Answers to the reviewer

Steffen M. Noe

January 22, 2014

What is actually measured?

The reviewer states " *...but the dominant part is thought to origin from specific storages...*" and refers to Lerdau and Gray (2003) [3] where is stated:

"*The light-independent emissions have proven the easiest to understand from both biochemical and functional perspectives.*"

These two statements use the terms "*is thought*" and "*proven easiest to understand*" because it is no clear statement about the origin of the emission possible. However, during the 90ties the mainstream believed, because of obvious and empirical evidence, that the majority of monoterpene emitted from species bearing specific storage organs (oil glands, resin ducts) is originated from those. Recent developments in measurement equipment such as use of PTRMS technique together with isotopic $^{13}CO_2$ as carbon source led to a more complicated picture with higher heterogeneity between the possible origins (recently formed and previously stored), see eg. Ghirardo *et al.* (2010). There will be always a mixture between both which is modulated by biological activities and physicochemical properties reacting to environmental changes.

We do not want to discuss this topic in greater detail here but we want to note what is typically measured if we speak about monoterpene emissions.

- Generally BVOC flux measurements are conducted by enclosure techniques where a leaf or branch are covered by some enclosure.
- By comparison of the difference between an air flow into and out-of that enclosure related to the surface area of the emitting tissue a flux over that surface is obtained. If related to mass, we obtain an emission rate.

In a simple sentence, we actually measure *a volume with an emitting tissue*. The tissue itself remain a black-box and we only can postulate from additional measurements (monoterpene content, physicochemical properties, tissue specific structural parameters, etc) how the share between possible origins of monoterpenes might compose an actual emission flux.

Storage in parameterized models

The previous section has put the focus to the way how the majority of parameterization data was and still is obtained. Given such data, we can now model them by using a pure temperature dependent or a mixed scheme where both, light and temperature dependent equations are used. These are typically offered by Guenther algorithm and MEGAN. As these rely on the parameterization data that already includes the mixture of monoterpene from different origin any model that manages to describe such emissions inherently includes stored and recently fixed sources of monoterpenes.

Let us assume we chose a temperature dependent algorithm, this will also work to some extent for light and temperature dependent emitters. This is because the solar radiation is the major energy input to the earth system and thus temperature and light are not independent from each other. On certain time scales, they will be rather well correlated and form a very similar environmental driver. We can now postulate that everything that is emitted by that driving factor may come from the storage pool but we will not be able to prove it.

Assumed we have a light and temperature dependent algorithm chosen, we may be able to capture different timescales in the emission dynamics and we can postulate that some portions may be originated from the storages and others from recently fixed carbon. Now, a nighttime separation as pure storage emissions can be formulated. But still, we can not prove finally how the daytime emissions are split at a certain moment.

To summarize, even a wrong assumption may lead to a reasonable well fitting algorithm that capture and describe storage bound emissions because the data already include that information.

Therefore, we grade the reviewers statement "*However, the models applied seem to consider only the light-dependent emission.*" as wrong.

Storage in the SIM-BIM model

The reviewer states: "*...the SIM-BIM model because this is developed for direct emissions only...*". This statement is only true for the original implementation of

the SIM-BIM model by Zimmer and coworkers (2000) [5] and subsequent developments done by Grote and coworkers (2006) [1].

We used a further developed equation system (see Appendix A) which could be named the "Tartu version" of SIM-BIM. One major feature that has been included are diffusion terms appearing in equations A11 and A13. These diffusion terms allow to control the amount of the monoterpene pool (in case of A13) that can be emitted. If the diffusion is as fast as the production we could postulate to describe a *light dependent* emission and if the diffusion is slower as the production it fills the monoterpene pool, in other words, a storage pool, and we describe a mixed emission scheme. If there is a big enough pool, ceasing the production will still lead to emission and we now can postulate that this is originated from *light independent* storage pools within the tissue.

Concluding remarks

Taking all together, we can not follow the reviewers grading "*If it is true that the emission from storages has been neglected, I would see it as a major flaw in the overall exercise.*" that the model exercise is flawed.

- By using parameterization data as described before, we have always a portion of monoterpenes originated from storage compartments within the tissue, even though these might not be explicitly formulated or the "wrong" driving factor was chosen.
- Inclusion of diffusion terms into the "Tartu SIM-BIM" version lead to dynamic build up of storage pools and emission from mixed origin.

Notes on birch emissions

From the work done in Järvselja, birch is a dominant monoterpene emitter (*Betula pendula* and *Betula pubescens*) see Noe *et al.* (2011) [4] as example. Hakola *et al.* (2001) [2] report also that birch species emit monoterpenes but few isoprene. Therefore, birch will contribute overall to the monoterpene concentration in boreal forests even though it is a deciduous tree.

References

[1] R. Grote, S. Mayrhofer, R.J. Fischbach, R. Steinbrecher, M. Staudt, and J-P. Schnitzler. Process-based modelling of isoprenoid emissions from evergreen leaves of *Quercus ilex* (L.). *Atmospheric Environment*, 40:S152–S165, 2006.

[2] Hannele Hakola, Tuomas Laurila, Virpi Lindfors, Heidi Hellén, Anca Gaman, and Janne Rinne. Variation of the VOC emission rates of birch species during the growing season. *Boreal Environment Research*, 6:237–249, 2001.

[3] M. Lerdau and D. Gray. Ecology and evolution of light dependent and light-independent phytogenic volatile organic carbon. *New Phytologist*, 157:199–211, 2003.

[4] Steffen M. Noe, Veljo Kimmel, Katja Hüve, Lucian Copolovici, Miguel Portillo-Estrada, Ülle Püttsepp, Kalev Jõgiste, Ülo Niinemets, Lukas Hörtnagl, and Georg Wohlfahrt. Ecosystem-scale biosphere-atmosphere interactions of a hemiboreal mixed forest stand at Järvselja, Estonia. *Forest Ecology and Management*, 262(2):71–81, 2011.

[5] W. Zimmer, N. Brüggemann, S. Emeis, C. Giersch, A. Lehning, R. Steinbrecher, and J-P. Schnitzler. Process-based modelling of isoprene emission by oak leaves. *Plant, Cell and Environment*, 23:585–595, 2000.