

Response to comments of both reviewers #1 and #2:

I. Response to RC1 (Violetta Velikova)

Thanks for the nice review and the questions raised. Since the details about the investigation conducted at the plant nursery in Freiburg in 2018 were rather scarce, we will add the following information to the present manuscript in line 170ff: „The distance between the edges of control and stressed group were approximately 10 m with 1 m distance between individual tree stems. Water was added by a watering pot by moderate flows to each of the trees in order to control the amount of water and to minimize the effects on neighbouring trees.“. The question concerning the soil water status of the seedlings is important. We have made two approaches, i.e. a) measuring the pre-dawn water potential using a Scholander bomb as well as b) approximating the local soil water content by reference measurements of the German Weather Service at the same soil conditions ca. 600 m in distance. The latter method used measurements down to 75 cm soil depth. We considered the water status of the seedlings as key parameter and transferred the derived values to soil water availability (SWA) taking the soil composition into account. By doing so, we hope to get close to the real SWA conditions and marked this by a notable errorbar, derived from the different approaches.

Regarding the minor comments:

L. 47: We'll replace Kelvin by degree Celsius as the units are only shifted by 273.15 K and slope is not affected.

L. 158-162: The following references will be added to „ROS detoxification/reduction of BVOCs“: Niinemets et al. (2014), Parveen et al. (2018), Piechowiak et al. (2019) and Yalcinkaya et al. (2019).

L. 203: Will be done.

References:

- Niinemetts, Ü., Fares, S., Harley, P. and Jardine, K.J. (2014). Bidirectional exchange of biogenic volatiles with vegetation: emission sources, reactions, breakdown and deposition. *Plant Cell Environ.* **37**, 1790–1809, doi: 10.1111/pce.12322. *(already in the manuscript)*
- Parveen, S., Harun-Ur-Rashid, M., Inafuku, M., Iwasaki, H., and Oku, H. (2018). Molecular regulatory mechanism of isoprene emission under short-term drought stress in the tropical tree *Ficus septica*. *Tree Phys.* **39**, 440-453, doi: 10.1093/treephys/tpy123.
- Piechowiak, T., and Balawejder, M. (2019). Impact of ozonation process on the level of selected oxidative stress markers in raspberries stored at room temperature. *Food Chem.* **298**, 125093, doi: 10.1016/j.foodchem.2019.125093
- Yalcinkaya, T., Uzilday, B., Ozgur, R., Turkan, I., and Mano, J. (2019). Lipid peroxidation-derived reactive carbonyl species (RCS): Their interaction with ROS and cellular redox during environmental stresses. *Environ. Exp. Bot.* **165**, 139-149, doi: 10.1016/j.envexpbot.2019.06.004.

First of all, thanks to the reviewer for the valuable comments made. We will address the individual issues step by step as listed in the review:

General comments

Indeed the studies used were digitalized by extracting the datasets from the individual publications as far as we could access it either personally or by web. It's a very good idea to put all data into a datasheet for the community to use it henceforth. This will be done as suggested as an extra supplementary dataset.

It's correct that most of the datasets were obtained from Europe, as a lot of studies on drought effects were conducted especially in the Mediterranean and the project is focussed on this area. However, looking for other studies with sufficient parameter measurements available for comparison (e.g. including specific soil params) were difficult to find. Potentially further information were obtained by the experimentalists i.e. authors of studies but were not accessible within the article or supplementary datasets. An inclusion of further studies is welcome anytime. As suggested the different studies will be classified by area of investigation (Mediterranean, temperate) in the overview table 2, where it is supposed to fit best. But one of the outcomes was actually that the plant response curves for individual species were physically i.e. molecular property based, but not species dependent. The plant species behaviour was reflected in the selection of the different intensities of processes and may be adapted in this way to the local environmental conditions such as typical wetness, temperature and oxidation strength. However, we do not have sufficient data to clearly prove this hypothesis in a statistically significant way.

The term 'biological growth curve' is based on a variety of fit curves to match best with observations. The fitting equation was derived from 'biological growth' behaviour, i.e. stresses to occur similar to the growth process. Those are supposed to be linked directly to processes involved in the plant status and establishing a metabolic balance. While some compounds such as isoprene may be less hindered by the stomata closure, its emission may be an appropriate adaptation measure of the plant to different conditions (e.g. energy fluxes). We will consider your comments by adding an explanatory sentence at the introduction of the fitting curve term in L. 150.

Regarding section 2.3, i.e. the fitting curves and the named driving forces: We do not have sufficient understanding of and information about the detailed physiological processes acting during production, emission and stress response in total. This is supposed to be gained in further experiments in the community. Thus, the present manuscript is only focussed on figuring out the plant response to drought stress for different compound classes. A review covering all the named aspects would probably be very extensive and fill a book, which would be nice to have. The controlling factor of emissions can be added as follows: „Isoprene is known to be emitted close to production (Guenther et al., 1995, 2006, 2012) and is therefore controlled by the production process itself as well as by diffusion gradients between plant and atmosphere. The larger sized monoterpenes at least partially require passage through the stomata, as their size does not necessarily permit diffusion directly through the plants *cuticula* (Sharkey et al., 1991). Therefore monoterpene emissions are controlled by the stomata opening to a larger extent than isoprene and less by the production. Parts of the produced terpenes may be stored if not emitted for later usage. This is even more evident for sesquiterpenes with 50% extra mass than the monoterpenes. Additionally the volatility is substantially reduced with increasing mass counteracting emission. Thus, the transport is the key for understanding the emission rate, if sufficient amounts of terpenes are available.“

Thanks for naming the issue of 'forest' air. This will be corrected to 'ambient' air, since the plant nursery is not placed in the forest but at the campus site. Figure S1 is added to the supplementary material, since it demonstrates a large variety of the behaviour of different VOCs and no lumping of functional

groups seems possible. It is not part of the paper, but to support following the analysis pathway. There is a tendency of an overall link predominantly by emission. Ozone and OH reactivity values were calculated based on reactivities as listed in Table (S1) and the individual emission rates. Thus, no further measurements like nitrogen oxides etc was needed. Temperature measurements have been taken from the nearby German weather service station named in L. 172f and temperature will be added to this paragraph. The BVOC measurements have been described in L. 176ff and methods have been referred to. While a further discussion would cause the manuscript to increase notably the measurements are used here to demonstrate only the behaviour of the plants emission reactivity at declining water status in addition to the match with other observations with respect to water availability and terpene emission rates.

Specific comments:

L. 16: OK.

L. 19: Correct. „On the contrary...“ will be changed to „On the contrary to declining soil water availability,...“

L. 26: Correct. To prevent any misunderstanding the term „and methane (CH₄)“ will be deleted.

L. 137: It should have stated as „last“ or „final barrier“ before being released to the ambient. The ‚e‘ will be deleted.

L. 189: Correct. „emissions of any vertical mixing“ will be changed to „effects of any vertical mixing“.

L. 191: Correct. It is tens not tenths and the second part of the sentence „...and the area of emission can be easily traced back within the next kilometers in distance (unpublished data)“ will be deleted.

L. 230: Done.

L. 263/331. Thanks. This will be done to prevent misunderstanding and misuse.

L. 293: True. This will be included in L. 292 as „SQT and diterpene (DT) (....)“.

L. 310: Thanks.

L. 344: We stated „tendency“ as this was found in the Rombach study and aimed not to generalize in total. We will put the references next to tendency to indicate that this is based on a limited amount of data, to be investigated further. The comment on the doi is absolutely correct and the doi will be removed from the thesis reference.

Fig. 5: The figure caption will be improved. Total monoterpene emission fluxes are plotted on the left, the corresponding of sesquiterpenes on the right. The individual emission fluxes will make the figure more complex but will be added in different colours. To display all 89 compounds identified would overload the plot.

Figs. 6 and 7: These results are gained by applying the derived parameterisation for isoprene and monoterpene emission rates for a) European beech (*Fagus sylvatica*, lower plots) and for all species present in the Black Forest according to their basal emission rate. This will be clarified in the caption adding „...using the parameterisations of Guenther et al. (2012) and the SWC dependency derived in this study.“ In Fig. 6.

Fig. 7: Will be done by adding a shaded box for the time of enforced drought.

We will ask for language editing.

Thanks a lot for the detailed suggestions made.

References:

Guenther, A., Hewitt, C. N., Erickson, D., Fall, R., Geron, C., Graedel, T., Harley, P., Klinger, L., Lerdau, M., McKay, W. A., Pierces, T., Scholes, B., Steinbrecher, R., Tallamraju, R., Taylor, J., and Zimmerman, P. (1995). A global model of natural volatile organic compound emissions. *J. Geophys. Res.*, **100**, 8873–8892, doi: 10.1029/94JD02950.

Guenther, A., Karl, T., Harley, P., Wiedinmyer, C., Palmer, P. I., and Geron, C. (2006). Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature). *Atmos. Chem. Phys.*, **6**, 3181-3210, doi: 10.5194/acp-6-3181-505 2006.

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) (2012). An extended and updated framework for modeling biogenic emissions. *Geosci. Model Dev.*, **5**, 1471–1492, doi; 10.5194/gmd-5-1471-2012.

Rombach, J. (2018). *Einfluss reduzierter Wasserverfügbarkeit auf die VOC-Emissionen bei Buchen*, Albert-Ludwigs Universität, Freiburg, 13, 14, doi: 10.1186/s13007-017-0166-6.

Sharkey, T.D., Holland, E.A., and Mooney, H.A. (1991). *Trace gas emissions by plants*. Academic Press, London, UK.