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Interactive comment on "Carbonyl sulfide (OCS) exchange between soils and the atmosphere affected by soil moisture and compensation points" *by* Rüdiger Bunk et al.

Rüdiger Bunk et al.

r.bunk@mpic.de

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REFEREE: This is an interesting study that investigates how the net carbonyl sulfide (COS) exchange between a set of soils (forest and agricultural) and the atmosphere is composed of two opposing component fluxes (a gross uptake flux of COS and a gross production flux of COS) that are regulated differently in response to variations in soil water filled pore space. They revisit the compensation point method of Conrad, 1994 and Lehmann & Conrad, 1996 to obtain estimates of COS production from observations of the net soil COS exchange measured under two different atmospheric COS concentrations of 50 and 1000 ppt. These concentrations are much lower than

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those used in Lehmann & Conrad, 1996 and likely more applicable to concentrations observed in the field. Although many studies have measured and modeled from theory the response of the net COS exchange to variations in soil moisture for a given soil and how differences in soil texture and bulk density play on the WFPS and subsequently the optimum net COS exchange (i.e. the maximum COS uptake rate measured), this is the first study to show that the COS production rate remains more or less constant over the entire range of %WFPS. Thus variations in the net COS exchange with WFPS are driven by the uptake component of the net COS flux. Between the different soils measured (3 spruce and 1 agricultural site) there were large differences in the magnitudes of the net COS flux and their component fluxes with largest fluxes for all components found in the Finnish forest and the lowest component fluxes observed in the agricultural soil in Germany. In addition a further experiment showed that after drying the agricultural soil for several months and re-humidifying, a very similar response of the net and component fluxes to % WFPS was observed, although no statistics were completed to test whether they were significantly different or not. In general there is some nice data in this study that is definitely worthy of publication.

AUTHORS: We thank the referee for his review, the good suggestions made and invested work.

REFEREE: Major comments Throughout the manuscript there is very ambiguous application of terminology regarding the exact flux being presented. When the paper is expressly about partitioning the components of a net flux, one has to take care to be precise and state clearly which flux they are writing about. I thoroughly recommend that the authors go through the paper and clarify exactly what each flux is that they refer to, when they refer to it in the paper. Simply referring to COS exchange is too ambiguous, this paper must always refer to the net COS flux (EOCS), the COS emission rate or production rate (POCS) and the COS uptake rate (UOCS).

AUTHORS: We agree clear terminology is very important here and will revise accordingly. All instances will be either be net release (EOCS), gross uptake (UOCS) or gross production (POCS)

REFEREE: Furthermore, the partitioning approach taken in the current study does not completely isolate the two component gross fluxes, rather the uptake term measured at a constant temperature as presented by the authors is still regulated to some extent by diffusion (not strictly enzymatic uptake of COS) and the production term as presented still incorporates a COS deposition velocity (Vd0) that occurs even when the COS concentration = 0. These details are developed in the Ogee et al., 2016 paper https://www.biogeosciences.net/13/2221/2016/bg-13-2221-2016.pdf and more relevantly to the current study in a recent publication in Atmospheric Chemistry and Physics Discussions by Kaisermann et al. https://www.atmos-chem-physdiscuss.net/acp-2017-1229/acp-2017-1229.pdf that demonstrates within the current methodological framework that a small additional analysis is required to obtain the gross COS production rate when COS concentration = 0 that must be solved iteratively see Eqs 2, 3 and 4 of Kaisermann et al.

AUTHORS: We agree that the most accurate POCS values would be obtained with the method described in Kaisermann et al., 2018. However, when we estimate the changes this correction would make to POCS in this study, these changes are below the uncertainty of obtained values: Derived from eq2 and eq3, we can say that the gross uptake (UOCS) is the consumption rate coefficient (kOCS) times the ambient OCS concentration (C). →UOCS= kOCS x C. For a rough estimation how much POCS will maximally be changed by Vd0 we can assume that the concentration C0 that the sinks in the soil are exposed to when the mixing ratio in the flushing air is zero can be derived from the (uncorrected) production rate. With the median production rates of 2, 4, 7 pmol g-1 h-1 and maximum kOCS of -0.005, -0.0015, -0.0013 and -0.0146 mol g-1 h-1 for Mainz soil, Waldstein soil with spruce or blueberry understory, and Finland needle forest litter, respectively, we get an estimated correction of 0.010, 0.011, 0.005 and 0.394 pmol g-1 h-1 These corrections would be smaller than the uncertainty of our measured exchange rates. As we do not have the accurate soil depth (zmax) and

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would have to use estimates (based on filling measurement chambers again with the amount of sample used in the experiments and measure soil depth), we think we can neglect this correction. Of course, the topic of these corrections will still be discussed in the revised version.

REFEREE: Another important point that was expressed several times in the paper is that the shape of the net COS exchange response to WFPS is unknown but probably caused by changes in the activity of the enzyme CA. However, this is not strictly true as in the past few years the community has made considerable progress in explaining the response of the net COS flux to variations in soil temperature, soil moisture, soil texture and soil microbial biomass collectively in the papers of Kesselmeier et al., 1999; Van Diest &Kesselmeier, 2007; Ogee et al., 2016 and Sun et al., 2016. From these papers it has been shown that the observed optimum with soil moisture content observed in the present study and many times in the literature can be modelled extremely well and is mostly caused by changes in the diffusion of OCS within the soil matrix that reduces the potential hydrolysis rate at a given temperature, microbial biomass and COS concentration. This can occur over the typically short time frames of these experiments and thus the net COS flux and the gross COS uptake rate does not need to be driven by changes in the intrinsic enzyme activity or size of the microbial population. Thus the discussion needs to take this in to account and furthermore considered in the interpretation of the data presented in the results. Subsequently, differences in soil texture can probably explain most of the differences in the absolute %WFPS values where the optimum net COS flux is observed. Unfortunately, no data is provided in the manuscript about the differences in texture between the sites, this should be added. As described above the data in this study are interesting however the presentation of the results could definitely be improved and synthesised.

AUTHORS: We agree that these aspects are underrepresented in the current version of the manuscript. We will cut back the deliberations about microbial communities and enzyme activity in the revised version and will scrap the discussion about het-

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erotrophs/autotrophs altogether. Instead we will expand the discussion about diffusion limitation by soil moisture, chemical properties of the soil (especially ammonium content). Unfortunately, only limited texture information is available.

REFEREE: I see no reason why figures 1 and 2 are not merged and a more synthetic analysis of the fresh vs dried/re-humidified soil results presented in a new Figure 2. This new figure could consist of 3 panels side by side. In panel (a) the authors would present the data from the 50ppt experiments, panel (b) the 500 ppt and panel (c) the 1000 ppt data. On the x-axis of each panel would be the net COS flux from the fresh soil against axis y the net COS flux from the same re-humidified dry soils. The authors could then colour the points by %WFPS (light blue = dry soils and dark blue = wet soils). Then they could also show the 1:1 line and do some regression statistics that way the audience can assess clearly and objectively the effect of the drying on the %WFPS response. The authors also point out that the consumption rate coefficient (k) follows the same pattern with %WFPS as the net COS flux and the partitioned COS uptake rate (Uocs) and present the variability of k with %WFPS. However, U and k must vary with soil moisture and temperature. . . etc as they are linearly related or even proportional if Vocs is always at the same COS concentration. Thus I would remove figure 5 and rewrite the discussion to address this point.

AUTHORS: We agree that Figure 1 and 2 can easily be merged and will do that. We thank the referee for the very interesting suggestion for a new comparison figure dry vs. fresh and will adapt this.

REFEREE: Also the compensation point does not affect the COS exchange rate and thus the title of the manuscript should be corrected.

AUTHORS: Yes, we agree that the title is misleading and will change it.

REFEREE: The authors state that the data from the Sun et al 2017 field study and this study are comparable and can be used to transfer the findings from the lab data to the field. However, the lab response to soil moisture content (green line) does not go through the middle of the points, but rather forms an upper envelope and there is no statistical test behind this statement. At the minimum the authors should calculate the mean deposition velocity for the relevant and comparable soil moisture values for their study and the Sun et al study and compare the means. Furthermore, I do not think it is appropriate to use the temperature optimum from the Mainz soil to make the field fluxes of the Finnish soil comparable. It has been demonstrated before that the net COS flux is strongly affected by the production rate and can cause a shift in the temperature optimum. As this study shows the Mainz and Finnish soils have extremely different production rates I do not think this is the most appropriate way to reconcile the two data sets and facilitate comparison.

AUTHORS: We agree that averages or a fit are a good idea and will implement this. We think that using the temperature curve from Kesselmeier et al., is the best available approximation. Another approach would be to follow the Kaisermann et al., 2018. They found 1.23 for kOCS Q10 value for 10° C and a large set of different soils. So for 5° C it should be 0.615. Either method is only an approximation, but in our opinion clearly better than no temperature correction at al. Naturally a discussion of the weaknesses of the correction method will be added.

REFEREE: Specific comments Page 3 of the introduction lacks a number of citations that describe the theoretical advances the research community has made in describing the response of net COS exchange to soil water, texture, soil temperature etc... there is also some internal contradiction within the text. Finally the introduction does not present any hypotheses on why they might expect shifts in sources and sinks relative to their experimental manipulation with COS concentration and soil moisture content.

AUTHORS: We will add a section about the theoretical advances, especially Ogee et al., 2015. A paragraph of the relationship of OCS uptake and ambient OCS concentration will be moved from its current position into the introduction, as has been suggested by another reviewer. We will also add research questions to the introduction, as detailed in our reply to R1.

REFEREE: The characteristics of the soils provided are partially useful. It would be better to provide the physical characteristics such as bulk density or texture rather than nitrate and ammonium fluxes taken from some other studies and not conducted on the soils at the same time as when measured. These values could be misleading as inorganic N concentrations are turned over rapidly and vary with season and management.

AUTHORS: We assume the referee means nitrate and ammonium contents as no fluxes are presented here. We would like to point out that our samples and the samples for the cited studies came from the same sample pool (out of the same bag of soil sample) including the measurement of N contents. Samples were stored at 4°C in the dark and sub-samples drawn for various studies. Therefore, we do not think the referees' concerns apply here. The bulk density will be added to the table.

REFEREE: Pg 4 line 10 what exactly can the author not exclude variability in over time? Was there no fixed protocol for the collection, storage and handling of the soils?

AUTHORS: A common protocol was in place. But the sampling was done by different persons and some of the steps contain a degree of subjectivity. For example, how long and vigorous a sieve is shaken influence how much of the particles sized close to the mesh size pass through. Similarly, the practical decision on the top 5 centimeters of soil contains some subjectivity. However, the problem is probably overstated. We think the statement about variability over time can be removed.

REFEREE: Pg 4 line 14 length of sample storage should be provided here and was it the same for each soil?

AUTHORS: we will add when each sample was collected (Mainz soil: January 2014, the other soils fall 2012). All experiments were performed in February 2014.

REFEREE: Section 2.4 and description of %WFPS protocol should come just after Section 2.1

AUTHORS: this change in sequence is a good idea. We will do that.

REFEREE: Pg4 line 20 state the temperature of the soil here and how constant.

AUTHORS: Soil temperature was 20° C. Temperature variation was less than 0.5° C. The information will be added.

REFEREE: Pg 4 Section 2.2 how many sample replicates are measured for each soil at a time and what do the error bars refer to in the figures?

AUTHORS: The error bars denote the standard deviation of 30 second averages. There have been no replicates. However, some experiments were done multiple times under similar conditions and we found that the resulting exchange over soil moisture was always very similar, though there were several months between these measurements and the soil chamber was moved to another lab and build up again. See the attached figure originally from Bunk et al, 2017.

REFEREE: Section 2.2 How long between wetting and gas exchange started? How long is the measurement sequence? How long is the airstream sampled? How do you check for steady-state? Information on when the soils were sampled would be useful e.g. time of year; before/after fertilization?

AUTHORS: The incubation and sampling technique was described in Bunk et al. (2017). We repeat it here for a better understanding. One measurement sequence (from 100 to 0 % soil moisture) is about 60 hours. Flushing of the chambers started about 1-2 minutes after wetting (the time it takes to screw the chamber tight). Chambers were measured in sequence for 10 Minutes. This means all chambers were constantly flushed and their outlet connected for 10 minutes to the analyzer each 40 minutes (3 samples plus one empty reference chamber). Our setup does not use true steady state conditions (which is not possible with a dynamic chamber approach) but a dynamic equilibrium that is for all purposes of this study analog to true steady state conditions: In a well-mixed chamber (ensured by the activity of a fan in the headspace of each cuvette in our setup) the concentration of a trace gas is determined by the concentration in the flushing gas, the rate of flushing and the release or uptake by the soil. The flushing rate is constant and monitored. Factors like soil moisture are very slow changing in relation to the measurement period averaged for calculation by eq1. Chamber effects are accounted for by using an empty identical chamber for reference. While true steady state conditions can only be archived with static chambers, a dynamic equilibrium was archived in flushed chambers that fulfills all conditions required to employ eq1. This dynamic equilibrium was ensured as described below and can/will be involved in the revised paper: Switching the analyzer inlet from one chamber to the next of course courses some disturbances in the chamber and also the inlet tube needed flushing. We did extensive pre-measurements by observing measured chamber concentration with chambers that were empty or loaded with soil samples, waiting for steady concentrations to be measured. With the length of inlet tube and flushing rate used, stable concentrations were always observed after less than six minutes. Based on these empirical determinations, the time each chamber was connected to the analyzer was set to ten minutes. Of these ten minutes, the first 7 minutes were discarded to allow for proper equilibration within the chamber and at the same time allowing proper flushing time for the analyzer and analyzer inlet tube. The following 2.5 minutes were averaged for 30 seconds intervals and used for exchange calculations. The last 30 seconds of each chamber cycle was discarded again to eliminate any chance of accidental overlap. Information about the sampling year and other available information about the soil sampling time and conditions will be added.

REFEREE: Section 2.4 remove the citation Bourtsoukidis et al

AUTHORS: Bourtsoukidis et al. were the ones from whom the method was adapted. The paper is now published so we think they should be cited.

REFEREE: Page 6 Line 24 is it the fluxes or absolute mixing ratios underestimated by 7%?

AUTHORS: The absolute mixing ratios. But since the fluxes are derived from the difference between the sample and reference mixing ratio, that propagates to the fluxes as well.

REFEREE: Page 7 line 12 I feel the last sentence of this paragraph is out of place and should appear later.

AUTHORS: We respectfully disagree. EOCS and POCS are described here, why would UOCS be out of place?

REFEREE: Section 3.2 really needs to be re-written. It is ambiguous, difficult to read and contains repetition. In places it is hard to work out what the authors are comparing.

AUTHORS: We will find a clearer phrasing.

REFEREE: Page 8 line 3 you should state that the soils had compensation points higher than the background atmosphere.

AUTHORS: We agree and will do that.

REFEREE: Section 3.3 again some repetition at the end of the paragraph. Also the authors should point out that compensation points in themselves are not particularly useful as they are not intrinsic properties of a soil as their value will vary with temperature and COS concentration. They are only useful for establishing whether a soil is a source or sink of COS to the atmosphere.

AUTHORS: We partially agree. The last bit of the paragraph can be pruned as it is said before. It is also true that CPs can only give limited information because they are co-dependent from many factors. However, such a discussion belongs in the discussion section. We will add it there.

REFEREE: Section 3.4 the figure 4 panel b production rate looks very strange. I am not sure why it has this appearance, but I am guessing maybe there is some interpolation being made between a limited number of measurements over the drying curve. It would be useful to explain what is going on here in the methods, results and the legend

AUTHORS: We agree that the production rate for this sample looks a bit strange. However, we could not find any indication that something is out of place. At around 50% WFPS the scatter of data points for the 50ppt net release (EOCS) measurement is a bit increased. This coincides with the “out of place” peak for the calculated production.

REFEREE: Page 8 line 19-20 This statement as described above is redundant as is the figure 5 and is not linked to any of the things proposed in Section 4.2

AUTHORS: As stated above we agree this sentence can be pruned.

REFEREE: Page 8 line 30 this correction may have some problems associated with it, if the authors insist on using it they should be more critical about why it is not ideal.

AUTHORS: We agree that this correction is not ideal. A discussion of the potential problems is warranted and will be added. However, we think it is the best possible approximation (see also comments to referee 1).

REFEREE: Page 9 Ln 3 the reason for the scatter is because many variables are changing at the same time, please be a bit more critical.

AUTHORS: Yes, this was also our conclusion. That is why we wrote in line 5: “We suggest that the stronger scatter in the field measurements is due to additional factors that were kept constant in our lab measurements, but unavoidably vary during field measurements”. Of course we will elaborate a bit more on that point.

REFEREE: Page 9 Ln 8 I would not hold this graph up as evidence that your study can simulate what is happening in the field.

AUTHORS: We disagree. Observed deposition velocity not only is very close in absolute value but also shows the same trend as the averages of the reported field data. This should become even clearer after modifying figure 6 as suggested by R2.

REFEREE: Page 9 line 11-22 This discussion is a little imaginative and is not so relevant to the results. The arguments do not follow a clear logic.

AUTHORS: We have decided to abandon this line of reasoning, instead focusing mainly

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on diffusion limitation, effects of the high ammonium content in the Waldstein soil and other physical and chemical properties.

REFEREE: Page 10 Line 11-13 I don't understand these statements. What exactly has it's activity reduced? What is the different uptake process and what evidence in your data do you have for this?

AUTHORS: That was referring to the aforementioned consumers. However, we propose to only point out there is a follow up study where microbial methods were employed. As we also decided to give up the discussion about heterotrophs and autotrophs in this study, we propose to prune away the lines 7 to 11.

REFEREE: Page 10 line 22 What about abiotic processes?

AUTHORS: "All known and unknown OCS production pathways" was supposed to include abiotic processes. We will revise to "All known and unknown OCS production pathways and processes".

REFEREE: Page 10 line 28-30 ambiguous statements about exchanges and observed values without being precise about what they are referring to. Which exchanges and values exactly?

AUTHORS: For the Waldstein Blueberry soil, the net exchange at 50 ppt is so close to 0 that, statistically, many data points cannot safely be distinguished from a zero exchange (the standard deviation of the sample and the reference chamber mixing ratios overlap). However, for all moistures, there are still some points for which the difference between the reference and sample mixing ratio exceeds the uncertainty. The data points where the difference between the sample- and reference mixing ratio is not bigger than the uncertainty still follow the same trend as the data points where the difference in mixing ratio does exceed uncertainty. Additionally, the Waldstein Blueberry soil shows the same general trend as the other soils at low ambient mixing ratios. We will revise to clarify the reasoning.

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REFEREE: Page 11 section 4.2 lines 7-15 I don't think any of these arguments are relevant to the results they are discussing. The authors do not appreciate the role of diffusion within the soil matrix and it's affect on the ability of soils to take up COS or not as WFPS changes. This should be explored first before jumping to the conclusion that autotrophic organisms have some role to play in explaining this pattern, especially as the authors have no evidence to support this hypothesis.

AUTHORS: Based on reviewer input and reconsiderations we have decided to remove the discussion about heterotrophs and autotrophs as well as strongly reducing most discussion of microbial communities. Instead, other options as physical and chemical soil properties shall be included in the discussion of the revised version. Especially a topic of diffusion limitation by high soil moisture and a brief discussion of the possible role of Ammonium oxidizing organisms, as there are significant differences in Ammonium content in the examined soils that coincide with curve shapes of UOCS and kOCS, will be included.

REFEREE: Section 4.2 last paragraph should be in the results section it is not discussion.

AUTHORS: We will move the paragraph to the results section

REFEREE: Page 12 line 3 also mention the other factors that will alter the compensation point.

AUTHORS: Agreed.

REFEREE: Page 12 line 14-16 your experimental data does not support this statement about the two compensation points of Lehmann & Conrad please modify the sentence.

AUTHORS: We will remove this sentence.

REFEREE: Page 13 Ln 2-19 this discussion again contains a lot of conjecture and fails to mention that the differences in texture between the spruce sites is probably important and should be accounted for before attributing differences in COS uptake rates to other

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factors

AUTHORS: We have decided to completely abandon the heterotroph/autotroph reasoning as not supportable with our data. Instead it will be discussed in the view of diffusivity and soil properties, especially Ammonium content (which is very high for the Waldstein soils). Also a topic of consideration will the origin layer (topsoil vs organic layer).

REFEREE: Page 13 line 23 I think this is important info that should appear in the methods section.

AUTHORS: The sampling year will be added for all soils to their description in the methods section and Table 1.

REFEREE: Page 13 line 28-30 don't you think this is because fundamentally the soil texture did not change over the 6 years at the Mainz site and thus you observe a similar pattern when you wet and dry the soil 6 yrs later?

AUTHORS: That's the point. The soil was sampled, then sampled again six years later. We get very similar patterns, so neither the physical properties nor the sink strength (determined by the microbial communities inside the soil) can have changed too much. And that is something that is good to know, as many budget calculations rely on soil studies that have only been done once. It may mean that agricultural soils do not change that much, unless their crop-cycle or fertilization mode is changed.

REFEREE: Page 13 line 18 LAI of 15? I don't believe this is possible

AUTHORS: This is the value given by the cited authors. Any debate of the accuracy of this value would have to be taken up with them. Besides, even if that number is not accurate it is still undisputed that the LAI can vary a lot from site to site. Either way, section 4.6 will be removed due to its tangential nature.

REFEREE: Section 4.6 I think this is a bit long and not so useful in the end I think it could be summarized in a sentence or two in the conclusion Conclusions are currently based on conjecture and not the results. Statement 1 The experiment was not designed to test and cannot prove that COS is driven by the litter layer. Statement 2 There are no data presented about fungi in this paper so again this statement seems redundant. Statement 3 They do not prove that COS uptake is driven by different enzymatic processes. Statement 4 Is an introduced error of 1% really significant? Statement 5 No evidence in this study that the correlation coefficient k is linked to the presence or absence of auto- or heterotrophic organisms. Statement 6 I agree trying to understand compensation point variability without a model that accounts for how it varies with T, moisture and COS concentration is frustrating. We should use the theory and models that now exist to address this issue. Statement 7 they did not demonstrate statistically that the storage issues introduced significant differences in the fluxes and what level of uncertainty is introduced.

AUTHORS: Based on the input by R1, R2 and R3 we decided to remove section 4.6. The main message will be added condensed to a few sentences at an appropriate point. The conclusions might be a good place for that, as the referee suggests. Based on referee feedback, the discussion will change quite a bit and the conclusions will be revised accordingly.

REFEREE: Merge Fig 1 and 2

AUTHORS: we will do that

REFEREE: Recommend a synthetic figure 2 with some statistical analysis.

AUTHORS: We will add a figure as suggested in the reviewer's general remarks. Thank you again for the suggestion.

REFEREE: Figure 3 should there not be some estimation of error on these points?

AUTHORS: The error of these points will be discussed within the text of the revised version and a reference to that discussion added to the figure caption in the revised version. Error bars will be added if they do not clutter the figure too much.

REFEREE: Figure 4 panel b looks weird also can you show that the inlet is constant during each of the experiments and that steady state was attained

AUTHORS: Please see our comment on the topic of steady state above. Regarding the inlet, inlet flow rate was logged and is constant.

REFEREE: Figure 5 redundant Figure 6 not sure this is necessary either

AUTHORS: We disagree

REFEREE: Figure 7 not sure this figure is explicitly referred to in the text or necessary in the paper.

AUTHORS: The figure is referred to in section 4.5

REFEREE: Table should state explicitly which nitrate and ammonium data are relevant to the gas exchange measurements taken during the actual present study experiment.

AUTHORS: please refer to our reply to your major comment section above.

Referenced literature:

Kaisermann, A., Ogée, J., Sauze, J., Wohl, S., Jones, S. P., Gutierrez, A., and Wingate, L.: Disentangling the rates of carbonyl sulphide (COS) production and consumption and their dependency with soil properties across biomes and land use types, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-1229, in review, 2018

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Ogée, J., Sauze, J., Kesselmeier, J., Genty, B., Van Diest, H., Launois, T. and Wingate, L., 2016. A new mechanistic framework to predict OCS fluxes from soils. Biogeosciences, 13(8), pp.2221-2240.

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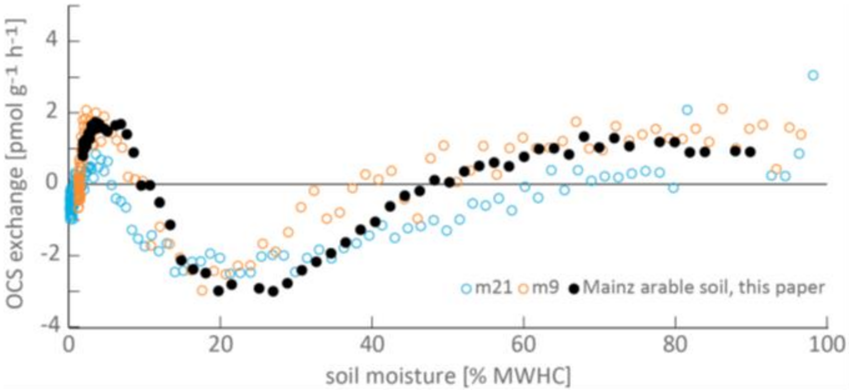

Fig. 1. Three measurements of EOCS at 1000 ppt OCS and 440 ppm CO2 under similar conditions in three different experiments. Fingure originally from Bunk et al., 2017

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