCS observations used for validation

5 Experiments and data analysis

5.1 Forward simulations for OCS

5.2 Forward simulations for CO₂

5.3 Optimization scenarios for OCS

5.4 Data analysis

6. Results (to be reorganized accordingly)

Etc...

We agree with the referee that the subsubsections were adding complexity to the paper structure and we thus followed his advice to make it simpler and clearer. We have thus dropped one level of sub-heading and decreased the number of subsections. This was possible given a substantial reduction of the text (mainly of the method section). We mostly followed the plan proposed by the referee, although with few differences:

- **The paragraphs describing the optimization scheme and the corresponding results have been maintained as we believe it is an important aspect of the paper that highlights the combine potential of OCS and CO₂ tracers in an optimization scheme (atmospheric inversion or model parameter optimization)**
- **The result section presents the simulated OCS surface fluxes in the same order as described in the method section, which makes the paper-structure more linear and easier to follow.**

Note finally, that the way to present or discuss the atmospheric concentrations of OCS in the different sections (forward modeling, optimization and general discussion) has been standardized. Each of them is discussed in this order:

- **The long term trend**
- **The amplitude and phase of the mean seasonal variations**
- **The annual mean concentrations of OCS and the north-south gradient**

Technical comments:

We thank the reviewer for all these suggestions. Overall, all technical comments have been addressed, including improvements of grammar, organization and clarity of the paper and adding several important definitions:

I wouldn't recommend abbreviations in the title. Replace 'of current vegetation models' to 'in current vegetation models'

done

P1 (abstract)

7. define LRU in the abstract where it is first used

done

8. velocities *of* plants

done

14 *in* soil and vegetation uptakes

done

P3

20: Here, we use OCS

done

29: For *this*, we modeled the

done

P4

9: In the next step, we define uncertainties

done

20: uncertainties *in* the surface fluxes

done

23: uncertainties *in* OCS surface processes

done

25: *In* the second *section*, we investigate

done

P7

16-17: seasonal cycle from each model and compute the uptake of OCS by leaves using GPP (as described above)

done

P8

5: define C4 plants

done

16: range on *initial estimate of surface fluxes* (Table 2)

done

P9

15: Here, we adopted a different approach *for* the uptake

done

P10

10: The last term in Eq. 3 is not defined

The definition of $[\text{OCS}]_{\text{atm}}$, the atmospheric concentration of OCS (in ppt) has been added.

14: Two different approaches for estimating...here. The first one is *from* Morfopoulos

done

19-20: The second approach is *from* Bousquet et al., (2011) where an atmospheric... H2 concentration is used < continue with the next paragraph here >.

done

24: *allowing* for the

done

27: remove 'being paid'

done

P11

2: Therefore, the *representation* of OCS emissions by anoxic soils in our model was largely

done

4: because of *the* large uncertainties associated with those fluxes, we finally

done

6-8: Rewrite as 'we used the seasonal maps of methane emissions by Wania... from both categories of soils as simulated ... model, to locate the hot spots of OCS emissions from anoxic soils temporally and spatially'

The new paragraph now stands as “Anoxic soil types were mapped accordingly to the representation used in the work by Wania et al. (2010) to represent seasonal methane emissions, as simulated using the LPJ-WHy-ME model. This way, anoxic soils activity were located via methane emissions and translated into hot spots of OCS emissions from anoxic soils, with similar temporal and spatial patterns. “

13: remove ‘a’ from with an assigned +-30%

done

P15

13-14: For each data point, we selected the closest monthly mean

done

21: “%” missing

done

P16

20: Mention “(refer Table 1 for details)”

done

25: replace “to changes in” to “for varying”

done

P23

14 & 22: Instead of “methods section”, give the section number

done

P24

24: Explain Fig 2 and then continue with the explanation

Before driving conclusions from the figure, the explanation of the figure is now clearly made at the beginning of the paragraph: “Figure 2 compares the simulated monthly mean atmospheric OCS concentrations with the observations at Mauna Loa (MLO). Mauna Loa (MLO) is a mid-latitude background station in the middle of the tropical Pacific Ocean (20°N, altitude 3500 m). The data therefore represents the integrated contribution of the surface fluxes from the entire Northern Hemisphere (Conway et al., 1994).”

P25

7: The annual trends of OCS in SPO is not shown in Fig. 3 : remove “(Fig 3)” from this line

done

10: Again, explain “smooth seasonal cycle” in the text of fig 3

As requested, the definition was added in the figure caption: “Raw data were fitted with a function including a polynomial term (1st order) and four harmonics. The residuals of the functions were further smoothed in the Fourier space, using a low pass filter (cutoff frequency of

65 days) to define a so-called smoothed curve (function plus filtered residuals). The mean seasonal cycle is defined from the smoothed curve after subtraction of the polynomial term. The corresponding smoothed seasonal variations obtained are displayed in lower panels. “

P28-29

26-28: Line incomplete. Rewrite

Now correctly standing as: “Table 2 summarizes the initial and the optimized values of the surface fluxes for the different optimization configurations. “

P32

8: Remove “remaining” and replace with “other biases still exist”

done

12: Replace the title of section 4.1 as “Additional biases in simulated atmospheric OCS concentrations”

done

P34

8: pointing out again the *possibility* of too *high* leaf uptake, which

done

18: arise due to the optimization of only one global annual scalar for each flux component

done

P36

8-11: Rewrite the sentence as “the version of the LMDz model used here is believed to have large mixing in the ..., which would thus dampen the amplitude...”

As requested, the new sentence now stands as “The version of the LMDz model used here is believed to have too large mixing in the planetary boundary layer (PBL) (Patra et al., 2011, Locatelli et al., 2013), which would thus dampened the amplitude of the seasonal cycle.”

P39

13: Replace “evidenced” with “shown”

done

Answer to reviewer number 2

Launois et al. present a modeling framework that evaluates the gross primary production (GPP) from three vegetation models (LPJ, NCAR-CLM4 and ORCHIDEE) using atmospheric OCS. The surface fluxes of OCS are optimized using an inversion system that optimizes the scaling factors for various fluxes within prescribed uncertainties. Furthermore, a series of sensitivity tests have been performed to study the influences of the model setup on the simulated results, and the amplitude and the phase of the seasonal cycle of OCS and other features are discussed. As GPP cannot be directly measured at a large scale, a number of methods have been utilized to estimate GPP, e.g. eddy covariance, fluorescence, isotopes, and OCS, which are all, to a certain extent, promising but have limiting factors. Specifically, the use of OCS is limited by our understanding of the correlation between the uptake of OCS and the gross uptake of CO₂, and of the other OCS budget terms. Furthermore, the transport uncertainty also plays an important role in such studies. It is therefore necessary to take into account the abovementioned uncertainties in the evaluations and interpretations of the GPP estimates.

We thank the reviewer for his useful comments that help to improve the manuscript and to highlight the strength and weaknesses of the OCS tracer to constrain GPP. All comments or remarks have been addressed and specific improvements of the text have been implemented in the manuscript (in the relevant sections). Details of all changes are described hereafter.

General remarks:

1) As the LRU from Seibt et al. (2010) with a global average value of 2.8 is in the upper range of estimates discussed in section 2.1.2 and section 4.1.1, wouldn't it make more sense to allow the scaling parameter `kplant_uptake` to vary more in the lower range than in the upper range? Actually the mean values of LRU from Berkelhammer et al. (2013) and Stimler et al. (2012) differ more than 30% from the value of 2.8. Lowering the LRU would have a significant impact on the optimized fluxes.

We agree that the global mean LRU value from Seibt et al. (2010) is in the upper range of the estimates reported by all recent studies that have evaluated this parameter.

However, it must be taken into consideration that:

- **the studies cited by the reviewer (Berkelhammer et al., 2013, Stimler et al., 2012) did not take into consideration as many biomes as Seibt et al. (2010), but only 2 or 3 specific ones. Therefore, for our global modeling study, the results from Seibt et al. (2010) were crucial to provide specific LRU values for different ecosystems. We thus choose to rely on a study that could provide "observation-based" ecosystem variations of LRU. Note that the LRUs in Seibt et al. (2010) vary from 1.5 for**

Xerophytic and Shrub vegetation to 3.6 for cold deciduous forest. Given such large variation range it is crucial to account for ecosystem specific LRUs.

- When our study was designed, early 2012, only few papers with LRU estimates were available and Seibt et al. (2010) was the most comprehensive one.
- With a 30% range of variation on the scaling of the plant uptake for the optimization, we are still able to obtain a set of fluxes that provides an equilibrated global budget for OCS (as seen by the atmosphere).
- Our initial objective was to test an idealized case where the LRU would be known at a 30% level of accuracy for the high error case and at 10% for the low error case. Choosing a 30% error on the initial surface fluxes simulates this low level of accuracy.

However, given the concerns raised by the reviewer and in particular the fact that the optimization of the leaf uptake coefficients for ORCHIDEE and LPJ as well as the soil uptake coefficient hit the bounds, we have redone the optimization with a larger range of variation for leaf uptake and soil uptake coefficients, defined as +/- 50% of the prior value. The new results are slightly changed compared with the previous ones (using a range of variation of +/- 30%), but the different scaling coefficients do not hit the bounds anymore. The leaf uptake coefficient decreases by 38% for LPJ and by 46% for ORCHIDEE. The soil uptake coefficient decreases by up to 36%. We have thus modified the text, the tables and the relevant figures according to this new optimization set up. The main conclusions of the paper remain the same.

2) The plant uptake and the net soil uptake of OCS are collocated sinks. Can the authors specify what differences have caused the inversion system to scale the plant uptake vs. the soil uptake? It is alarming to see that both the soil uptake and the plant uptake of OCS are reduced to the lower limit (30%) for the LPJ and the ORC models in section 3.3.2. It may indicate that both are still overestimated in the optimized fluxes.

The reviewer raised two complementary points.

Concerning the first point (what caused the inversion system to scale the plant uptake vs. the soil uptake) we agree that this is a crucial point of the proposed “OCS” diagnostic:

At first order, the uptake of OCS by soils and plants are indeed co-located in space and they also co-vary in time since both processes lead to maximum uptakes during the respective summer period of each hemisphere (December to February in the Southern Hemisphere, June to August in the Northern Hemisphere). However, there are significant differences between the two absorption fluxes that are crucial:

- **First, according to our description of soil OCS absorption (a function of H₂ deposition in the soil), the seasonal variations of this component are much smaller than the seasonal**

variations of the leaf uptake of OCS. For instance, there is still absorption of OCS by soils in the mid-latitude regions during the coldest months of the year. These seasonal differences are illustrated in Figure 4 (by comparison to Figure 2) and with the supplementary figures (Figures A2 and A3). The use of different ecosystem models with different amplitude of the GPP seasonal variations lead to significant differences in terms of OCS leaf uptake and atmospheric OCS concentrations (Figures 2 and A3). On the other hand, changing the coefficient of proportionality between “soil H₂ uptake and OCS uptake” or the estimate of H₂ uptake (Morfopoulos et al., 2012 or Bousquet et al., 2011) does not change significantly the amplitude of the OCS fluxes and atmospheric concentrations (Figures 3 and A2).

- Second, at high latitude (>50°S and >50°N) the vegetation is absorbing OCS, while soils are acting like net sources, especially in regions dominated by anoxic soils.
- Finally, there are regional differences in the spatial distribution of OCS soil and leaf uptake (see Figure 1) and for the leaf uptake, significant differences also occur between the 3 vegetation models (see for instance the likely overestimation of the GPP in the Northernmost regions for ORCHIDEE). Thanks to these differences, the inversion system allows to scale differently the plant uptake from the soil uptake.

Therefore, given that the inversion system only optimizes a global coefficient for soil and plant uptakes, the constraint brought by the atmospheric OCS seasonal cycle applies differently to these two components. To the extent that OCS uptake by soil follows the estimated seasonal variations of H₂ uptake, these variations are small enough so that the inversion system will optimize preferentially the leaf uptake to match the atmospheric signal. However, we acknowledge that the lack of comprehensive and global observations/measurements of soil OCS uptake calls for caution and that the hypothesis adopted for OCS uptake by soils may need to be revisited in the future.

Although the discussion and conclusion were already pointing out the importance of the differences between the seasonal amplitude of the leaf versus soil OCS uptake, we have slightly modified the text to insist on this crucial aspect, especially in the result section 3.2 and 3.3.

Concerning the second point raised by the reviewer (the extreme reduction of soil and plant uptakes):

The fact that both the soil uptake and the plant uptake of OCS are reduced to the lower limit (30%) for the LPJ and the ORC models could indeed be a concern since an optimal solution for both uptake components was not provided by the optimization. In the revised manuscript the range of variations was changed to +/- 50% for the scaling leaf/soil parameter (“High error” case). This new set up leads to optimized fluxes that are no longer at the lower limit for the soil

and leaf OCS uptake for all models. Note that even with the limits set at +/- 30% several points were already showing that the optimization results were close to optimal:

- The optimization, when using the CLM4CN, leads to a reduction of the vegetation uptake to 772 GgS yr⁻¹ (not at the limit) a value that is somewhat similar to those obtained with ORCHIDEE and LPJ (708 and 663 GgS yr⁻¹, resp.)
- The global budget after optimization was balanced for all three models

3) Given the coarse spatial resolution (3.75degree x 2.5degree) of the model, it is true that the representation errors for sites in the Northern Hemisphere should be larger than for those in the Southern Hemisphere, as defined in section 2.4.3.

- However, I wonder whether the value of 26 ppt for the Northern Hemisphere is too large. It is 5% of the annual mean value, however, is up to 15-20 % of the seasonal amplitude.

We agree with reviewer #2 that the observation errors selected for the optimization scheme are relatively large and that too little justification of this choice was provided in the text. We should first recall that the choice of the so-called “observation error” is difficult. It should gather the measurement error as well as the model error including the flux model error, the transport model error and the representation error (scale mismatch between the observed concentration at a given location and the model concentration at coarse scale. Usually the measurement error is relatively small compared to the modeling error. A proper assessment of model error could be done with the use of different models with different parameterizations. However, for transport modeling studies this is usually not feasible and simpler approaches are used. As a first approximation, we can use the RMSE of the prior model-observation concentration differences at each station. We choose such simple approach and further averaged the RMSE by latitudinal bands to avoid the complexity of longitudinal differences in model skills. In this case, high-latitude stations such as ALT were displaying large prior MSE (nearly 2000 ppt² year⁻¹, see Fig. 11)) and were therefore assigned with a large observation error in the inversion. Note finally that we took slightly larger errors to account for the fact that the “observation error matrix” in the inversion is assumed diagonal and thus neglect all error correlations. Given that these correlations are likely positive between different months of a given site, inflating the standard deviation in that matrix is a way to account for the missing correlations (and thus not to overweight the information content brought by the data).

However, we have done several sensitivity tests (see below) and kept our initial choice but with improved justifications (see main text).

- This may be also part of the reason why the annual mean differences in the Northern Hemisphere sites are larger than those in the Southern Hemisphere shown in Figure 10?

We agree with the reviewer that the large error on the observations (26 ppt in the mid- and high-latitude sites in the Northern Hemisphere) might be part of the reason why station-to-station gradient differences remain poorly captured in the North.

However, we would like to raise several other issues that could explain the differences seen in Figure 10:

- First, the observed station-to-station gradients in the Southern Hemisphere are smaller than in the Northern Hemisphere and thus easier to model. We should recall that the simulated mean concentration across all stations has been reset to the observed mean
- Some of the station-to-station gradients in the North are also well captured as between BRW and ALT, between MLO and KUM or between MLO and NWR.
- The station-to-station gradients in the North that are not well reproduced involve contrasting sites that monitor continental conditions (LEF,) versus “background” conditions (NWR, BRW, MLO). The associated gradients are relatively large and not always well captured because of transport modeling issues: i.e. the difficulties to represent local sources/sinks influences (LEF), topographic effect (NWR), coastal versus continental influence (MHD),...

We have nevertheless tested the impact of the observation errors on the optimization. We tested different configurations, including one with equal errors for all sites (set to 18ppt). The results are not substantially modified and at least the main conclusions of the paper remain. We thus decided to keep our initial choice and to better justify in the method section the selected values.

Detailed remarks:

P27670, L1: removing “during photosynthesis”. The hydrolysis of OCS is expected inside the leaf, but independent of the photosynthesis process.

We agree with the reviewer and as suggested we changed the sentence.

P27670, L10: what does DGVMs stand for?

The acronym stands for Dynamical Global Vegetation Models as commonly used in the land surface modeling community. It has been detailed at its first occurrence, both in the abstract and in the core of the article (introduction)

P27671, L2-3: Where do the ambient concentrations of OCS and CO₂ come from?

These data were taken from the NOAA-ESRL database. Note that in section 2.3 the source for these data was made explicit: “Atmospheric OCS and CO₂ concentrations used in the present work are from the NOAA/ESRL (National Oceanic and Atmospheric Administration/Earth

System Research Laboratory/Global Monitoring Division Flask Program) database, where OCS measurements from 10 stations have been gathered since 2000 (Montzka et al., 2004).". Following reviewer#1 comments, we have shortened and restructured the method section and we have thus moved the description of the data sources to the beginning of the section.

P27672, L12: replacing "soil water content" with "the fraction of water filled pore space".

Done

P27678, L21: "range and uncertainty of -30/+50%" means the variation range from -30% to +50%? **Indeed there was a missing "%" sign. We now change to "-30% to +50%".**

And what is the prior uncertainty?

The prior error was set to 30%; this has been explicitly added in the text.

P27679, L9: how are the forward model errors estimated?

As described above, the transport model errors are difficult to estimate. For the optimization scheme, the quantity that needs to be estimated is the sum of the transport model errors, the measurement error and the representation errors (scale mismatch between model and observations). As commonly done in other atmospheric tracer inversion studies (see for instance a review of atmospheric inversions by Peylin et al. 2014), we took the average RMSE of the prior model-observation differences as a proxy of this overall error. As a further simplification we average the RMSE for the stations by latitudinal bands. We agree that too few details were provided and as detailed above we have improved the text to better describe the observation errors.

P27680, L9: "EDGARD-v4.1" → "EDGAR-v4.1"

done

P27681, L3: "Table 3" → "Table 1"

done

P27682, L24: "Table 1" → "Table 2"

Done

P27687: the section of 3.2.1 should be shortened or be removed. It is so obvious that unbalanced fluxes lead to annual trend in the simulations, isn't it?

The paragraph has been shortened by roughly 50%, dropping especially the obvious conclusions. However, the paragraph could not be removed, as one important point was to discuss with the help of sensitivity tests the fact that ocean and soil flux corrections can be scaled

to improve the annual trend of the atmospheric OCS concentrations without affecting the amplitude of the seasonal variations, contrary to the uptake of OCS by leaves.

P27727, Figure 10. What does the sentence mean? “Note that the global mean for each mixing ratio series has been set to the global mean of the observations”.

The previous labeling was indeed unclear, as noticed in a previous question by the reviewer #1. The misleading labeling has been changed to “Note that the global mean across all stations for each modeling experiment has been set to the global mean of the observations.”

P27728, Figure 11. The axis labels are hard to read. An increase of the font size will be helpful.

This has also been pointed out by referee #1. The labeling has been changed, with larger fonts.