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Interactive comment on "FORest canopy atmosphere transfer (FORCAsT) 1.0: a 1-D model of biosphere–atmosphere chemical exchange" *by* K. Ashworth et al.

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Review of "FORest canopy atmosphere transfer (FORCAsT) 1.0: a 1-D model of biosphere-atmosphere chemical exchange" Ashworth et al., GMD (2015)

We thank Reviewer #1 for their positive, constructive and thorough review of our manuscript.

General Comments "Updates to the CACM mechanism: While the CACM mechanism is evidently not as good as RACM, it offers the advantage of coupling with an aerosol module; thus, a lot of time is spent fixing issues with CACM. While this is important, my concern is that Sect. 2.6.3 seems out of place as it is very long and involves analysis of field observations. It may be more appropriate to just give the important details in Sect. 2.6.2 and move most of this to the beginning of Sect. 4."

While CACM has not performed as well in reproducing observed concentrations under the particular conditions (low-NOx, high isoprene) experienced at UMBS in 2009, numerous case studies in different environments have shown CACM well capable of capturing tropospheric VOC chemistry. We thank the reviewer for their suggestion that Sect. 2.6.3 is out of place as it involves analysis against observations, which would be better covered in Sect. 4 with the discussion. However, this manuscript is a comprehensive description of the development of FORCAsT from its predecessors (CACHE and CACM0.0). We wanted Sect. 3 and 4 to consider FORCAsT1.0 only, as this is the "final" model version that is being made available to the community. However, we also felt that it was informative to set out in the manuscript how the model has been adapted from the forerunners. We therefore leave Sect. 2.6.3 in place, but make clearer the reasoning behind the layout of the manuscript.

"Also with regard to this subject, the titration of NOx by CACM0.0 is really striking. The lack of HO2 in this simulation is likely b/c of insufficient cycling via RO2 + NO, though this is not mentioned in the discussion. What is the primary source of NOx in the model? Advection? The accumulation of peroxy radicals as described in the last sentence of P. 5200 should shunt all of the NOx into NO2, but then where is the NO2 going? PAN? In short, it might be helpful to add a paragraph about the sources and sinks of NOx in the model."

Insufficient recycling of HO2 via the RO2+NO reaction pathway is implied in the discussions of the poor model performance under low NOx conditions in Sect. 2.6.3. However, we have now explicitly stated this in the initial evaluation of CACM0.0 against observations (p5198). Advection in FORCaST is dependent on wind-direction, with NO2 advection occurring when air masses are southerly (with high rates from SW-SE, and lower rates from E-SE and W-SW). Thus low levels of NO2 are advected to the site from 00:00-05:00 LT on the first day of the simulation period, and again from 23:00 on

Day 1 through to 03:00 LT on the second day. For the remainder of the simulation the wind was from the N-NW bringing ‘clean’ air and no NO2 advection. The only primary source of NOx after this time is soil NO emissions; some NOx is regenerated through oxidant recycling and chemical regeneration. The accumulation of peroxy radicals occurs under low-NOx conditions in CACM0.0 following the depletion of NOx. The little NO remaining does react with RO2* to regenerate NO2, but at an insufficient rate to match NO2 loss rates. The primary losses of NO2 are via direct reactions with RO2* leading to the formation of PAN and other unreactive species with atmospheric lifetimes well beyond canopy retention time. High NOx conditions only occur at UMBS as a result of transport of pollution, and advection rates are sufficiently high to overcome these losses and retain NOx in the system. A couple of sentences explicitly outlining the sources and sinks of NOx in the model have been added to Sect. 2.6.3.

“Conclusions: a little time should be spent re-iterating what insights are gained from using the resolved canopy model versus using a 0-D model or a 1-D model without all the fancy canopy widgets. Hopefully the results presented in the last few sections can back up such statements.”

We have re-iterated the insights gained from using a resolved canopy model and highlighted specific processes that our sensitivity studies have shown to be of particular importance in canopy-atmosphere exchange. The following text has been added to the conclusions: . . .“Recent laboratory experiments and field measurement campaigns have shown that we still lack understanding of many of the fundamental processes involved in the exchange of gases and particles between the forest canopy and atmospheric boundary layer: from primary emissions (e.g. Jardine et al., 2013), to VOC oxidation chemistry (e.g. Rohrer et al., 2014; Perring et al., 2013, Surratt et al., 2014; Mellouki et al, 2015), to deposition of reactive species (e.g. Nguyen et al., 2015) and the mechanisms of turbulent vertical exchange (e.g. Steiner et al., 2011). It is only through the application of 1-D canopy models such as FORCAsT, in which all of the processes are prognostically included, that we can fully investigate the relative importance of each of these processes and assess the validity of proposed mechanisms. Insights gained from the application of FORCAsT can be used to improve 3-D models of regional and global atmospheric chemistry and climate. . . . . . The sensitivity studies and chemistry mechanism updates included here have provided valuable insight into the importance of peroxy radicals and organic nitrates in VOC oxidation under low-NOx conditions, and further suggest that nighttime chemistry plays a vital role in controlling the oxidative capacity of the atmosphere within and above forest ecosystems. We find that peroxy radical self and cross reactions dominate VOC degradation under low-NOx conditions, but due to complexity are necessarily crudely modelled either by considering a small subsection of the possible permutations or by representing many peroxy radicals as a single species. This study points to the urgent need to constrain concentrations of key short-lived radical species such as organic peroxy radicals in and above forest ecosystems, and to elucidate the mechanisms and processes governing their production and loss.” . . .

Specific Comments “Sect. 2.4: Does the deposition scheme consider loss to surfaces other than leaves and soil (i.e. bark)?” Deposition occurs only in the crown space (to the leaves) and the ground surface in the current implementation of the resistance deposition model in FORCAsT1.0.

“Sect. 2.5: How is BL-FT exchange handled, and how is the height of the mixed layer determined?” Vertical exchange is driven by incoming radiation and surface heating of the foliage and ground. Energy balances are carried out for each canopy layer at every time step. This drives vertical mixing and turbulent exchange throughout the height of the model domain. FORCAsT is not influenced by synoptic conditions and the mixed layer depth is around 1km during the day, decreasing at night as turbulence decreases. The height of the mixed layer is not explicitly calculated, but can be estimated from the height at which Kh approaches zero.

“P. 5197, L.15: “The similarity of the modelled concentrations suggest that differences in terpenoid oxidation pathways between the two chemistry schemes is of little importance compared to the magnitude of emissions and efficiency of vertical turbulent transport at this site." This sentence seems a little confused. Chemically, only the lifetime of primary emissions should affect their concentration, so they shouldn't care about subsequent chemistry except via feedbacks through radical cycling. Suggest restructuring to clarify what is meant here. There is a similar sentence at the top of P. 5210." These lines have been altered to clarify the intended meaning: … "The similarity of the modelled concentrations suggest that the differences in terpenoid oxidation pathways and hence oxidant availability between the two chemistry schemes is of little importance compared to the magnitude of emissions and efficiency of vertical turbulent transport at this site." Given this clarification the sentence at the top of P. 5210 has been retained.

"P. 5202, top: What is the assumed yield of isoprene nitrates? This information is not in the supplement (or at least I couldn't find it). If it is much larger than 12%, this would explain some of the remaining issues with the mechanism described later." Isoprene nitrate yields are temperature dependent and have been conservatively estimated here. For the range of temperatures at UMBS, yields are small: ∼3-5%; this has been added to the end of the sentence at the top of p5202.

"P. 5202, L. 13: The Muller (2014) paper is a theoretical study, not a lab study (though it does re-analyze some older lab experiments)." This statement has been corrected to read: "A new theoretical study based on previous laboratory experiments has also demonstrated that …" "P. 5202, L. 24: ISOPO2 + HO2 does form carbonyls, but with a small yield (Liu et al., 2013)." ISOPOO+HO2 does not form carbonyls in either RACM or CACM. However, this statement has been modified to read: "As reactions between peroxy radicals and HO2 do not produce a significant yield of carbonyl compounds as first-generation products (e.g., Liu et al., 2013), …"

"P. 5203, 2nd paragraph: In most cases, 1st-generation isoprene hydroxyhydroperoxides should react with OH to form epoxides instead of photolyzing (the lifetime of ISOPOOH against OH reaction is a few hours). The omission of this pathway, which

has been known since 2009 (Paulot et al., 2009), seems like a major shortcoming of CACM and RACM and could be problematic for low-NOx environments." We acknowledge that the isoprene oxidation mechanism currently implemented in both CACM and RACM within FORCAsT1.0 requires updating. This is the focus of the next stage of development work, which is currently underway. The specific improvements that will be made are itemised in the Conclusions.

"P. 5210, L. 3: In Fig. 4b, the model concentrations are at the low end of observations until noon of the second day." While the concentrations are toward the low end, they do fall within the range of observed values, suggesting good model performance.

"P. 5211, last sentence: ozone has a larger reservoir and a long lifetime, so this isn't especially surprising. Likely, most of the ozone measured at UMBS was made upwind, so one would not really expect a 1-D canopy model to accurately capture ozone variability." This final sentence has been removed. We have tightened up a similar sentence on P.5197 to clarify the buffering effects we were referring to, in line with the reviewer's comments.

"P. 5212, L. 9: Is the same data shown in Figs. 4i and 4j? If so, is it really fair to compare modeled HO2 to observed HO2*?" Figs 4i and 4j show the same observations (HO2*) but Fig 4i shows modelled HO2, and Fig 4j shows modelled HO2+ISOPO2 (from ISOP+OH only). We consider it instructive to include both plots as: (a) the precise nature of the detected "HO2*" is speculative (it is thought likely that it includes ISOPO2 but it is not certain what fraction nor whether other RO2 species are also detected – e.g. Bryan et al., 2012, Fuchs et al., 2011); (b) comparison of the plots highlights the relative overproduction of ISOPO2 by the CACM mechanism (vs. RACM). Hence both panels have been retained.

"P. 5214, L. 10: Why do SOA concentrations maximize here?" The location of maximum SOA concentrations is near the top of the mixed layer and coincides with the build-up of keto-propanoic products from oxidation of MVK. The sentence referred to by the reviewer and last sentence of the same paragraph have been edited to make this point more clear.

Technical Comments “Figures: there are a lot of problems with the figures. - For all figures, the font size should be increased. I had to magnify some to 300% to read them.” Apologies, the font size has now been increased to be readable at 100%.

“- Given that only 2 days are shown, it might be better to use hour-of-day, rather than day-of-year, as the x-axis coordinate.”

The x-axis coordinates have been altered to show elapsed time since the start of the simulation period (i.e. 00:00 (EST) on 4th August 2009).

“- Fig. 2: what is the shading? What are the vertical lines?” As stated in the text, the shaded area is the model spin-up time; this time period is not included in the model evaluation or discussion but was retained in the Figures for completeness. This has been added to the caption. The vertical lines mark dawn, dusk and midnight; a statement has been added to the caption.

“- Fig. 3: Air temperature in C, not K” The axis label has been corrected, thank you for spotting that.

“- Fig. 7: Might consider adding a dashed line showing height of mixed layer.” Fig. 7 has been modified so that the y-axis shows absolute altitude rather than altitude relative to canopy height. As noted above, the model does not explicitly calculate mixed layer height, although the vertical profile of the eddy diffusivity coefficient kh provides an approximate diagnostic. As a result, we chose to not include it in the plot.

“- Fig. 8: Might look better as cumulative-area plot” Fig. 8 shows the concentrations time series, which is typical way of showing contribution of various SOA components. We have kept the figure the same.

Text: “p. 5187: A few others that didn’t make the list of canopy models are ACCESS (Saylor, 2012) and SOSAA (Zhou et al., 2014). The latter is particularly relevant as it is

another canopy model with embedded aerosol mechanisms.” This was not intended as an exhaustive list; however in line with the reviewer’s comments the reference to SOSA has been extended to include SOSAA and ACCESS has been added to the list.

“p. 5198, L. 4: It might be better to refer to the sum of MVK and MCR as “MVK+MCR.” MVK-MCR has been replaced by “MVK+MCR” throughout the manuscript as suggested.

“Sect. 4.3: The discussion of the aerosol vertical profile might be better placed in terms of height relative to mixed layer depth rather than relative to canopy height. Or, just use absolute altitude. Using multiple height coordinates is confusing.” As suggested by the reviewer, Section 4.3 has been revised so that the discussion is in terms of absolute altitude. Fig. 7 has also been modified to show absolute altitude on the y-axis.

“Supplement: There’s a lot of info here (which is good!). A table of contents would be helpful.” We have added a table of contents to the SI.

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