Interactive comment on “Simulating atmospheric composition over a South-East Asian tropical rainforest: Performance of a chemistry box model” by T. A. M. Pugh et al.

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Received and published: 10 November 2009

The paper presents a box model study of the photochemistry above the Borneo forest using the observations conducted during the OP3 field campaign to constrain these simulations and also interpret some of the observations. The originality of the work appears to be mainly the fact that it describes issues involved in tropical boundary layer chemistry for the Asian rainforest where most of the research until now has been conducted in Amazonia (and the African tropics). This area is obviously of great interest because of some of the ongoing land use changes in this region (peat land drying, palm oil plantations, etc.). The paper is well written, addresses an interesting dataset and
describes some challenges involved in closing our understanding of tropical boundary layer chemistry. However, it does not address according to me very novel issues; it is basically considering the previously identified issues on tropical forest photochemistry but then for this particular, interesting location. For example, the discussion on the issue of the overestimation of the VOC oxidation products possibly due to a misrepresentation of dry and wet deposition has been already been discussed previously, first of all by von Kuhlmann et al. (Sensitivities in global scale modeling of isoprene, von Kuhlmann et al., Atmos.Chem. Phys., 4, 1-17, 2004). We also discussed this extensively in the evaluation of the Guyana Gabriel campaign observations. (Ganzeveld et al., ACP, 2008) but also indicated that making assumptions on a more efficient removal of these products only reduced the surface/canopy layer concentrations and not the concentrations aloft. I recall that having met the first author on a workshop beginning of October that we have extensively discussed the issue on the possible importance of the residual layer chemistry for this misrepresentation of the MVK and MACR concentrations (as we also hypothesized in the Gabriel modeling paper). Having the 75m observations (still in the inversion layer?) of OP3 it would be interesting to shortly discuss in this manuscript what these observations show on nocturnal MVK/MACR mixing ratios.

By the way, more a minor comment; In the discussion on the MVK/MACR deposition process referring to the sensitivity analysis we conducted for the Gabriel campaign there are some misunderstandings. The numbers mentioned for the dry deposition velocities, 1 and 0.1 cm s⁻¹, refer to those explicitly simulated with the multi-layer canopy exchange model for O₃ and SO₂. Those for MVK and MACR end up being smaller based on the Wesely approach and we actually conducted an analysis modifying the dry deposition calculations using in one simulation (near) zero surface uptake resistances.

I also would like to comment on the link between the modeling and the observations. The presented study comes as an accompanying paper that describes in more detail
the observations. It is mentioned in the introduction of this ms in one line that the measurements used for the interpretation are collected 5m above the ground. However, later on reading the section on the soil NO emission modeling I am became quite concerned about how to link simulations of NOx exchange to observations that were apparently done at 5m height above the ground in a clearing of the forest. This extra information is quite essential in appreciating these observations and as well the model simulations. With some of the longer-lived trace gases one could possibly deal with this heterogeneity of the site but with NO/NO2 issues of different photolysis regimes, local heterogeneity in soil emissions (enhanced by the clearing? direct rainfall on the soil due to missing interception etc.) become essential. Moreover in the discussions about the possible relevance of the intensity of segregation to explain the discrepancy between the isoprene and OH concentrations a reference is made to using the observed concentrations at 5m height. Do the 5m isoprene concentrations reflect a footprint of the in canopy, understorey emission regime whereas the OH reflects full sunlight (comparable to above forest canopy) conditions? I am very much aware of the main logistical challenges in conducting these kind of measurements in tropical forests but also think that this 5m reference height measurement really poses many limitations on the presented model assessment, e.g. the role of the intensity of segregation. Consequently I suggest that these subtle features of the measurement footprint must be more properly described in the introduction. In addition, the limitation of the interpretation of the observations and potential implications for the interpretation with the model should also be more explicitly discussed.

In the introduction of the model system it is indicated that the box model uses a maximum volume up to 800m altitude also based on the LIDAR observations. In some of the other reviews/comments this feature of the analysis has also been raised and want to add to this that it would be useful to shortly discus this rather shallow tropical forest boundary layer depth by comparison with previously reported BL depths on the order of 1500m over the Amazonian rainforest (e.g. Fish et al., Eerdekens et al. ACP, 2009). According to me such a substantially shallower boundary layer could point at
less efficient turbulent mixing (smaller presence of strong down- and updrafts?) compared to the conditions seen for GABRIEL with an inferred BL depth of about 1500 m topped by a shallow cumulus layer with enhanced transport. For these conditions a very/unrealistically large intensity of segregation between OH and isoprene of 50% was proposed whereas the numbers for the required Is in this manuscript are comparable or even larger.

Finally, I especially disagree with the opening statement of the Summary and conclusions section; “This is the first modelling study of the chemistry occurring in the tropical rainforest PBL to utilise such a wide suite of ground-based measurements, particularly VOC fluxes, to inform the modeling”. It were actually already the colleagues involved in the ABLE campaigns in the 80’s (recall the Jacob and Wofsy, J. Geophys. Res., 95, 16737-16754, 1990 paper) that were already doing this pioneering work on interpreting the observations of chemical composition and exchange over tropical forest.

Minor comments;

Pp 19246; line 14 remove comma

Pp 19247; line 4, It is not only a challenge to test photochemical models; the challenge is to understand the role of photochemistry in relation to the role of surface exchanges and boundary transport processes.

Line 9; as indicated in my comments above; it would be useful to give some background information on the particular features of the Borneo tropical forest site in terms of similarities and differences with other tropical forest sites subject of previous studies.

Pp 19248, line 5; 5m above the ground; is this about the soil surface, inside the canopy or above the canopy? See also my previous comment how essential it is know where exactly these 5m observations were collected. You have to be more specific about this and it would be useful to give a short description of the canopy properties in relation to these reference heights.
Pp 19249, line 2; what has been measured; the measured photolysis rates or the measured ozone/OH?

Line 14-15; Apparently you assume that there is an nocturnal inversion layer 200m deep. Since you apply a box model system it implies that you assume that this is a well-mixed volume in direct contact with the surface. Do you calculations of deposition consider the limiting turbulent transport from the reference height of the 200m deep box (so 100m height) to the surface. And how do you consider to limited transport of the emitted species to this 100m reference height. What are your assumptions on the mixing in between the two different layers? no mixing at all?

Pp 19250; lines 1-2 and one ward; You just present the numbers on the NO and BVOC emissions without presenting any justification for the selection of these values (references). Later on some comparison of these numbers with the literature are presented but I propose to provide some short introduction of typical tropical forest NOx and BVOC emissions fluxes. In addition, with respect to the NO biogenic emissions it is essential to indicate if you refer to canopy-top emissions (of NOx) or to soil NO emissions. Since you apply one box it appears that you don’t treat the canopy interactions you apply the canopy reduction factor approach as proposed by Yienger and Levy. This feature of the model set-up is essential and should be explicitly mentioned. On the other hand, this issue about how to treat/ignore the canopy interactions comes also back to my comment about the reference height of the concentrations; 5m above the ground of the clearing.

Line 9-10; This estimate of 75% canopy reduction (expressed by a Canopy Reduction Factor of 0.25 according to Yienger and Levy, this always leads to confusion) of the soil-biogenic NOx source was based on a some very simple first order estimate of the potential role of NO2 dry deposition. The LBA (Amazonia) observations and modeling work (Ganzeveld et al., JGR, 2002) resulted in an canopy reduction factor of 0.5 that does not only reflect NO2 dry deposition but also mixing and in-canopy chemistry for tropical forest. Apparently, this doesn’t help you in explaining the difference between
the canopy top flux soil NO emission flux of 93%.

Pp 19256; In the discussion on the importance of wet deposition in removal of some of the products I wonder if you considered the fact that the convective precipitation might only over a smaller fraction of total area that your model somehow represents (e.g. the typical grid size of a large scale model).

Pp 19258, line 24; Rephrase; “Considering this extra source of OH involving the carbonyl groups resulted only in a 4% increase in the peak [OH]”

Pp 19260, line 23; “Furthermore halving the daytime zmix from 800m to 400 m” Based on the previous comments it already seems that an 800m deep daytime BL seems to be substantially smaller compared to other tropical forest sites which would exclude the possibility of even having a shallower mixing volume.

Pp 19263; “Some care however, must be taken in using the modelled MACR/MVK concentrations as support for the rate constant reduction . . . reaction”, coming back to the previous comment about MVK/MACR deposition it should be added here that use of the concentrations of these isoprene oxidation products to interpret the OH-isoprene oxidation is limited due to potentially important and not well understood role of removal by deposition as well as diurnal boundary layer dynamics, e.g. entrainment of residual layer air masses.

Overall I appreciate the manuscript as an interesting paper that further explores the issues on chemical exchange over tropical forests which as main novel component the fact that it describes the Borneo forest photochemical regime. However, as can be inferred my comments I also have some serious concerns about the interpretation of the observations, in particular the fact that short- and long lived interacting species have been measured at 5 m reference height above a clearing and not above the forest itself. I would therefore recommend the authors to consider the above listed comments and suggestions for a revised version of the manuscript.
Interactive comment on Atmos. Chem. Phys. Discuss., 9, 19243, 2009.