

Interactive comment on “The Atmospheric Chemistry and Canopy Exchange Simulation System (ACCESS): model description and application to a temperate deciduous forest canopy” by R. D. Saylor

Anonymous Referee #2

Received and published: 16 November 2012

This paper describes the application of a 1D column model to describe the chemistry and physics of a forest canopy ecosystem. The modeling results predict concentrations and fluxes of gas phase organic compounds. Independent measured vertical profiles of isoprene, are compared to model predictions. Sensitivity simulations suggest NO_x-influenced photochemistry is non-linear, which is consistent with conventional wisdom. The author's main contention is that these findings could have an impact on the relative importance of aqueous- versus gas-phase SOA formation. While I am inclined to agree, the link between this assertion and the model findings should be stronger

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and described more efficiently. I recommend acceptance of this paper provided the comments below are addressed.

overall comments: In the abstract, the author states "Model results of isoprene profiles and fluxes are found to be consistent with previous measurements made at the simulated site and with other measurements made in and above mixed deciduous forests in the Southeastern United States". This statement is not strongly supported by the presented results. Measurement data for isoprene fluxes are not presented in the main body of this manuscript. In Figure 7 there is comparison of measured isoprene mixing ratios at different heights, compared to model predictions. It is difficult for the reader to fully realize the abstract's assertion concerning profiles as some measurements differ from the model predictions for the same elevation from a factor of 3 to >6. The degree to which the model is reasonably consistent with measurements requires an understanding of the bNO_x for each measurement. If the measurement data was color coded the same as the model predictions for bNO_x, this would provide insight as to whether the dependence in model predictions of isoprene vertical profiles to bNO_x is supported by field measurements, and in fact if BVOCs are reasonably represented by the modeling system.

The statement at the end of the abstract "...canopy which may have an impact on the relative magnitude of SOA formed through aqueous- versus gas-phase pathways as a function of anthropogenic influence..." is due to the fact that predicted biosphere fluxes of water-soluble organic compounds (that form SOA in laboratory experiments) is dependent on simulated NO_x values. This is central to the author's point, but is never explicitly stated.

The literature search presented in the introduction is extensive, but difficult to connect to the abstract's objective related to aqueous and aerosol phase chemistry. The articles presented here are arguably the important and relevant literature for describing the physics of sub-canopy flux, and it is clear the author understands this body of work. It is difficult for the reader to keep track of the important chemical details regarding

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differences in development of model chemistry and its application because a synthesis of key details affecting critical uncertainties is not identified, in particular related to gas and/or aerosol phase chemistry. Further, the introduction is largely a discussion regarding canopy focused on differences in tree type (loblolly pine plantation, idealized forest, different types of real forests) and oxidants (NO_x and OH). The author states, "most previous investigations have focused on gas-phase chemical processes and the effect of BVOC emissions.... multiphase modeling and measurement studies of forest-atmosphere exchange are needed to....", but then describes how this work also only considers gas phase chemistry.

The author's contention that the relative importance of aqueous versus gas phases processes for SOA formation is important, is not strongly linked to the previous discussion in terms of chemistry or physical nature of the studied environments.

detailed comments: The author explains in detail how several BVOCs are included in the chemical mechanism(s). However in the emissions used in the modeling exercises here, only isoprene emissions are considered. Discussion as to why this is suitable for the area being studied needs to be justified. What are the effects and specific limitations of neglecting a large fraction of the total emissions?

In the model description it is noted that there is a 60 sec integration time step for sub-canopy processes. Has the author performed sensitivity studies that demonstrate this time step is sufficient? The discontinuity in soil NO_x emissions at 30 deg. C, suggests some possibility of instability.

As the author notes, anthropogenic influences on biogenic SOA have been mentioned in the literature (e.g., Hoyle et al applies the idea to forests in Finland). Murphy et al., (2008) and Carlton et al. (2010) describe anthropogenic NO_x effects on biogenic SOA specifically to the Eastern U.S. Would the model results presented here support or contradict those findings applied to the same area the author studies here?

On page 24783 (and elsewhere in the paper) there is description of how model-

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prescribed NO_x is changed, but prescribed ozone is not. Can the author explain why this is a realistic and valid test? In isoprene dominated areas, one would expect that ozone formation (and concentration) is NO_x-limited. Changes in NO_x inputs would be expected to have an impact on predicted ozone mixing ratios, both for the initial boundary conditions.

page 24786 ... the author describes debate in the literature regarding isoprene oxidation in high and low NO_x environments. Is 'low-NO_x' an appropriate description of the area studied here?

page 24788. In what ways are Walker branch and the Ozarks similar, is it in terms of isoprene dominance or in terms of NO_x, ozone and vertical mixing structures also?

table 2.... more water soluble products at higher NO_x. It is very interesting to note, that despite a variety of differences in the applied models, this finding is consistent with Ervens et al., (2008), GRL, who found that for a given set of conditions, when the VOC:NO_x ratio is lower (e.g., higher NO_x conditions) a larger fraction of the total organic gas phase carbon is more water soluble.

figure 8 - isoprene can be chemically produced?

Can the canopy height be added to the vertical profile plots?

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 24765, 2012.

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