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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Modelling bi-directional fluxes of methanol and acetaldehyde with the FORCAsT canopy exchange model" *by* Kirsti Ashworth et al.

Anonymous Referee #1

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The authors present a canopy modeling study aimed at better understanding the mechanisms driving emissions and deposition of methanol and acetaldehyde between the forest and atmosphere. They use a 1D canopy model and test the sensitivity of the simulation (and resulting agreement with observations) to various assumptions related to emission pathways (e.g., importance of direct versus storage emissions, and degree of stomatal control over the storage emissions). These are relevant questions with implications for our ability to model these emissions and predict the sensitivity of emissions to environmental changes.

The study is well thought out and carefully executed, with a range of sensitivity tests presented. It merits publication in ACP. Some suggestions are below.

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Discussion paper

Discussion of all the sensitivity analyses tends to run on a bit to excess. The manuscript could be more effective and concise if some secondary material were moved to a supplement. The "threshold stomatal control" section is one example. Just prior to this, you state that for both methanol and acetaldehyde, stomatal control is not needed to explain the canopy-scale observations. This seems to be one of the main take-home messages and is a useful finding. But once that point is established, it doesn't seem we learn anything substantially beyond that from the section looking at the subtleties of threshold effects. I.e. the take-home from Sections 3.5 and 3.6 seems unchanged from what we had in 3.4; the bulk of this could be moved to SI with a brief summary in the main text.

7L3-6, "While we acknowledge that the magnitudes of the recorded night-time fluxes during summer 2012 may have large associated errors, we are confident in the direction of the exchange as we see variation between different species suggesting no systematic bias." Unclear what this means. Please clarify.

7L30-31, "although its reactions are limited to oxidation by OH to produce formaldehyde". That's the only relevant chemical sink in any case. Perhaps change to "its source/sink reactions. . ." to clarify that you are not including any chemical sources of methanol (e.g. peroxy radical reactions) . . . I concur that these would not be important in this context.

"FORCAsT includes a physical representation of a forest canopy, with the lowest eight model levels set as trunk space and the next ten as crown space. The ten crown space levels contain the foliage". So this neglects any shrubs etc near the ground, is this an ok assumption for Harvard Forest?

10L26-28, "rs" is capitalized in Eqn 3 but not on line 26

Please be more explicit about what assumptions are embedded in the lack of treatment of advection for methanol and acetaldehyde. Both are both sufficiently long-lived that in reality there is a substantial advective component.

13L12-14. Note that acetaldehyde only accounts for 25-40% of the bidirectional VOC flux in the Guenther 2012 scheme, depending on PFT

"The emission factors were then modified to best reconcile modelled and observed concentrations and fluxes at 29 m whilst ensuring that total canopy emissions for all simulations were within ±10%." Unclear what this means, please be more specific.

13L25-27. "The greater the contribution from storage the higher the overnight and the lower the daytime peak" Wording is a bit odd. A higher storage flux by itself doesn't necessarily decrease the daytime peak. E.g. if you kept the direct flux constant and increased the storage flux, you'd still have a daytime peak (and it would occur at higher concentration). What you're saying occurs because you've constrained the 24-h integrated canopy emissions to be the same between simulations. Right? Could say "... and the lower the diurnal amplitude".

14L21, where does the limiting nighttime value of 3000 come from?

14L22, what does the scaling factor n physically represent?

Figure 2 is hard to decipher. Font very small. Heights hard to read.

Fig 3, perhaps show the corresponding model correlations that emerge from the simulations?

16L15-32. Figure 3 seems hard to interpret due to convolution between the independent variables. For instance, you state "the data appear to show a strong linear correlation at low conductance, suggesting that at small stomatal aperture the stomata exert control over fluxes of methanol to the extent that it is observable at the canopy scale." But couldn't it equally have nothing to do with stomatal conductance, and just arise from temperature/light affecting both emissions and stomatal conductance simultaneously?

16L21-22. "A similar relationship between canopy-top methanol fluxes and concentrations is likely due to the influence of atmospheric concentrations on dry deposition

to surfaces within the canopy." Similar comment. Temp/light would increase Ch3OH fluxes which would in turn increase the concentrations. And wouldn't your point about deposition work in the opposite direction? At the lowest CH3OH concentration deposition should be lowest so that net emission is highest. But the data go in the opposite direction.

"the level at which model and measurements are compared can also affect the measured-modeled bias, an effect compounded by the instantaneous nature of the model output fluxes". But aren't the model fluxes averaged over the same 30-min intervals as the data?

None of the simulations appear to capture the nighttime concentration decline for methanol. Is this a mixing effect or does it point to some shortcoming in the model treatment of deposition?

Perhaps I missed it, but if not please clarify how atmospheric mixed layer dynamics are treated. Are those entirely prognostic within the model energy balance? How do we know how well the model captures the ML depth and growth/collapse timing, since those would clearly affect the diurnal concentration profiles that you interpret.

Please double- check references.

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