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Interactive comment on "Emission of monoterpenes from European beech (*Fagus sylvatica* L.) as a function of light and temperature" *by* T. Dindorf et al.

T. Dindorf et al.

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Author's response to anonymous referees 1 and 3:

First of all we would like to thank the anonymous referees for their comments on the manuscript. The main concern of these referees is related to the representativeness of the experimental data presented, as well as the utilisation of branch enclosure measurements and our experimental setup in general. Since we do not agree with the fundamental critic of the referees, we would like to contribute to the discussion and add some annotations to the referee's comments:

1. Absence of replica measurements/ representativeness:

To our knowledge, these are the first measurements demonstrating the light dependence of monoterpene emission from Fagus sylvatica under natural conditions. The

light dependence of monoterpene emission from beech is an important finding, which was clearly demonstrated by the field measurements including a darkening experiment under daytime conditions causing an immediate cease of monoterpene emission. Emission recovered immediately after the branch was illuminated again by sunlight. Demonstrating the dominating role of light for the monoterpene emission and uncovering the significance of the standard emission factor of this deciduous tree species in the European budget may initiate research activities at other locations, in other seasons and during longer time scales in future studies. We agree with the referees that confirmation by true replication on other specimens will help to reduce the uncertainty of our measurements, specifically concerning the variation in the standard emission capacity. Regarding our measurement research strategy, it is worthwhile to point out that this was done under environmental conditions in the natural habitat with an old adult beech tree, not with a young one and not on a single leaf basis. We regarded it as highly important to get a consistent track of emission rates under natural conditions over several days within several weeks on the same tree and branch in order to avoid any discrepancy of tree-to-tree or branch-to-branch variability. Furthermore, we assume that a branch with a high number of leaves reflects per se a natural mixture of heterogeneous single leaves. We do not think that this branch or this specimen was a special case. We have to emphasise (also in a revised version of the manuscript) that emission factors measured in 2002 were confirmed by simultaneous measurements by means of another enclosure system applied on a different branch of Fagus sylvatica. The latter work focused on the seasonal development of monoterpene emission from European beech (Holzke et al., in preparation) at the same site. As already outlined in the manuscript, the representativeness of the 2003 branch enclosure measurements (in terms of light-dependence) was additionally confirmed by contemporaneous flux measurements (eddy covariance) and a top-down model approach including fetch calculations and species distribution at this site by Spirig et al. (2004), calculating monoterpene standard emission factors for European beech of 7 μg g-1 h-1. For details of this approach we refer to the respective literature. The revised version of the

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manuscript by Spirig et al. (2005) included more detailed information and therefore monoterpene emission factors were calculated to 10 μg g-1 h-1. These emission rates even exceeded the emission rates obtained by the enclosures by a factor of 2. We agree with the referee # 3 that the top-down scaling problematic is an issue that has to be carefully discussed in a revised version of the manuscript. However, the emission factors observed during our field campaign in 2003 are in close agreement with two other publications reporting on enclosure measurements under artificial laboratory conditions (Kahl et al. 1999; Schuh et al. 1997). In this way, the classification of European beech being a low emitter by the referees seems no longer acceptable to us. With an observed standard emission factor of 13 μg g-1 h-1 (observed in 2002) only 11 plant species among a total of $>$100 with specified monoterpene emission rates exhibited higher rates than European beech (see Kesselmeier and Staudt 1999). Taking into account the lower standard emission rates of 4 μg g-1 h-1 in 2003 , European beech is still among the top third of all monoterpene emitters screened so far. With both of our numbers being validated by independent methods we think it significant that the emission of the same individual branch over two years covers such a wide range. Moreover, we do think that the effects of temperature (as measured in the field) and drought (only derived from precipitation data, see Figure 9) are of importance when discussing the emission variability of European beech (see comment by referee # 3). However, the predawn water potential was not recorded during the present study. We will include this information in a revised version of the manuscript.

2. Technical details of the branch enclosure system applied:

We do not regard this cuvette system to be poorly controlled. Avoiding a too high load by leaves and adapting the flow to the amount of biomass, CO2 concentration, as well as the temperature adsorption, we were able to realize cuvette conditions which followed the ambient conditions quite closely. The focus of our field studies was to learn about the diel courses of emission patterns of sunlit leaves representative of environmental conditions in its natural habitat as close as possible to ambient

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(although we realize that identical conditions are never matched). Recently, the growth environment/history became more and more focus of research of the variability of the emission capacity, which is why we tried to avoid artificial change of environmental conditions in the enclosures as far as possible. Self-shading of leaves in a branch enclosure system is an issue, as compared to single leaf enclosures. An advantage of the utilisation of dynamic branch enclosures rather than leaf enclosures is, that here an average of several heterogeneous leaves with different physiological characteristics (e.g. photosynthesis, stomatal conductance and also monoterpene emission) can be obtained under non-artificial conditions- e.g. not influencing the natural movement, growth, and/or the direction of the leaves. Moreover, since the application of a branch enclosure is less invasive than leaf cuvettes, branch enclosures were preferred for this study (see Marler and Mickelbart 1992). However, shading of leaves in a branch enclosure can not be avoided and we agree with the referees that this systematic error may in part explain the observed scattering of our data. A comment on this detail will be added in a revised version of the manuscript. Photosynthesis, transpiration, and stomatal conductance were monitored during the complete measurement period by application of an infrared gas analyser (Model Li7000 [Licor, USA]) and allowed a constant examination of plant's physiological status. The environmental parameters of light and temperature were measured by standard sensors. As outlined by the referees, we will include a detailed description of the measurement system in the experimental part of the manuscript file, additional to the literature already cited. PAR measurement were made by application of two (in 2002, one in 2003) PAR sensors (Model SB 190, [Licor, USA]) that were placed on two sides directly above the enclosure (outside of the sample cuvette, and not shading the leaves, the FEP Teflon foil of the enclosures was proven to be fully light permeable in the spectral range of 300-900 nm). Air temperatures were measured by ultra fine Teflon covered thermocouples (0.005 inch;, Chromel-Constantan, [Omega, UK]) outside but next to the sample cuvette (ambient measurements, in addition to the ambient temperature measurements performed at the meteorological tower, presented in Figure 9), in the reference cuvette and in the

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sample cuvette. Leaf temperatures were measured by application of the same type of thermocouples. Data reported in the manuscript represent the average of 4 leaf temperatures measured simultaneously on the upper- and lower sides of 2 representative leaves inside of the branch enclosure. Therefore temperature and PAR measurements were obtained continuously by application of several sensors and were not measured on spot only as outlined by referee # 3. Daily variations between both PAR sensors (in 2002, see above) were typically $<$10% of the average value. However, small differences in light intensity as observed between both years (decrease of maximum values by some percent in 2003) might be an effect of not exactly placing the sensors at the same place as outlined by referee # 3. We will include this information in a revised version of the manuscript. Variations of leaf temperatures were typically $<$5% for the upper and lower side of the leaves, respectively. We will include this information in a revised version of the manuscript.

3. Midday depression and hysteresis:

We agree with the referee # 1 that midday depression is not easily detectable. However, we found a noticeable reversible decrease of stomatal conductance and photosynthesis in both years (see Figure 2 for photosynthesis, data of stomatal conductance are not shown) in relation to temperatures increasing over the day. By means of this midday depression the plant is able to keep the water loss rate unaffected. Therefore the transpiration rate was not affected by the latter regulation. The daily correlation of photosynthesis and stomatal conductance to PAR (presented in Figure 7) shows a hysteresis structure typical for midday depression which is in accordance with Tuzet at al. (2003). For a better understanding, it should be mentioned in the figure legend and in the relevant chapter that the daily maximum values shown here were derived during the morning hours, while lower values were measured during the afternoon. We will include some comments on this in a revised version of the manuscript. Unfortunately, the available time resolution of monoterpene measurements (1 h) makes it impossible to relate a midday depression to monoterpene emissions from the direct daily progression

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as shown in Figure 2. However, in correlation to PAR we found a decrease of monoterpene emission during the hottest hours of the day and discussed such behaviour (hysteresis) in agreement to photosynthesis as to be related to a midday depression, owing to a restriction of photosynthetic CO2 fixation. A simple reduction of emissions rates due to stomatal closures seems to be excluded as an internal concentration increase of monoterpenes will lead to a steeper concentration gradient which causes compensation of stomatal closure (see,e.g Loreto et al. 1996; Niinemets and Reichstein 2003, Niinemets et al., 2004). Monoterpene emission from this beech tree generally followed the progression as shown in Figure 8 (top graph) and since this pattern was measured several times during our 2003 campaign (also found for tropical tree species during our previous campaigns in Amazonia, data not published) we assume this progression to be the "normal" case. Unfortunately, monoterpene measurements performed during the other days in 2002 did not show an adequate fast increase of radiation intensities in the early morning and thus do not allow to observe this "normal" progression as observed in 2003. We will add this detail in a revised version of the manuscript. As shown by Figure 8 (bottom graph) the daily progression was different for the measurement day # 7 in 2002 and in correlation to leaf temperature a similar hysteresis can be observed for the latter day as well. We will include this information in a revised manuscript. Since the above described pattern was observed for one day in 2002 only, it's beyond our possibilities to drive general conclusions for the year 2002 and we agree with the referees that the data basis for these measurements is rather small. Moreover, we are not able to show that the daily progression in correlation to PAR as observed in 2003 is valid for the year 2002 as well, and our conclusions depend on the assumption that the general pattern observed in 2003 and other campaigns was valid for 2002. Furthermore we were not able to prove that sabinene emission was reduced by down regulation of RUBISCO and photosynthesis. Taking this into account, we can only speculate about the reasons leading to the different daily progressions. We agree with both referees to shorten this chapter to avoid misunderstanding and speculations.

4. Monoterpene mixing ratio of ambient air:

In chapter 3.2. we reported the maximum monoterpene mixing ratios that were measured in the years of 2002 and 2003. Maximum mixing ratios measured during both measurement campaigns were 1.8 ppb (2002) and 1.1 ppb (2003) and thus differed by factor of 1.6. We do not think that a factor of 1.6 represents a small difference. However, we agree with referee # 3 that interpretation of ambient mixing ratios demands a careful discussion. Variation of ambient monoterpene concentrations ranged between zero and the respective maximum values for both years and normally showed a pronounced diurnal course. We will include the ambient mixing ratios of monoterpene compounds in Figure 2 for the respective measurement days in a revised version of the manuscript. However, meteorological parameters like wind direction were not measured directly at this tower site and our conclusions that can be derived from these measurements are limited. We state that the trend of variation in emission capacity is reflected by the change of respective ambient mixing ratios. However to avoid misinterpretation, we will discard the sentence “This result is indicative for the strong influence of beech trees on atmospheric gases in the vicinity of the tower site”.

5. Decomposition of sabinene on GC-MS adsorbent traps:

In a laboratory study it was observed that sabinene was decomposed on the GC-MS adsorbent traps as a function of storage time. Sabinene decomposition reached a saturation effect after 7 days. At this time 45% of the sabinene was decomposed to other monoterpenes. All samples that were analysed during the ECHO campaigns were stored longer than 7 days before GC-MS analysis was performed. Thus, we multiplied sabinene concentrations measured by GC-MS with the latter factor. We agree with referee # 3, that the uncertainty induced by this correction should be reported in the manuscript. However, the overall conclusion that sabinene is the major monoterpene compound emitted by European beech will not be changed, since daytime sabinene emissions exceeded that of other monoterpene compounds even if no correction factor was applied. Of course also this result was confirmed by contemporaneous measurements with a different enclosure system on different branches of Fagus sylvatica

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(Holzke et al., in preparation).

6. Implications for the European Budget:

We do not regard the influence of light on monoterpene emission (that was shown by laboratory studies conducted by Schuh et al. 1997 and during the present study) as a weak fact as outlined by referee # 3. Moreover, standard emission factors reported for 2003 are in good agreement to Schuh et al. (1997) and other studies. Standard emission factors obtained for 2002 are in good agreement to measurements performed by Holzke et al. (in preparation). We do not agree with referee # 3 that the standard emission factor reported is a "generally accepted mean emission factor". Monoterpene emission factors that were assigned to temperate forest ecosystems (see Olson 1992; Guenther et al. 1995) account to 0.9 μg g-1 h-1 only. Regarding the measured monoterpene emission factors for Fagus sylvatica (4 μg g-1 h-1 in 2003 or 13 μg g-1 h-1 in 2002) we do regard an increase of a factor of 4 to 14 that is induced by a single tree species of this ecosystem type as highly important. Our intention of presenting several graphs was to give an overview of the different steps of the model scenarios and not to overemphasise the chapter as outlined by referee # 3. Therefore, we will discard Figure 10 since this figure presents the data used by the default model only. As outlined by referee # 3 we will include a table instead of this figure in a revised version of the manuscript file. With our model approach we want to stress the potential importance of a single species (in this case the third-most distributed in Europe) on the monoterpene budget calculations.

7. Minor comments by referee # 1:

Minor comments will be corrected in a revised manuscript. Regarding the measurements shown in Figure 2, the uptake of carbon is shown as a negative-, emission of carbon or water as a positive value. Photosynthesis shown in all other diagrams was multiplied by a factor -1. We will include this information in the manuscript. However, regarding the figures we would prefer to use the term "CO2 exchange" instead of "CO2

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drawdown" since this is a term commonly used.

8. Technical corrections by referee # 3:

Technical corrections # 3, 5, 6, 7, and 9 will be included in a revised version of the manuscript. Other corrections seem to be not reasonable to us or are beyond our possibilities:

a. Emission rates are reported in units of μg g-1 h-1 in the text of the manuscript file as this is a commonly used unit for terpene emission rates (e.g. see the BVOC database of ACD, IGAC, NCAR, NSF, EPA, and Lancaster University http://www.acd.ucar.edu:8080/voc/vocIndex.jsp or see the review by Kesselmeier and Staudt 1999). However, we already provided the conversion factor and specified the unit μmol m-2 s-1 in Table 2 to allow better comparison to other studies.

b. The reference "Holzke et al. in preparation" is not cited in the reference list and appears as a footnote only. This is consistent with the general instructions of manuscript preparation published by "Biogeoscience" (see paragraph "References" at http://www.copernicus.org/EGU/bg/guidelines_for_manuscript_and_article.html#chapter3). Furthermore, we are confident that the cited manuscript will be submitted at the end of the evaluation process of the actual manuscript.

c. We would prefer not to add the reference "Schnitzler et al. (1997)" on page 139, line 27, since our general experiment is described at this paragraph. We will include the reference at another paragraph in a revised version of the manuscript.

c. Unfortunately it is not possible to report the same units for monoterpene emission rates given by König et al. (1995) and Tollsten and Müller (1996) who reported emission rates on a dry weight basis, and Schuh et al. (1997) and Kahl et al. (1999) who reported emission rates on a leaf area basis, since these authors gave no conversion factors (specific leaf area). However, comparison to the measurements of the present study is still possible and is discussed in the respective paragraph, since we reported

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conversion factors (see Table 1) as well as emission rates on leaf area basis and on dry weight basis (see Table 2).

Literature cited:

Guenther, A., Hewitt, C.N., Erickson, D., Fall, R., Geron, C., Graedel, T., Harley, P., Klinger, L., Lerdau, M., Mckay, W.A., Pierce, T., Scholes, B., Steinbrecher, R., Tallamraju, R., Taylor, J., and Zimmerman, P.: A Global-Model of Natural Volatile Organic-Compound Emissions, Journal of Geophysical Research-Atmospheres, 100, 8873-8892, 1995.

Kahl, J., Hoffmann, T., and Klockow, D.: Differentiation between de novo synthesized and constitutively released terpenoids from Fagus sylvatica, Phytochemistry, 51, 383-388, 1999.

Kesselmeier, J. and Staudt, M.: Biogenic volatile organic compounds (VOC): An overview on emission, physiology and ecology, Journal of Atmospheric Chemistry, 33, 23-88, 1999.

König, G., Brunda, M., Puxbaum, H., Hewitt, C.N., Duckham, S.C., and Rudolph, J.: Relative Contribution of Oxygenated Hydrocarbons to the Total Biogenic Voc Emissions of Selected Mid-European Agricultural and Natural Plant-Species, Atmospheric Environment, 29, 861-874, 1995.

Loreto, F., Ciccioli, P., Cecinato, A., Brancaleoni, E., Frattoni, M., and Tricoli, D.: Influence of environmental factors and air composition on the emission of alpha-pinene from Quercus ilex leaves, Plant Physiology, 110, 267-275, 1996.

Marler, T.E. and Mickelbart, M.V.: Repeated Mechanical-Stress from Leaf Cuvette Influences Leaf Gas-Exchange, Hortscience, 27, 432-434, 1992.

Niinemets, U., and Reichstein M.: Controls on the emission of plant volatiles through stomata: Differential sensitivity of emission rates to stomatal closure explained. Journal of Geophysical Research, 108, D7, 4208, doi:10.1029/2002JD002620, 2003.

Niinemets, U., Loreto, F., and Reichstein M.: Physiological and physicochemical controls on foliar volatile organic compound emissions. Trends in Plant Science, 9, 4, 2004.

Olson, J.: World ecosystems (WE1.4): Digital raster data on a 10 minute geographic 1080‘ 2160 grid square. In Global Ecosystem Database, Version 1.0: Disc A Ed. NOAA National Geophysical Data Center, Boulder, 1992.

Schuh, G., Heiden, A.C., Hoffmann, T., Kahl, J., Rockel, P., Rudolph, J., and Wildt, J.: Emissions of volatile organic compounds from sunflower and beech: Dependence on temperature and light intensity, Journal of Atmospheric Chemistry, 27, 291-318, 1997.

Spirig, C., Neftel, A., Ammann, C., Dommen, J., Grabmer, W., Thielmann, A., Schaub, A., Beauchamp, J., Wisthaler, A., and Hansel, A.: Flux measurements of biogenic VOCs during ECHO 2003, Atmospheric Chemistry and Physics, Discussions, 4, 6603-6643, 2004.

Spirig, C., Neftel, A., Ammann, C., Dommen, J., Grabmer, W., Thielmann, A., Schaub, A., Beauchamp, J., Wisthaler, A., and Hansel, A.: Eddy covariance flux measurements of biogenic VOCs during ECHO 2003 using proton transfer reaction mass spectrometry, Atmospheric Chemistry and Physics, 5, 465-481, 2005.

Tollsten, L. and Müller, P.M.: Volatile organic compounds emitted from beech leaves, Phytochemistry, 43, 759-762, 1996.

Tuzet, A., Perrier, A., and Leuning, R.: A coupled model of stomatal conductance, photosynthesis and transpiration, Plant Cell and Environment, 26, 1097-1116, 2003.

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