

## ***Interactive comment on “Observations of the uptake of carbonyl sulfide (COS) by trees under elevated atmospheric carbon dioxide concentrations” by L. Sandoval-Soto et al.***

**Anonymous Referee #2**

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This paper is focused on plant acclimation to elevated atmospheric CO<sub>2</sub>, causing reduced activity of the enzymes involved in CO<sub>2</sub> uptake and fixation, including carbonic anhydrase (CA). CA has a high affinity for carbonyl sulfide (COS) and has been shown to be the major enzyme involved in COS uptake by plants. As a result of acclimation to increasing atmospheric CO<sub>2</sub>, plant uptake of COS should decline, causing atmospheric COS to increase. Based on leaf-level CO<sub>2</sub> and COS exchange measurements from two tree species at two CO<sub>2</sub> levels (350 and 800 μmol mol<sup>-1</sup>), the main conclusion of the paper is that plant uptake of COS does indeed appear to decline under conditions of elevated atmospheric CO<sub>2</sub>.

C418

The paper is appropriate for the journal. The writing could be improved considerably – the paper is hard to follow in many places, and it is easy to get lost in all the detail and the nine tables. Writing deficiencies go beyond simple English translation issues.

While this is an interesting paper, with potential implications for the amount of COS in the troposphere and the contribution of this COS to the stratospheric sulfate aerosol layer, there are multiple issues that the authors need to address before this paper can be published.

Two specific hypotheses put forward by the authors in the introduction warrant further discussion in the introduction, and should be brought up again in the discussion, in relation to the results of the study:

P 2126, Lines 5-6 state, “Elevated CO<sub>2</sub> will trigger a decrease of the enzymatic activities which is balanced by a higher CO<sub>2</sub> availability. Thus, the CO<sub>2</sub> uptake will not decline, but a CA acclimation may lead to a reduction of the COS uptake due to a lower metabolic sink as long as the uptake is not also enhanced by higher substrate (COS) concentration.” If carbonic anhydrase activity indeed decreases under future atmospheric CO<sub>2</sub> as the authors suggest, plant COS uptake may not decline in response if atmospheric COS increases, as an increase in atmospheric COS could offset a lowered plant sink. While there is some evidence that global COS sinks and sources are in balance at present (Kettle et al., 2002; Watts et al., 2000), and that atmospheric COS has declined in recent years (Montzka et al., 2004), atmospheric COS has increased significantly since the industrial revolution (Aydin et al., 2008; Montzka et al., 2004), just like CO<sub>2</sub>. Thus, there is no guarantee that atmospheric COS will continue to remain constant and/or decline in the future, particularly where some COS sources are anthropogenic and highly uncertain (Watts, 2000). The papers by Aydin et al. (2008) and Montzka et al. (2004) should be cited and the results should be discussed in relation to the higher substrate concentration statement made by the authors. The discussion about global relevance in section 3.4 seems overly speculative given these issues.

C419

P 2126 line 10: "Furthermore, increased CO<sub>2</sub> without an increase of COS leads to a competitive inhibition of the COS consumption." This is in direct contrast to recent results from Stimler et al. (2010, *New Phytologist*, not cited by the authors), who observed no cross-inhibition effects between COS and CO<sub>2</sub> during leaf uptake by three different species over wide ranges of COS and CO<sub>2</sub>. Granted, the measurements by Stimler et al. (2010) were not made on plants exposed to elevated CO<sub>2</sub> for an extended period of time. However, the data from Stimler et al. (2010) indicate that uptake of COS may not be inhibited by competition with CO<sub>2</sub> under conditions of elevated CO<sub>2</sub>. Even though the plants used by Stimler et al. (2010) were not exposed to elevated CO<sub>2</sub> before measurements were made, the paper should be cited and the results should be discussed in relation to the competitive inhibition hypothesis proposed by the authors.

One of the central arguments that follows from the main conclusion (i.e. COS uptake declines as ambient atmospheric CO<sub>2</sub> increases) is that the tropospheric COS mole fraction should increase, potentially leading to a higher flux of COS to the stratosphere, where COS is thought to be the principal contributor to the stratospheric sulfate aerosol layer during volcanically quiescent periods. The authors indicate that this would lead to increased shortwave radiation reflection by the stratospheric sulfate aerosol layer, thereby counteracting the anthropogenic enhancement of the atmospheric greenhouse effect. However, the authors do not discuss the enhancement of the greenhouse effect that would be caused by an increase in the tropospheric mole fraction of COS, as COS is a strong greenhouse gas. This topic is dealt with in a recent paper by Bruhl et al. (2011), cited by the authors (now published in final form, 2012). The relevance of results from Bruhl et al. (2012) in relation to the work being done by the authors should be discussed.

The usefulness of comparisons of measurements from the two different CO<sub>2</sub> treatments are subject to question because the time periods when the measurements were made do not overlap (Table 1), or only overlap for a few days, as was the case for two of the data sets collected for *Quercus ilex*. Thus, there are two variables in the

C420

experiment: ambient atmospheric CO<sub>2</sub> and time. What evidence is there to suggest that differences between CO<sub>2</sub> treatments are really a CO<sub>2</sub> effect and not a seasonal effect? Aside from CO<sub>2</sub> deposition velocities following the expected trend of lower deposition velocity under elevated CO<sub>2</sub> (Table 3), it seems there is little evidence to rule out a possible influence of time of year.

The conclusions as stated in the final section of the paper are much too strong and/or incorrect given the experimental data. The *Fagus sylvatica* COS deposition velocity under elevated CO<sub>2</sub> was not statistically significantly different from the deposition velocity under ambient CO<sub>2</sub> (Table 4). The *Quercus ilex* COS deposition velocity under elevated CO<sub>2</sub> was only statistically significantly different than the deposition velocity under ambient CO<sub>2</sub> for two out three time periods (Table 4). Additionally, there was a statistically significant shift of the *Quercus ilex* compensation point in only two out of three time periods (Table 6), but not the same two time periods as the significant difference in COS deposition velocity. Also, none of the carbonic anhydrase activities, for either species during any of the time periods, were statistically significantly different. Thus, the enzymatic capacity was not decreased, contrary to what is reported in the conclusion section. Additionally, as discussed in section 3.3, the linear regression analysis only indicated acclimation to higher CO<sub>2</sub> in only one case (*Quercus ilex* in 1999), yet acclimation under elevated CO<sub>2</sub> is one of the main topics of discussion in the conclusions section. The conclusions section of this paper should be rewritten to reflect the actual results and corresponding statistics provided in the data tables, and it should be stated that these measured results only weakly support the proposed hypotheses (given in the introduction) for *Quercus ilex*, but not *Fagus sylvatica*.

Given the tenuous results of COS uptake differences under the two different CO<sub>2</sub> treatments, and the fact that the results were from only two tree species, the global impact calculations presented in section 3.4 are little more than simulations of a conjecture in relation to the possibility of reduced COS uptake under conditions of elevated atmospheric CO<sub>2</sub>.

C421

Specific Comments:

The word adaption (which implies genetic change) should be changed to acclimation (which implies phenotypical changes within a given genotype) throughout the paper.

Carbonic anhydrase activity was measured with some sort of pH-based method, as indicated by the heading to Table 5, but no detail for this method was found in the materials and methods section.

Methods section in general – not all equipment detail is provided – some instruments and manufacturers are specified and others are not

2125 line 27 - 2126 line 2 – these alternative pathways are interesting but their relevance to this manuscript is weak

2126 line 6: – the enzymatic role is likely not obvious to everyone, reword

2126 line 7: “well-established” is better here than “well-reported”

2126: units of ppm are not concentrations (mole/volume), they are mole fractions (mol/mol)

2128 line 1: “Measurements . . . were”

2128 line 2: “to deal with. . .” – sentence ends in an awkward way

Table 9 “best estimate” is better than “best guess”

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