

Responses to co-editor on “Effects of ozone-vegetation coupling on surface ozone air quality via biogeochemical and meteorological feedbacks” by M. Sadiq, A. P. K. Tai, D. Lombardozzi and M. Val Martin. (MS No.: acp-2016-642)

Our point-by-point responses are provided below. Comments are *italicized*, our new/modified text is highlighted in **bold**, and highlighted in **blue** in the manuscript.

I have read in detail once more again the revised ms as well as the reviews, including the one public comment and your response to these two reviews and comment. Overall, it seems that you have tackled well the various raised comments and that the revisions you have included in the manuscript properly addresses some of the main criticism. My own criticism is that there are a number of occasions where the actual models response cannot be well explained leaving you to hypothesize what might be the explaining mechanisms behind that response, e.g., changes in chemistry over the EU, a stronger response in transpiration compared to the Lombardozzi et al. (2015) study, the reduced N-resource limitation. This is also further confirmed in your response to, especially some of the comments raised by reviewer #2, e.g., the link between CUO and surface ozone concentrations and the explanation for the different responses in isoprene in the various regions. It calls according to me for a more detailed analysis for specific sites, e.g., with 1-D model approaches of the similar system so that you can really nail down the various explanations for all these mechanisms. That would, however, be enough material for another paper and appreciate this paper providing a strong motivation to indeed further pursue such further in-depth process understanding studies also for those regions where these interactions and resulting feedbacks mechanisms seem to be most relevant. In this process of reading over the comments, your response and the resulting revisions, I still came across a couple of editors' comments that I would like you to address for my final decision.

You could consider to include the reference by Super et al., 2015JG002996, Cumulative ozone effect on canopy stomatal resistance and the impact on boundary layer dynamics and CO₂ assimilation at the diurnal scale: A case study for grassland in the Netherlands, J. Geophys. Res. Biogeosci., <http://dx.doi.org/10.1002/2015JG002996>, after the statement on the impact of entrainment on ozone, lines 91, 92

(L91-92) Added as suggested:

“... but may enhance entrainment, which either increases or decreases surface ozone depending on the vertical ozone profile (**Super et al., 2015**); and higher temperature that ...”

Line 179: “multiplied with..”

(L179) Revised.

Line 184: “Therefore, online ozone-vegetation coupling and feedback are included”, I would propose here to state that the “previously discussed feedbacks are mostly included” also since you might not cover yet all the potential feedbacks (like ozone deposition impact on NO_x exchange).

(L187) Revised as suggested.

Line 220: this finding that the ozone impact in the simulations with an interactive calculation of ozone concentrations and using prescribed ozone is an interesting one since it already suggests that the overall impact of the coupling is not that large, resulting in substantially differences in ozone, or is it more a coincidence dependent on what prescribed ozone climatology you used?

As pointed out by editor, overall impact of the coupling on CUO is not that large, even with substantially higher ozone level. This can be understood using Eq. (3), which shows that CUO depends both on surface ozone concentration and stomatal resistance, such that the effect of higher ozone concentration is partially counteracted by larger stomatal resistance, leading to proportionally smaller changes in CUO and the corresponding impacts on vegetation. The text is modified to clarify this point (L221-223):

“... **leads to a similar pattern of ozone uptake by vegetation to the case using prescribed ozone due to the compensation between higher (lower) concentration and higher (lower) stomatal resistance, as reflected in Eq. (3).**”

Line 230-232; Discussing your results by comparison with other global scale climate-chemistry studies; were you aware of the Ganzeveld et al. 2010 study on the impact of land cover and land use changes on O₃ and atmospheric-chemistry climate interactions? I am mentioning this since in that study we showed that the impact of future land cover and land use changes on ozone was small also due to the role of compensating effects by inclusions of some of the same interactions as you consider in this study, e.g., changes in LAI (line 258), micro- and boundary layer meteorology and biogenic emissions, except of the ozone uptake impact.

This paper is now cited and discussed in the relevant section:

(L235-237) “**This coupling effect is smaller than the potential ozone changes driven by anthropogenic emissions (up to +30 ppbv), but it more likely reflects compensation among various pathways (e.g., Ganzeveld et al., 2010).** These simulated increases...”

(L343-345) “This coupling effect is significant in view of the 2000-2050 effects of climate and land cover changes on surface ozone (+1-10 ppbv) as found in previous work (Jacob and Winner, 2009; **Ganzeveld et al., 2010**; Tai et al., 2013), and should be considered in future air quality projection studies.”

Line 280: “biogenic isoprene (or VOC?) emissions”; I think that you should stress that you consider only changes in biogenic emissions of isoprene and did not consider changes in biogenic N-emissions. By the way, so far you mainly referred to isoprene emissions but do changes in biogenic emissions also include changes in terpene and other BVOCs emissions? This is not completely clear. You should possibly stress this.

Revised accordingly in Sec. 4 (L286):

“A comparison between Fig. 6(a) and Fig. 3(a) shows that the changes in **biogenic VOC emissions** account for ~0-60% of the ozone increases over Europe, North America and China, ...”

Changes in isoprene emission, among all biogenic VOCs, are dominant in affecting tropospheric ozone concentrations. Isoprene emission alone comprises roughly half of the total emission of biogenic VOCs (Guenther et al., 2012) and is considered to be the most important biogenic hydrocarbon in atmospheric chemistry due to its highly reactive nature (L48-51). However, in our simulations, the embedded MEGAN module simulates emission fluxes of 19 different categories of biogenic compounds, including isoprene, monoterpenes and others. Although not dominantly, terpene reactions are linked to ozone. Therefore, turning off MEGAN and comparing between [PHT+COND_nM] and CTR_nM have indeed isolated the impact of total biogenic VOC emissions.

Line 287: “which provided the sensitivity of simulated ozone to perturbed dry deposition only without the complication of stomatal coupling with hydrometeorology”. This modification doesn’t read well. I would propose to rephrase this to: “which provides an approximated sensitivity of simulated ozone to perturbed dry deposition velocity only to separate this impact from that due to changes in hydrometeorology associated with changes in stomatal conductance, e.g., changes in mixing layer depth”. This also links this statement better to the follow-up discussion/statement on mixed layer depth changes. I also think you should carefully refer to the term dry deposition being or the dry deposition velocity or dry deposition flux. Since you are referring in this part of the analysis to figure 5a, it should all read “dry deposition velocity”.

Revised accordingly (L291-293, L301).

Line 291-292: “...feedback whereas over western Europe, where the lower chemical loss rate following reduced transpired water might have further enhanced the positive feedback”; see my suggestion for sentence change but also want to remark that such a statement about what might have caused a stronger response does not give a strong impression about the level of understanding of the actual explaining mechanisms. Are not there not additional diagnostics to indeed confirm that it is change in the chemical loss rate that explains the simulated response?

Revised as suggested (L296-298). As for additional diagnostics, unfortunately we did not archive the chemical loss rates as outputs in our 20-year simulations, but we were able to arrive at these explanations due to an examination of spatial patterns of changes in the associated hydrometeorological variables and an understanding of the interactive processes represented and not represented in CESM.

Line 314; here I read for the first time a reference to the role of soil moisture changes whereas I think this is an extremely essential component in this coupling mechanism. Has there indeed been hardly any change in soil moisture, has it not been focus of your study or was it already included in more detail in the other preceding studies? I am inquiring, also since I am informed that for example the soil moisture signal might be an essential component of the Mediterranean pollution response (the ecosystems being much less sensitive to ozone when stomatal closure due to reduced soil moisture is considered, see studies by Emberson et al. on the DOSE modelling and EMEP).

Changes in soil moisture and precipitation are shown below and also included in the supplement. We find increases in soil moisture (along with increases in precipitation) in many places where GPP and LAI also (counterintuitively) increase with ozone-vegetation coupling, hence we conclude that, by the understanding of model mechanisms, more favorable hydrometeorological conditions have contributed to this vegetation growth in these regions. This effect appears to dominate over the higher sensitivity of vegetation to ozone at higher soil moisture (as Emberson et al. suggested), which is also accounted for in the model though only weakly. Changes in soil moisture and precipitation tend to reinforce each other, however, leading to strong coupling both in the real world and in the model that is particularly difficult to isolate. We decide not to lengthen the discussion on this since soil moisture has not been our research focus, due to its less comprehensive parameterization in CLM and weaker linkage to photosynthetic processes (Bonan et al., 2014), and because as we have shown LAI changes have only a minor feedback effect on ozone. We now include the plots for soil moisture and precipitation changes in the supplement to clarify these points:

(L317-320) “... and relaxation of resource limitation as nutrients and water become less limiting upon lower photosynthetic and evaporative demands, as well as favorable hydrometeorological changes following ozone exposure (enhanced soil moisture and precipitation as shown in Fig. S5).”

(L400-402) “... hydrometeorological feedbacks introduce strong nonlinearity in the interactions between atmospheric chemistry, **soil moisture** and vegetation that is more difficult to isolate.”

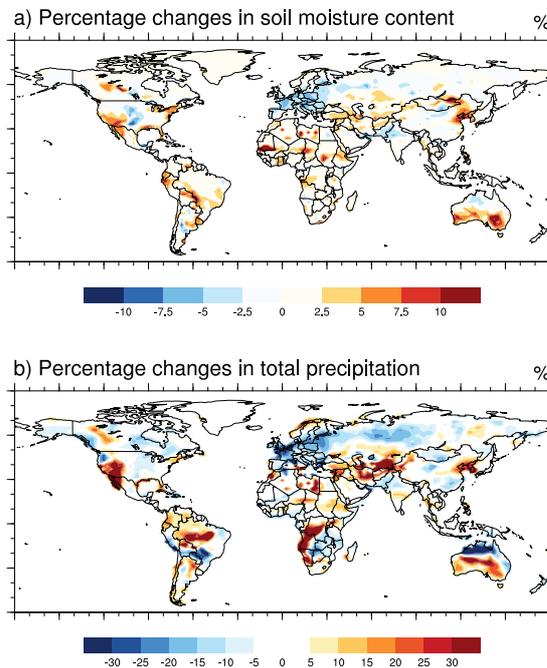


Figure S5. Percentage changes in (a) volumetric soil water content and (b) total precipitation rate in the [PHT+COND] case, where both photosynthetic rate and stomatal conductance are modified by ozone uptake, relative to the control case (CTR).

Regarding some of the revised figures, also the ones you included in the supplement; there is one striking one, Figure S3 which shows the relative changes in the O_3 dry deposition velocity, indicating differences of -20% over Greenland. I am aware that there it is reflecting a relative difference for a very small absolute difference in V_d , which is mainly reflecting the snow-ice uptake rate and the turbulent and diffusive transport to the surface. You would guess that the snow-ice uptake would not change in your model (which is the case in most models) and that the change would reflect a small change in V_d in turbulent and diffusive transport. This is possibly also reflected in quite large differences in surface temperatures and heat fluxes expressed in Figure S4. Anyhow, so showing these relative differences including this “surprising” signal in such areas calls for a better explanation of the mechanism, or you might prefer to only show the relative differences for those regions where this is some significant vegetation cover. This would serve to avoid getting too much distracted from the main point you want to make and requiring a more detailed explanation of such additional, complicated system responses.

Fig. S3 (a) and S4 (b,c) are updated accordingly by only showing differences where there is vegetation cover with annual mean LAI above 0.01, as shown below also:

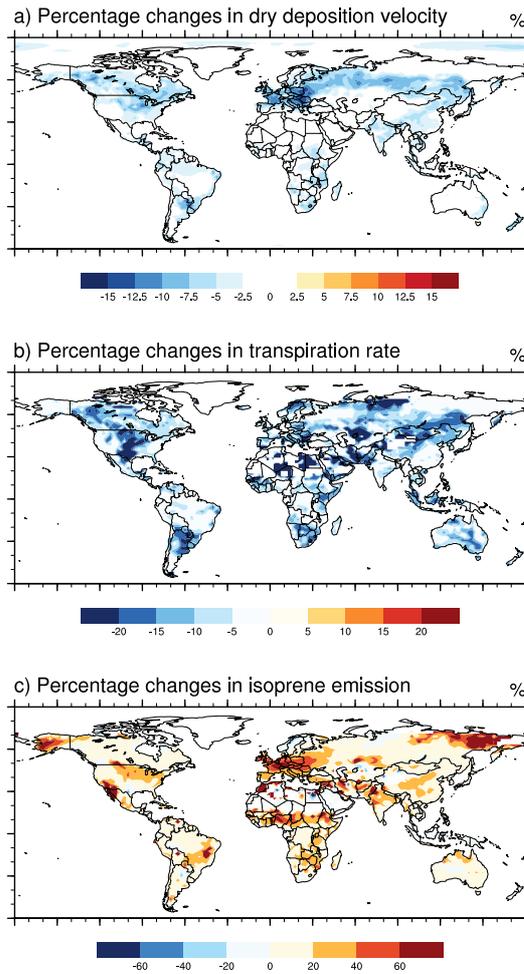


Figure S3. Percentage changes in (a) dry deposition velocity, (b) transpiration rate and (c) isoprene emission in the [PHT+COND] case, where both photosynthetic rate and stomatal conductance are modified by ozone uptake, relative to the control case (CTR).

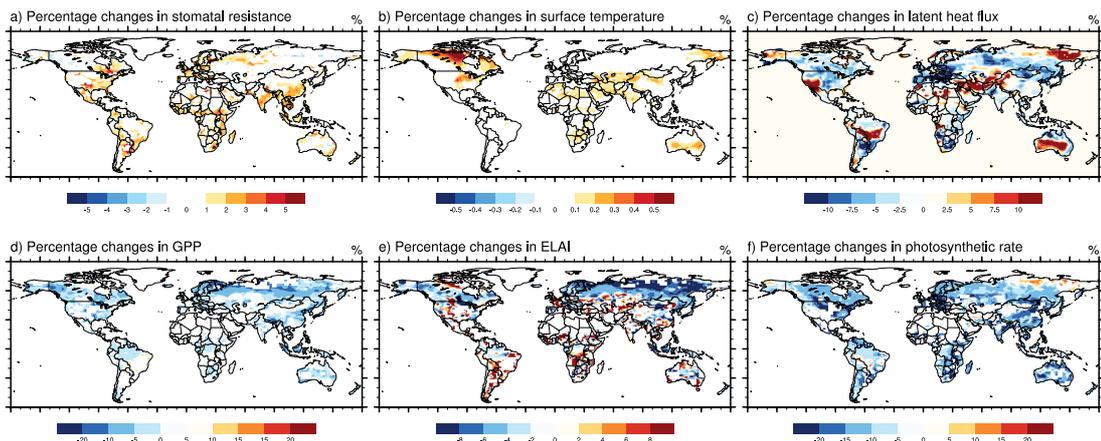


Figure S4. Percentage changes in (a) stomatal resistance, (b) surface temperature, (c) latent heat flux, (d) gross primary production (GPP), (e) effective leaf area index (ELAI) and (f) photosynthetic rate in the [PHT+COND] case, where both photosynthetic rate and stomatal conductance are modified by ozone uptake, relative to the control case (CTR).

I also checked the revised Figures 6 which were changed to tackle one of the comments by one of the reviewers about statistical significance resulting in a recommendation of major revision. The dots should indicate the regions where the changes are significant but have to say that these came out poorly and were only visible by a very careful check of the figures. I recommend that you still try to find a more optimal way to indeed indicate the significance levels, e.g., only showing the contours for those locations where the signal is indeed significant.

Fig. 3 and Fig. 6 are revised (shown below also) with different plotting parameters to make markers more visible:

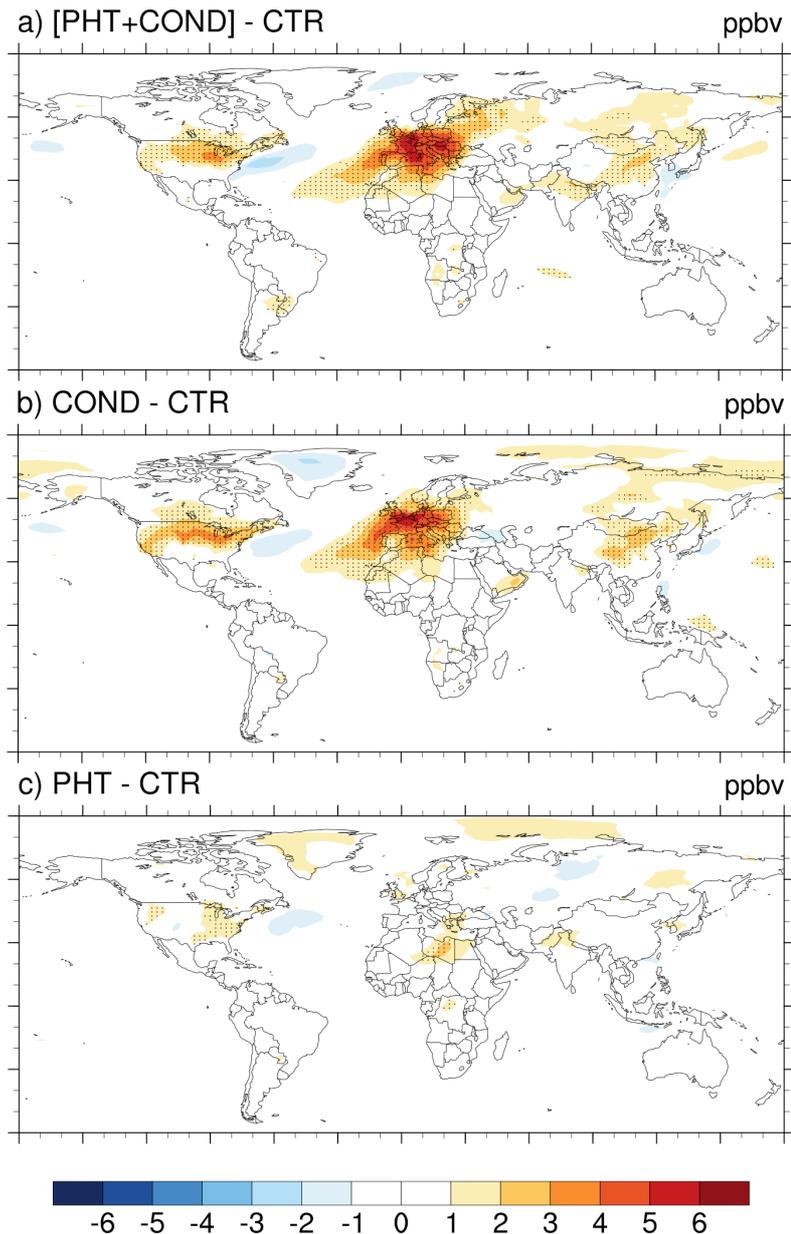


Figure 3. Changes in summertime surface ozone concentrations in different simulations: (a) the case where both photosynthetic rate and stomatal conductance are modified by ozone uptake; (b) modified photosynthetic rate only; and (c) modified stomatal conductance only, all relative to the control case (CTR). Stippling with dots indicates significant changes at 90% confidence from Student's t test.

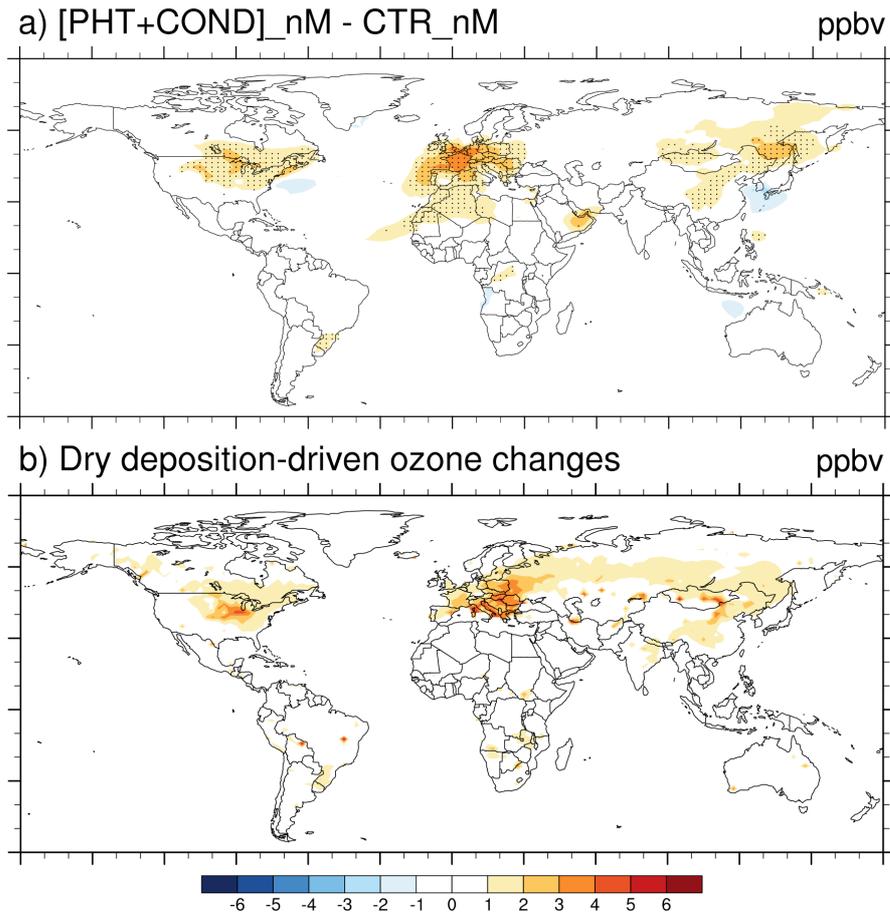


Figure 6. Changes in surface ozone concentration in: (a) the case where both photosynthesis and stomatal conductance are modified by ozone uptake, but with prescribed isoprene emission from the original control case (CTR) by turning off MEGAN (stippling with dots indicates significant changes at 90% confidence from Student's t test); and (b) theoretical changes calculated by multiplying our simulated dry deposition changes with the change in ozone concentration per unit change in dry deposition from Val Martin et al. (2014), which did not include ozone damage on vegetation.

Regarding this specific comment by reviewer #1:

p8 l275-277: As mentioned above in connection with figure 6, I don't quite understand the line of argument here. Why can the ozone impact on its own deposition not be diagnosed directly from the model simulations? Please explain further.

Your response: See our response to point (3) above.

I was also wondering about this point. I have already provided some additional comments on how you tackled this comment #3 and where I understand why these diagnostics might be complicated.

However, I don't think it is computationally complicated. It points to me that you have not available in your output diagnostics the separate terms that all at the end determine the actual change in V_d which are $R_a + R_b + R_{surf}$. If you would have those terms written out you could directly calculate the actual resulting changes in V_d and the diagnose the contributions by the micro-met terms relative to the surface uptake term. Somehow, the currently provided explanation does not seem to be very satisfactory and would still like to invite you to see if this can be more optimally tackled.

As we examine the reviewer's question again, "Why can the ozone impact on its own deposition not be diagnosed directly from the model simulations?" if this question is interpreted literally,

the answer is in the original manuscript because the impact of ozone exposure on dry deposition velocity (v_d) is already shown in Fig. 5(a), which is the actual changes in v_d resulting from online ozone-vegetation coupling. Such changes as we diagnose from the model outputs are attributable mostly to changes in stomatal conductance, and to a lesser extent to subsequent hydrometeorological changes that further modify boundary-layer (g_b) and aerodynamic conductances (g_a). Unfortunately, we did not archive g_b and g_a in our model diagnostics to make an exact attribution, but this does not alter the key conclusion that ozone-induced changes in dry deposition velocity can create major feedback effect on ozone concentration itself.

However, from the lines the reviewer was referring to (now L288-291), we infer that what the reviewer has meant to ask was why the changes in ozone concentration due to changes in v_d alone could not be diagnosed directly by a sensitivity simulation in the same way as that due to changes in biogenic VOC emissions alone. This we believe has been addressed, namely, that due to restrictions in the model structure, there is no simple way to do a sensitivity simulation where v_d is kept unaffected by ozone as in the control case while other pathways are affected. It is computationally challenging to prescribe fixed v_d to feed into CLM in the same way biogenic emissions can be prescribed. We therefore resort to make use of the sensitivity of ozone concentration to v_d changes from another set of simulations with essentially the same model configuration.

I hope that this last round of editors comment will help in arriving at a final version of the ms that optimally describes the main outcome of your study and that will further trigger research with a focus on this theme of ozone deposition impacts, interactions and feedback mechanisms.

The manuscript has been revised, as detailed above, in response to the comments and requests from the editor. Please also note that the model description (Eq. (3) mainly) has been updated to correct a formerly overlooked typographical mistake and a more detailed description has been added to explain the equation better (L161-173).

We thank the editor again for his insightful suggestions that have helped improve the quality of the manuscript.

References

Bonan, G.B., Williams, M., Fisher, R.A., and Oleson, K.W.: Modeling stomatal conductance in the earth system: linking leaf water-use efficiency and water transport along the soil–plant–atmosphere continuum, *Geosci Model Dev*, 7(5), 2193-2222, doi:10.5194/gmd-7-2193-2014, 2014.

Guenther, A. B., Jiang, X., Heald, C. L., Sakulyanontvittaya, T., Duhl, T., Emmons, L. K., and Wang, X.: The Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1): an extended and updated framework for modeling biogenic emissions, *Geosci Model Dev*, 5, 1471-1492, doi:10.5194/gmd-5-1471-2012, 2012.