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Atmospheric Chemistry and Physics Discussions

Interactive comment on "A top-down approach of surface carbonyl sulfide exchange by a Mediterranean oak forest ecosystem in Southern France" *by* S. Belviso et al.

Anonymous Referee #3

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General comments:

The authors present atmospheric OCS concentrations during a few days in June 2012 and June 2013 and tried to explain the variability by considering the processes in soil, vegetation, and atmospheric transport. Large changes in atmospheric OCS are observed at the site, including large decreases in the early morning, and large increases in the afternoon for data from 2013. I think the authors can do further analysis to show that the reasons that they give for the increasing OCS concentrations are indeed plausible.

The lack of an afternoon peak in 2012 is explained by the fact that for these days the backward trajectories show that the air was transported mostly from the South, and not

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from the industrialized area in the Rhône Valley. I suggest the authors consider doing a windrose analysis to show if the source of OCS is persistent from the same direction.

The authors suggest that the early-morning drop in OCS is caused by vegetative uptake and that it increases shortly after that due to entrainment of air from above the boundary layer. The authors could try to make the existence of entrainment more plausible by looking at for example H2O concentrations. The air above the atmospheric boundary layer is generally drier than within the boundary layer. If the increased OCS concentrations are indeed driven by entrainment, then also a decrease in water vapor concentrations can be expected.

Another dynamical process that should be considered is the sea breeze. Due to large convection over land there is generally lower pressure over land, which causes air to move from sea to land during daytime. The authors suggest that the high peak of ozone in the afternoon data in 2013 is transported by the sea breeze with the source in the Marseille area. I wonder why the ozone peak should come from the Marseille area, and not from the Rhône valley. The wind direction should be shown to indicate the presence of a sea breeze and the correlation with the ozone peak. Furthermore, if the enriched air of OCS is coming from an industrial area a correlation with CO would be expected, was this visible at the site? An analysis using wind direction and other tracers (e.g. H2O for entrainment, CO for advection from industrialized areas) must be done to better characterize the sources (and sinks) of OCS.

In general I wonder why the authors only show data from a few days in June 2012 and June 2013. Did they only measure during these few days? Please point out if these were only two short campaigns. If the authors have a longer measurement period available they should explain why they chose to show only a few days and I suggest they consider including a longer time series of data. This would have added value in characterizing the atmospheric dynamics and the sources and sinks of OCS at the site. For example, by considering a longer time series of data the authors can discuss if the afternoon peaks observed in June 2013 are actually a rare event or if they occur more

often. Besides that, they can discuss if sea breezes are a general characteristic of the atmospheric dynamics at the site. The currently presented measurement period is rather short to draw conclusions on the suitability of the site to study OCS as a tracer for GPP. In fact, the current data show that the ecosystem OCS uptake is not a dominant process for most of the day (e.g. influence of entrainment in the morning and pollution in the afternoon). The authors showed that ERU calculations were limited to only a few hours, which actually suggests that this site is not ideal to study OCS as a tracer for GPP.

Specific comments:

Abstract: Page 1, line 27: I would think it is relevant to say from which absolute concentrations the values drop. E.g. say "... and synchronous steep drops of OCS from ... ppt down to ... ppt". The same holds for O3.

Introduction: Page 2, line 17: "Atmospheric OCS is also removed from the atmosphere by other pathways, such as nighttime uptake by plants...". I would not use the word "pathway" here, as the nighttime uptake by plants follows the same pathway as the daytime uptake by plants, that is, through open stomata. Only the difference with CO2 is that the OCS uptake is not light-dependent, and therefore it is not corresponding with photosynthesis.

Material and Methods: Section 2.1. Site description: It would be worth mentioning the canopy height.

Page 4, lines 16-21. Can the authors briefly explain the method to partition GPP and Reco?

Page 5, lines 1-9: What is exactly a calibration gas provided by U. Seibt and K. Maseyk who purchased it from Air Liquide? $\sim$1 ppm or $\sim$500 ppt OCS? How did the authors find an agreement better than 0.2%?

Page 5, lines 10-11: What was compared/evaluated? Was the cylinder air from NOAA-

ERRL used as target? Replace “certified” with “calibrated”.

Page 5. Did the authors observe a dependence with water vapor when comparing OCSLGR with OCSGC? Kooijmans et al., (2016) found that for the Aerodyne laser spectrometers there can be spectral interference between H2O and OCS, depending on the spectral fit. Did the authors observe something similar?

Page 5, line 28: I think “this manuscript” refers to Yver et al. (2015)? In that case I suggest the authors say “that manuscript”.

Page 6, line 24-27. Both methods seem to be used under wet conditions: “Penman Monteith for RH > 70 %” and “Under wet conditions the stomatal conductance was estimated following Lamaud et al. (2009)”. Did the authors mean to say that one of the two methods is used under dry conditions?

Results: Page 7, line 6: “The two campaigns took place in June of 2012 and 2013.” I suggest the authors mention this earlier in the manuscript, e.g. in the introduction or in the methods. This would make clear already in the methods section that some instrumentation for one variable differs over the two years. Besides that, please explain why only the data from a few days in 2012 and 2013 were used and not a longer time series.

Page 7, line 21: “. . . same for ozone”. Better say: “. . . and the same holds for ozone.”

Page 8, Line 17-21: Is there any relation between the increased water flux and CO2 fluxes? What does this information tell us? I do not see a further discussion about the latent heat fluxes in the discussion session, so does this information have added value?

Page 8, line 21: Maybe the authors can introduce already before what the relation is between isoprene fluxes and CO2 fluxes. That would make clear why the authors measure this. Discussion:

Page 9, line 12: As explained in the general comments I suggest the authors look at

H2O concentrations to see if the morning rise of OCS coincides with a decrease in H2O concentrations, which may be an indication of entrainment.

Page 9, line 17-20: Please explain this better, was there a typical event of excessive biomass burning in North America that could potentially have explained the OCS increase?

Page 9, line 21: "... it is clear that the OCS and O3 peaks have distinct origins". The air has the same origin, but the OCS and O3 enrichment has different sources.

Page 9, line 23: "Backward trajectories at 300 m above ground level ending at 12 UTC, when OCS levels at the O3HP in June 2013 were over 600 ppt, show that the circulation of air masses during both periods was at low altitude...". Define "both periods". Do the authors mean 2012 and 2013? The sentence before points to only 2013 data.

Page 9, line 24-30: It is not clear here what message the authors try to convey. The authors point out two different trajectories: one is from the Rhône Valley, where anthropogenic emissions could cause a rise in OCS. The other is the sea breeze, which (I presume) could transport the high O3 concentrations from the Marseille area, but this peak does not coincide with the OCS peak.

Page 9, line 28-29: The authors state that polluted air from the Marseille area is transported by a sea breeze, leading to an increase of ozone above the boundary layer. Why would a sea breeze cause transport above the boundary layer? I would say this transport happens within the boundary layer as a sea breeze causes horizontal movement from the sea towards land. Please also show why it is plausible that there is a sea breeze, did the wind direction change? Why would the Marseille area cause an ozone peak and not the Rhône Valley? And did ozone correlate with CO for the 2013 data?

Page 10, line 10: the authors probably mean to refer to Fig. 5 instead of 3.

Page 10, line 14: ERU is defined as the ratio of the relative drawdown of OCS to CO2.

Only when the plant uptake is the dominant flux, the ERU is proportional to the ratio of GPP/NEE with a proportionality constant that is the LRU (Campbell et al., 2008). Please make clear that the formulation that the authors use is only valid when the plant uptake is the dominant flux. After that the authors can explain that this is only the case at the OH3P site for a few hours in the afternoon (because at other moments the ecosystem is not the main driver but rather the boundary layer dynamics) and that ERU could only be calculated using the OCS and CO2 gradients for these few hours. Please give the numbers reported by Blonquist et al. (2011). I am also aware of ERU values presented by Maseyk et al. (2014). What do these ERU values tell us about the plant uptake? (Like the authors state in the third reason given in the beginning of section 4.3, see also my next comment).

Page 10, line 26-28: please clarify all three reasons to reach the conclusion that OCS uptake is the only relevant biospheric flux. This is not clear yet.

Page 11, line 31-33: Please rephrase, it reads as if the authors refer to the difference between the three open oak woodlands. But the authors probably mean the difference between these woodlands and the O3HP site. Also be more explicit how this conclusion is obtained: “The fact that no large nighttime drop of OCS is observed at O3HP suggests that the soil is not a net sink of OCS.” The soil temperature and moisture have not changed from 2012 to 2013, and a early morning drawdown was indeed observed in 2012.

Page 12, line 10. Remove “If”

Conclusions and perspectives: Page 13, line 15. Which requirements? Introduce them in the introduction and repeat here. Did the authors refer to the spring in 2012 only?

Page 13, line 15-17: The authors state that the soil uptake of OCS is negligible compared to the uptake of this gas through the stomata, however, I think this conclusion is made too easily. In fact no net exchange of OCS during the night is observed, which could either mean that there is no soil and leaf flux during the night, or that the sources

and sinks (either from the soil or leaves) compensate each other. State clearly that this is just a speculation.

Page 13, line 21: which “second method” do the authors mean? Which is the first?

Page 13, line 19-22: The authors discuss here that LRU is needed to derive GPP from OCS fluxes, and then continue saying that there were difficulties in determining ERU. To my knowledge LRU can only be derived from leaf-level measurements with branch chamber/bag measurements (e.g. Berkelhammer et al., 2014), how do your perspectives tackle the issue of getting LRU values?

Figures

Fig 2. 2012 data are shown, but the 2013 data are at least as important due to the high afternoon OCS peaks. I suggest the authors show both the 2012 and 2013 data. Also interesting to see would be the wind direction as an indication for a sea breeze and H2O as indication for entrainment.

Fig 3a. This can already be seen from Fig 2c and 2d. I suggest the authors include meteo and concentration data of 2013 in Fig 2, then remove Fig 3, and include the average daily cycle of ozone in Fig 4 (to still be able to make the comparison between OCS and ozone).

Fig 5. Please show uncertainty bars for OCS as for CO2.

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