**Atmospheric
Chemistry
and Physics
Discussions**

Interactive comment on "Tethered balloon measurements of biogenic volatile organic compounds at a Boreal forest site" *by* C. Spirig et al.

Anonymous Referee #2

Received and published: 27 November 2003

The paper is very interesting, of high quality, well structured, excellently written, it reads easily and the arguments are easy to follow. I recommend it for publication in ACP after a some minor changes (and (suggested) additions, see below).

General comments.

The paper by Spirig et al. presents interesting results of VOC measurements from a tethered balloon platform performed during the OSOA field campaign (August 2001, Hyytiälä, Finland). Two flux evaluation techniques ("Mixed Layer Gradient" and (MLG) and "Mixed Box" (MB)) have been applied to concentration data of terpenes and iso-prene, and to corresponding meteorological data. Results of both techniques are compared, supported by photochemical modelling of VOC degradation and proper footprint

Full Screen / Esc

Print Version

Interactive Discussion

Discussion Paper

© EGU 2003

analysis. Special attention is called to the section of error analysis and extended footprint discussion. The paper closes with some solid ground estimates and discussion of the production of condensable vapours, which indeed can not explain the formation of secondary aerosol particles (as observed by others at Hyytiälä), but represent very valuable material for further discussion in this field of research.

Specific comments.

When making use of data (obtained at a tethered balloon platform) for the application of MLG and MB methods, one of the most important quantites (besides precisely measured concentrations) is the height of the mixed layer z_i (or the depth of the CBL, as called by the authors on page 6). This parameter displays in eqs. (1), (2), (4), (5), and (6). However, there is only one single (vague) statement in the entire paper, how z_i was determined (page 10 last para, 1st sentence : "Temperature and humidity profiles obtained from flights with continuous sampling during ascent and descent were used to determine boundary layer depths"). Given the well known problems in determining mixed layer heights from profiles of temperature and humidity, the authors should include (at least) a paragraph how they have done it (e.g. setting certain criteria on the vertical derivates of potential (virtual potential) temperature and/or relative humidity (or specific or absolute humidity). Within this context it is also pretty important to know

(a) what was the nature of temperature and humidity sensors ? Especially temporal stability of light weight relative humidity sensors may be limited for high resolution measurements needed for z_i-criteria on the vertical derivative of (relative) humidity

(b) how temperature and humidity sensors have been calibrated ? The statement on page 4 (last para, 2nd sentence), namely "Pressure and temperature readings of the different sampler packages were referred daily against the official pressure measurement in the SMEAR II station" seems not to be sufficient (by the way, it is not easy to understand how a temperature reading can be referenced against a pressure measurement)

Full Screen / Esc

Print Version

Interactive Discussion

Discussion Paper

(c) what was the precision of temperature and humidity measurements of the different sensors ? This will be important in the context of the required precision to establish proper z_i-criteria on the vertical derivative of these quantities

(d) what was the temporal resolution of temperature and humidity sensors ? The e.g. 90%-time of each sensor determines the "simultaneousness" of temperature/humidity measurements as well as their proper attribution to a specific height (again, an important issue how to determine z_i). In this context, what is the meaning of "vertical resolution usually 1-2 m" (page 4, para 3) ?

page 4, para 1. Were the "ML integrating" balloon flights and the "Gradient" flights performed successively or simultaneously ? This may be important when comparing MGL and MB flux results.

page 4, para 2. What was the nature and purpose of "conditioning" the cartridges in the week before the experiment ?

page 4, para 4. "VOC sampling packages are attached to the tether line at heights of about 120 m, 250 m, and 500-600 m". If one considers Figure 3, one may ask where the shown data in the altitude ranges 150-200 m, 300-400 m and $>$ 650 m are coming from?

page 5, para 2, last sentence. This formulation may be a bit confusing. It is not clear, at least to the reviewer, what was the accuracy of isoprene and monoterpene measurements and what was the precision of isoprene and monoterpene measurements for the Hyytiälä experiment. Please clarify, since this is an very important issue for the error discussion (section 3.2.1, first para). In this context, especially considering the "small VOC levels encountered at Hyytiälä" (see page 14, para 1), the reader is certainly missing a clear statement about the detection limits (definition, number) for the (individual) isoprene and monoterpene measurements.

page 7, para 1. Finally, how exactly was the entrainment velocity we determined from

the mixed layer growth ? (from corresponding temperature/humidity profiles ?)

page 8, para 2. "entrainment flux" rather than "entrainment".

page 9. eq. (7). Is X_g equivalent (identical) with CF of eq. (3) ?

page 10. last para. "Part of the flights did not exceed the top of the boundary (mixed?) layerĚ" Within the context of the (important) determination of CF (eq.3) and X_g (eq.7), it may be interesting to be informed how many ballon flights really made their way to levels (well) above z_i.

page 11. last para. Despite the presented argumentation and the information of Table 1, it is still not clear, at least not to the reviewer, which values (ppt) have finally been used for the CF 's (eq.3) and X_g 's (eq.7) of each individual flux calculation.

page 13. para 1+2. 12 profiles of 32 individual VOC samples have been used. This suggests that flux evaluation of most profiles is indeed based on a 3-level gradient (but some only on a 2-level gradient ?) For a proper judgement of the MLG equation fitting procedure : what about to show one (or more) typical examples (in an additional figure) ?

page 13. para 3. last line. "average" : average over which data ensemble ?

page 14. para 1. Without any statement about actual detection limits (definition, number) and even with the already given statement about accuracy and precision of the concentration measurements (page5, para 2, last sentence) it is hard to see, that the "uncertainty of terpene measurements at these levels" should be (only) ś 30%. Please clarify. It will be important for the rest of the (very valuable) argumentation and discussion of section 3.2.1.

page 14. para 2. last 3 sentences. "potential stratification effects due to the lake", and "Ěas stratification has the tendency to shift the footprint further upwind" : the non-meteorologists among the ACP readership may have (severe) problems to understand what this will meanĚ.

page 16. para 1 and Figure 6, respectively. The term vertical mixing velocity is intruduced in the legend of Fig. 6. What is it, and how was it measured/inferred/deduced ? Please clarify.

page 16. para 2. 2nd sentence. "Ěsuitable for flux calcualtions". Obviously, some rejection criteria have been applied to the total data set of "integrating" tethered balloon concentration measurements ?

page 16. para 4. 2nd sentence : Ě.. hard to follow.

page 17. para 1. last sentence. "and measurements were takenĚ" rather than "measurements were taken" ?

page 18. para 1. first line. "Ě..footprint areas of 200 to 500 km2" : considering the further discussion within this section (and within the rest of the paper), it would be interesting to get (even a rough) idea, what the composition of the landscape around Hyytiälä beyond the (limited) area shown Fig. 1 is.

page 18. para 3, last 2 sentences. It is suggested to re-consider these statements, especially when considering the interquartile ranges of the modelled biogenic VOC emissions for areas of 1 ` 1 km and for 10 ` 10 km : it is not easy to see the significance of the difference between 1 ` 1 km and 10 ` 10 km results.

page19. para 2. last sentence. Since it is essential for the further argumentation : it should be made (more than) clear which numbers (in units of molecules cm-3s-1) refer (a) to the calculations of the authors, which numbers refer to the estimate/measurements of others, and which number is 10% of what. The present formulation seems to be a bit confusing Ě

page 20. para 1. last sentence. in this context, the reviewer likes to draw the author's attention to a very recent article by Bonn & Moortgat (2003) (Sesquiterpene ozonolysis: Origin of atmospheric new particle formation from biogenic hydrocarbons, J.Geophys.Res., 30 (11), 1585, doi: 10.1029/2003GL017000, 2003)

Figure 5. Despite the fact, that the meaning of boxes and bars is explained in the text of the manuscript, it should also be mentioned in the figure legend.

References. "Madronich, S., and Calvert, J. G.: Permutation reactions of organic peroxy radicals in the troposphere, J. Geophys. Res., 95, 5697-5715, 1990" is not cited in the text of the manuscript.

———————————————

Interactive comment on Atmos. Chem. Phys. Discuss., 3, 5357, 2003.