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Interactive comment on "The Chemistry of Atmosphere-Forest Exchange (CAFE) Model – Part 2: Application to BEARPEX-2007 observations" *by* G. M. Wolfe et al.

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The paper shows a comprehensive detailed evaluation of a multi-layer canopy exchange model, coupled to a representation of the lower boundary layer, by comparison with observations of fluxes and concentrations of a range of reactive compounds during the BEARPEX-2007 campaign in the Sierra Nevada, US. The paper is well written, addresses the application of an interesting dataset to not only conduct an quantitative but also a more qualitative analysis to further identify some of the main challenges involved in understanding canopy chemical exchange processes. It is clear from the presented detailed evaluation that in particular the dry deposition process including the

role of non-stomatal removal processes is still poorly understood. In addition, recognizing the potentially rather large exchange velocities of many species it suggests that the exchange also largely depends on turbulent exchange which is known to be generally not well represented in chemical exchange models. This is discussed in the model but no sensitivity analysis is presented to demonstrate the impact on simulated exchanges as a function of different assumptions on turbulent exchange. I have read this paper without reading the accompanying paper on the model development as might be the case for other readers that are not per se interested in all the model details but rather want to focus on the model application. This raises a number of comments where it would be useful to shortly give some background on some model features as indicated below in my comments.

Comments;

Pp 21794, line 14-15; Since you are also in this introduction referring to the relevance of in-canopy interactions for the effective exchange of reactive compounds in regional and global scale modeling (and mentioning feedback mechanisms, see below), you could include some reference to studies that aimed to assess the significance of in-canopy processes on these scales in coupled chemistry-climate studies, e.g., modifying the sentence "gradients in radiation, O3 and turbulent transport on fluxes of NOx (Gao et al., 1991; Ganzeveld et al., 2002a and b, Dorsey et al., 2004; Duyzer et al., 2004)".

Ganzeveld, L., J. Lelieveld, F. J. Dentener, M. C. Krol, and G.-J. Roelofs, Atmosphere-biosphere trace gas exchanges simulated with a single-column model, J. Geophys. Res., 107, 2002.

Ganzeveld, L., J. Lelieveld, F. J. Dentener, M. C. Krol, A. F. Bouwman, and G.-J. Roelofs, The influence of soil-biogenic NOx emissions on the global distribution of reactive trace gases: the role of canopy processes, J. Geophys. Res., 107, 2002.

Line 23; "biosphere-atmosphere feedbacks"; could you provide an example of such feedbacks? e.g., how ozone stomatal uptake can effect photosynthesis which in turn effects the energy balance and turbulent transport of ozone to the surface and into the canopy.

Pp 21795; line 2-3 “CAFE is the first model of its kind to incorporate the extensive master chemical mechanism (MCM)”. This is an interesting claim recognizing the observation that most likely the main limitations in understanding the role of canopy interactions in atmosphere-biosphere exchange are related to our understanding and quantification of biogenic emissions, deposition and, especially, turbulent exchange, and not so much a very detailed representation of chemical transformations.

Pp 21796, line 15; “36 layers within the forest canopy”; what is the main motivation to use such a large amount of canopy layers recognizing the fact that characterization of the vertical distribution of biomass and sources and sinks is probably mostly limited to characterization of first-order differences between under- and upper storey. Is this main motivated by numerical issues or by the representation of particular processes, e.g., turbulent transport or extinction of radiation?

Pp 21798, line 16, here and before there is a reference to the over- and understorey inside the canopy suggesting a distinction between these two parts of the canopy. It is then indicated that the upper- and understorey deposition resistances are scaled to the 36 layers of the canopy scaling with the LAD profile. But would it be not more essential to scale with the radiation extinction profiles jointly with the LAD profiles? In other words, I am asking a short description of the disaggregation method to scale the coarsely calculated source and sink distributions to the 36 layers of the numerical system.

Pp21800, line 4-5; Rather than applying a advection rate constant, did you ever consider to “nudge” the model simulated surface or boundary layer concentrations of the “long-lived” species such as ozone, CO (and NOx) to the observed concentrations (e.g. Ganzeveld et al., 2006) ? In this way you implicitly add the advection tendency to the vertical transport tendencies and secure the model to simulate realistic surface and

boundary layer concentrations. This would have also been an optimal approach to consider the role of advection of isoprene and its oxidation products as indicated at pp21802.

Line 10; it is stated that the diffusion parameters are initialized based on input meteorological parameters. It makes me wonder to what extent the model also simulates online the development of meteorological properties, e.g., temperature and moisture profiles, radiation etc. I am also asking that this would be essential to know in order to study potential feedbacks in the system which cannot be studied in an offline model system with imposed meteorology. Possibly that some more detail on this is given in the accompanying paper but it would be useful to include some short statement about this feature of the model set-up in this paper.

Line 16; what is the reason to consider the soil NO emission into the diffusion operator whereas the other source/sinks are considered in the chemical operator?

Pp 21803, line 10-12; what would be the uncertainty in VOC emissions associated with issues on the representation of the extinction of radiation of the coniferous forest? What is the main issue with the representation of this feature relevant to VOC emissions (and deposition)?

Pp2104, line 25; it is not completely clear from the site information but I could imagine that the soil is relatively well covered resulting in a presence of an inversion inside the canopy with cooler soil temperatures compared to the canopy temperature, which could partly explain a misrepresentation of the deep-canopy profiles. Is there any experimental information available to confirm the presence and relevance of such an in-canopy stratification?

Pp21805; line 8; when you use the term “negative-in canopy gradient” you should indicate if a negative gradient reflects a decrease in concentration with height or vice versa. Would it be more useful to apply the terms “deposition gradient” and “emission gradients”?

Line 14; why do you not consider the canopy-/ground deposition of MVK and MACR although references are listed that have reported these numbers??

This also brings me to a comment that I want to make with respect to the Zhang et al. 2003 reference on “improved” dry deposition parameterizations. I want to express my concern that it appears that this model has been somehow established as a benchmark model system (also being referenced in the recent Karl et al. 2010 paper on VOC deposition) to treat dry deposition in CAFÉ and regional air quality models system. Possibly that this review is not the most optimal medium to express this concern about other scientific activities but since your model relies largely on this work, I want to stress that according to me the Zhang et al. parameterizations, especially those on non-stomatal uptake, are based on non-existing correlations. The evaluations were only based on reporting the fact that model provided reasonable numbers (the minimum that you would expect from a model) generally being seen for O3 and SO2 dry deposition velocities but not based on a direct comparison of simulated and observed Vd’s.

Pp 21806; the role of HCHO in explaining the discrepancy between the observed and simulated OH reactivity for the cold period is interesting also reading that measurements show noontime maximum mixing ratios up to 12 ppbv. What would be the lifetime of HCHO for the specific conditions in terms of temperature and radiation. Would it be long enough to point at a distant anthropogenic source or biogenic source from high-emitting isoprene landscape. How did the diurnal cycle in HCHO look like? Does it point at an important role of nocturnal processes and downward mixing of HCHO associated with elevated concentrations higher up in the BL?

Pp21807, line 12; here you actually make the point that I raised before by explaining what the positive gradient reflects.

Pp21815, line 1-3, the discussion about the underprediction of O3 deposition is interesting and especially raises the issue on the model representation of the non-stomatal sinks based on the already mentioned Zhang et al. dry deposition model. This model

is supposed to do a better job on non-stomatal uptake compared to the classis Wesely approach but this is apparently not the case for the BFRS site. What is the value of the selected soil uptake resistance? What are the soil properties at this site, is it a site with a relatively high organic content (which would likely enhance soil deposition). It would be useful to see how sensitive the simulated O3 deposition is to assumptions on the ground versus the cuticular resistances.

Pp21816; especially with the NOy you would expect a very significant contribution by advection due to relative long lifetimes of the various contributing compounds; how much is the relative contribution by advection compared to the local production and downward mixing? This also brings me to a point that I should have raised earlier; how did you initialize the vertical concentration profiles all the way to the top of the vertical domain and how do you treat the concentrations higher up? Did you fix the concentrations at 800m height?

Pp21818; line 11-12; you state that “if above canopy NO2 fluxes are a good indicator of the soil NO flux suggesting that the soil NO flux is overestimated by about 50%. This is initially confusing since from the initial introduction of the model soil NO emission flux I inferred that this was scaled to reproduce the observed soil NO flux but this is apparently not the case. It would be useful to already mention this fact with the introduction of the treatment of the soil NO emission flux. Why would the NO2 flux be a good indicator of the soil NO emission flux? assuming that all NO would be transformed into NO2 and that a substantial fraction of this would be deposited inside the canopy, you would expect a NOx/NO2 canopy top upward flux (apparently, the contribution by advection in NOx is small compared to the soil NO source) that would be substantially less than the soil NO emission flux. In other words, what is the canopy reduction factor (CRF) for this site?

Pp21822; in the discussion on the chemical versus the exchange velocities of some of the APN compounds it would be useful to indicate the chemical timescale of the production of the various APN compounds relative to the turbulent timescale as a function of height inside- and above the canopy. According to the discussion the chemical timescales should be really short to effect the exchange velocities but to put this in perspective it would be useful to see how the models turbulent timescales compares to values reported for other sites.

Pp 21825; in the discussion about AN and HNO3 deposition, with inferred Vd's large as 2.7 (AN) and 3-4 cm s-1 (HNO3), which should be close to the turbulent limit, you would expect a negligible role of surface uptake resistance including the stomatal resistance. How do you explain then that you still see a role of the difference in stomatal resistance between the cold and warm period?

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 21791, 2010.