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Atmospheric Chemistry and Physics Discussions

Interactive comment on "On the role of monoterpene chemistry in the remote continental boundary layer" *by* E. C. Browne et al.

Anonymous Referee #1

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In this study the authors have investigated the contribution and role of monoterpene derived organic nitrates to the NO_x mixing ratio and the oxidation capacity above Canadian boreal regions. In order to do so they applied the WRF-chem model to ambient observations during the ARCTAS campaign. As the campaign is focussed on the boreal region where coniferous trees are the dominant tree type monoterpene emissions outmatch the isoprene ones by far and thus control the local tropospheric chemistry. Any NO_x reactions and concentrations therefore will be notably affected by their presence and the resulting nitrates will undergo phase transfer that will alter the regional impact of nitrogen oxides.
So far the authors have done well and tackled an aspect worth investigation. There are some points worth a more detailed discussion:

1.) The approach is done using the 2 product approach by Odum and colleagues in 1997 although more advanced however reduced chemistry schemes exist (Derwent et al., 2003; Bonn et al., 2004; 2005). This will certainly impact strongly on a) the amount of SOA formed and b) on the detailed regional impact as the aerosol is no permanent sink anymore but acts as a temporal sink with a release in a later stage that influences the gas-phase chemistry again. Please comment much more on this!

2.) There are a couple of different chemical schemes on the market (RACM, STOCHEM, MCM derived ones) that display notable differences. How do these intercompare here with respect to the results shown?

3.) The authors basically state the major monoterpene species would be α- and β-pinene. This highly matters on the tree species involved and a large number of differently structured monoterpenes will be emitted especially when focusing on the whole region of Canada. This will impact on the reactivities with the ambient oxidants, i.e. ozone, OH and NO_3 in detail that are hardly matched by a single scheme used in common CTMs. I am aware of the complexity of the problem an the challenges for modelling. But this will have particular consequences on the oxidation capacity, the SOA contribution and the aerosol as well as the gas phase chemical speciation! Please consider this much more in the discussion. This certainly is difficult in a regional or global modal. However this will modify the chemistry from the emitting needle towards the boundary layer towards the free troposphere. Please consider this at least in the considerations. Please mention this at least in more detail. But let's discuss more because its an online journal that offers the possibility and the tools are available. Please consider

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this in more detail when elaborating your results although they are certainly interesting, nice and mainly correct.

4.) I actually miss the possibility of trajectory analysis when using aircraft measurements and models. That allows a very nice set-up of detailed investigations in this context. Why is this not done?

5.) The water solubility mentioned. I kindly ask to consider and to take into account the huge amount of carbon molecules of monoterpenes and the large functionalities of their oxidation products. Assuming Henry's law coefficients between 10^3 to 10^5 may be a little bit to elevated. Have there been any sensitivity studies with a range of water solubilities?

I suspect considering some of the points mentioned and the aspect risen the observations shown may be explained to a large degree especially the differences of vertical OH and NO behavior.
Last but not least please correct a list of typos with respect to the references provided:
Peeters et al. (2012): Insert full pages.
Rollins et al. (2012): Write NO_x correctly.
Thornton et al. (2000): Remove '?'.
Tunved et al. (2006): It's Mika Dal Maso as Dal Maso, M. instead of Maso, M. D..
Xie et al. (2012): Please change ACPD reference to ACP.

Citations:
Bonn, B., von Kuhlmann, R., and Lawrence, M. G. (2004), High contribution of biogenic hydroperoxides to secondary organic aerosol formation. Geophys. Res. Lett., 31, L10108, doi:10.1029/2003GL019172.
Bonn, B., von Kuhlmann, R, and Lawrence, M. G. (2005), Influence of Biogenic Secondary Organic Aerosol Formation Approaches on Atmospheric Chemistry. J. Atmos.

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Chem., 51, 235–270.
Derwent, R. G., Jenkin, M. E., Johnson, C. E., and Stevenson, D. S. (2003), The global distribution of secondary particulate matter in a 3-D Lagrangian chemistry transport model, J. Atmos. Chem., 44, 57–95.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 22297, 2013.