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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Boundary layer concentrations and landscape scale emissions of volatile organic compounds in early spring" *by* S. Haapanala et al.

Anonymous Referee #1

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General comments

Haapanala and co-authors present aircraft and balloon based VOC measurements over boreal forests and estimate landscape scale VOC surface emissions. While such efforts have already been made earlier over various ecosystems (including boreal forests), the new element in this work is the first measurement of fluxes on this scale over boreal forests during spring time - the period with most frequent observations of new particle formation events in the area.
The topic is of interest for the ACP readership, but the authors are encouraged to revise their contribution by adding more details on the methods and significantly extending the

discussion of the results. Unless the QUEST special issue claims a lower than the normal ACP quality standard, the paper requires improvement before publication.

Specific comments

Introduction

The authors argue that information on the distribution of VOCs throughout the boundary layer is important in the context of the particle formation events. This point should be extended and clarified - it sets the stage for a later discussion of the VOC results and their significance in relation to the particle formation (a discussion that unfortunately is not included in the manuscript yet, but should be added!).

2. Materials and methods

Figure 1: Indicate the location of SMEAR II on the map. Is Table 1 not giving the landuse information for the whole area shown in Fig.1. (km^2 instead of m^2)?

P.10570, line 17: Please give information on the location of Juupajoki relative to SMEAR II.

P.10571, line 10: Does this reference only describe the inlet system or the whole VOC analysis? It is also not clear whether the VOC sampling here is the same as the one described later for the sampling on the balloon. If not, a quick description of the VOC sampling on aircraft should be added.

line 27f: Some evidence for a complete trapping with this setup should be presented, e.g. by including typical ratios of the concentrations in the second cartridge relative to those in the first cartridge.

General comment to the description of the VOC analysis systems: A description of the accuracy and detection limits is required. Maybe adding this information in the form of a table would be helpful. As it stands now, it is hard to oversee the complexity of different sampling/analytical devices. The information presented later in table 2 is insufficient.

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P. 10572, line 11 f: The assumptions should not only be mentioned but also be commented and discussed. For example, under what conditions is the assumption of balanced emissions and sinks reasonable? Is the 20% underestimate due to entrainment negligence a general fact? It would be particularly important to include statements on what points are of particular interest when applying this method for conditions at this time of the year (implications of reduced mixing, possibly different lifetimes due to lower oxidant levels, etc.).

Lines 24f: A good estimate of the chemical losses is obviously the key for surface flux estimates with this method. Therefore, more details about the model used for calculating the OH concentrations must be included. Was the model driven by observations? Uncertainty estimates for the OH concentrations? Also, it is not clear from the description what O_3 concentrations were used for the calculation of chemical losses, whether they were derived from measurements (what type, surface/airplane/balloon) or modelled.

3. Results and discussion

P. 10573, line 11: It is not clear what the averages represent. Please indicate whether they are means over several flights(indicate number!) or even flights on various days.
lines 13f: Please clarify the reasoning for the very low isoprene concentrations. It would be interesting whether light/temperature conditions alone are sufficient to explain these low concentrations or if an additional seasonal variation of the emission factors is necessary.
line 27: When comparing to the data of Hakola et al., temperature conditions during 2001 should be mentioned.
P10573/4: I am missing a summarizing, concluding remark after this paragraph. The MT concentrations measured here during spring time were of similar magnitude and of the same distribution. Is this according to/contradicting to expectations (based on knowledge about seasonal variations of emissions)?
Lines 9f: Here and in several other instances it is not evident whether the profiles origi-

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nate from airplane or balloon measurements. For interpreting the shape of the profiles, it may be important to distinguish between the results of different platforms. Also, details about the flight patterns of the light aircraft need to be included somewhere.
Lines 15f: I agree that the uncertainty derived from a 2h sampling must be a lower bound estimate for the airborne measurements. Still, some more reasoning about the use of the uncertainty of a surface measurement for qualifying the airborne measurments should be added. This type of questions hopefully become obsolete if the description of the sampling/analytical systems in the M+M section is clarified and extended.

P.10575. Flux estimates: It is not clear what kind of flux values are presented here. Is it the range of fluxes derived from individual profiles or from certain averages? An estimate of the uncertainties of the fluxes should be presented. The authors mention analytical uncertainties, but the boundary layer heights and the modelled OH concentrations in particular seem to be at least as important for the overall uncertainty of the estimates.
More details on the modelled VOC emissions should be given. It is not clear whether a seasonal dependence of emission factors was accounted for. Please clarify the discussion about landscape vs ecosystem scale emissions. The flux estimates presented here obviously represent the landscape scale, and the emission model, too, no?
If the discrepancy between modelled and estimated landscape fluxes holds even after a careful uncertainty estimate of the flux estimates, it needs to be discussed in more detail. Is there enough evidence to conclude that emission models for this area do not adequately describe the real emissions? Is it true for this season only?

Conclusions

The conclusions should be significantly extended. It now reads like a mixture of summarizing results and single non-structured interpreting statements. Despite cold weather, boundary layer concentrations of monoterpenes were not lower than during summer - this calls for a conclusion. Do the authors want to make a point that even

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though emissions are lower in spring, the resulting concentrations of (condensable) vapours might be of the same magnitude as in summer due to the reduced mixing, enhanced lifetime of the compounds?
Line 19f: It should be discussed earlier why gradient methods were not applicable in this case. It also should be discussed why the mixed box technique was still applicable - e.g. in the discussion of assumptions for this method in chapter 2. As the original point of this work is the performance of measurements in this particular season, an extended discussion (and conclusions) on whether and under what conditions these methods are still applicable, would be of high interest. The conclusion on the preference of aircraft vs. balloon platforms needs to be explained further, it is not evident from the results presented here. For example, one could argue that the more stationary balloon platform is advantageous because it has a constant footprint whereas the aircraft measurements imply a varying footprint. Again, a clear distinction of airplane and balloon results in the previous section and information on the flight pattern will help to follow this interpretation.
Finally, after mentioning the significance of biogenic VOCs on particle formation in the introduction, a statement or comment on the implication of the results presented here on this topic would be valuable.

Tables:

Table 3: Some information on the number of profiles used for these estimates should be added either in the text or in the table.

Technical corrections

There are many examples of missing articles or cases where articles should be omitted. The following list points to some of them:

P.10568, line 18, P.10569, lines 1, 12, 21, 23 P 10570, lines 23, 26 P 10571, lines 7, 12, 15, 20, 27 P. 10572: line 1 P. 10573: lines 2,3, 18, 20, 21

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Other linguistic corrections: P. 10569: line 3: "on" different scales line 8: "proved" line 10: "on" .. scale line 25: "in early spring"

P.10571, line 4: "at" seems wrong P. 10573, line 23: reactivities

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Interactive comment on Atmos. Chem. Phys. Discuss., 6, 10567, 2006.