Reviewer #1

The author sincerely thanks the reviewer for a careful and thorough review of the manuscript. Responses to specific comments are presented below in italics.

General Comments

Rev#1: In the abstract, it is stated that a key goal of this effort is to "better understand the formation of SOA mass from forest emissions...". This is misleading, as the model is solely focused on gas-phase chemistry as presented. To avoid confusion, this should be moved to the bottom of the abstract or saved for the conclusion section. It should also be noted that no 1-D canopy models (to this author's knowledge) currently contain SOA mechanisms, and this is a crucial next step in their evolution.

Response: *The reviewer's point is well taken. The abstract and summary have been modified in the revised manuscript to make it clearer that this work reports on an initial, gas-phase-only version of the modeling system.*

Rev#1: Regarding turbulent transport: on page 24779, l. 28, the author states that "simplification of the turbulence model, while far from being a trivial impact, does not substantially change major conclusions drawn from these investigations." One might argue, however, that if key conclusions are unaffected, then the impact of using K-theory (versus something fancier) is effectively trivial. To stimulate a discussion on this issue, I am wondering if the author could comment on what, if anything, might be gained from a more complete turbulence representation. In other words, what questions can we not answer owing to a limited turbulence representation?

Response: *The reviewer raises a valid challenge. The author now believes that the phrase "… does not substantially change major conclusions drawn from these investigations" is overstated. By necessity, most previous canopy chemistry modeling studies have either implicitly or explicitly assumed that K-theory is adequate to represent the environmental conditions most relevant to the particular questions on which each investigation is focused. In reality, the computational burden imposed by higher-order treatments of reactive turbulent transport has been too large to overcome and has been the primary motivator for more simplified approaches. This work faces the same burden, especially because of the desire to avoid the use of condensed chemical mechanisms and the resulting difficulties that then arise when comparing model results of condensed mechanism surrogate species with real ambient measurements. It seems that most previous 1-D canopy chemistry modeling investigations have assumed that K-theory, on average, can account for the majority of turbulence regimes that are important for forest-atmosphere exchange. However, the recent work of Steiner et al. (2011) for the 2009 CABINEX study has convincingly demonstrated that intermittent coherent structures (canopy-scale "sweep-ejection" cycles) can account for a large fraction of momentum (40-50%) and heat (44-65%) transport. Of course, K-theory is completely inadequate to model these events. So, if it is assumed that the other, roughly half, of heat and momentum transport can be adequately modeled by K-theory (or some slight modification of it, as for example, the near-field theory of Rapauch (1989)), then 1-D models such as ACCESS can still be useful. It is the author's belief that understanding of the impact of within- and above-canopy turbulent structures and dynamics on reactive trace gas fluxes is at best incomplete at this point and deserves much more attention from the research community. At the very least, any future forest canopy atmospheric chemistry measurement campaigns should include a full suite of turbulence-related measurements, preferably at multiple levels within and above the canopy.*

To address the reviewer's concern raised in this comment, the discussion on this topic has been modified in the revised manuscript to better reflect the discussion provided above.

Raupach, M. R. (1989) A practical Lagrangian method for relating scalar concentrations to source distributions in vegetation canopies, Quarterly Journal of the Royal Meteorological Society, 115, 609-632.

Steiner, A. L., Pressley, S. N., Botros, A., Jones, E., Chung, S. H., and Edburg, S. L. (2011) Analysis of coherent structures and atmosphere-canopy coupling strength during the CABINEX field campaign, Atmospheric Physics and Chemistry, 11, 11921-11936.

Rev#1: The supplement is probably unnecessary, as all relevant information for the chemical scheme can be found on the MCM website.

Response: *In general, the author agrees with the reviewer, except that the supplement explicitly documents the mechanism used in the current study including changes to thermal rate coefficients from that suggested by MCM. Additionally, the code used to calculate more complex rate coefficients is presented at the end of the supplement. The author will abide by the editor's decision whether or not the supplement should be included in the final publication.*

Specific Comments

Rev#1: Introduction – The discussion of previous work is quite thorough. You might also include results from Rinne et al. (2012).

Response: *The author thanks the reviewer for the additional reference. A citation and brief discussion of this work is included in the revised manuscript.*

Rev#1: p. 24770, l. 6-9: I have several comments here:

1)   I suspect that the cited reference should be Wolfe et al. (2011a).

Response: *The reviewer is correct. The error is corrected in the revised manuscript.*

2)   Mao et al. (2012) have demonstrated a positive artifact in the OH observations from the PSU LIF instrument. Correction for this artifact decreases [OH] by a factor of 2 for the hottest conditions of BEARPEX 2009, and a similar bias likely exists in the 2007 observations. As such, the magnitude of the missing OH source is probably smaller than originally proposed (OH was off by a factor of 6 in the CAFE study (Wolfe et al., 2011a)).

Response: *Mao et al. (2012) was published after the original submission of this manuscript, hence the omission of this very important result. The revised manuscript includes a citation and discussion of this work.*

3)   Also of relevance is the potential for aerosol formation from O3 + BVOC reactions, discussed in Wolfe et al. (2011b) but not mentioned here.

Response: *The author thanks the reviewer for pointing out this oversight. A brief discussion of this topic is included in the revised manuscript.*

Rev#1: p. 24773, l.6: what is meant by "dynamic mode"?

Response: *The author agrees that this terminology is non-standard and may be confusing. The sentence has been modified to read … "The model can be run in either steady-state mode at a prescribed fixed time or over a full diurnal cycle or in prognostic mode from a given set of initial*

conditions and allowed to evolve deterministically over time as a function of dynamically-varying input data." The term *prognostic* is more familiar and should be easily understood by most readers.

Rev#1: p. 24773, l.28: what is the reason for mapping J-values to MOSAIC? Does this somehow include cloud cover or better radiative transfer? How different are these values (e.g. compare J(NO2) and J(O3) for MCM and MOSAIC)?
Response: Using J-value approximate functions from MOSAIC was simply a convenience. The author of the current work created these computationally efficient J-value parameterized functions originally for Easter et al. (2004). Zaveri et al. (2008) then employed revised versions of the functions in MOSAIC. The revised parameterizations in MOSAIC are now being used in ACCESS. Rather than explain the long (and largely irrelevant) history of these functions, but still give proper credit to the authors of Zaveri et al., only the last link in this chain was referenced.

Easter et al. (2004) MIRAGE: Model description and evaluation of aerosols and trace gases, Journal of Geophysical Research, 109, D20210, doi:10.1029/2004JD004571.

Zaveri, R. A., Easter, R. C., Fast, J. D., and Peters, L. K. (2008) Model for Simulating Aerosol Interactions and Chemistry (MOSAIC), Journal of Geophysical Research, 113, D13204, doi:10.1029/2007JD008782

Rev#1: p. 24775, l. 7: This parameterization is originally from Yienger and Levy (1995). Also, it is specifically for dry soils and may not be appropriate for WBW. Analogous parameterizations are given in the above reference.
Response: The additional reference is noted in the revised manuscript. Since there is no information available (at least to the author's knowledge) concerning soil NO fluxes at the WBW site, the simulations presented in this manuscript used only a small basal soil NO emission rate (5 ng N m^{-2} s^{-1}) which was identical in each of the background NO_x experiments. It is not expected that the small flux employed would significantly alter the results or conclusions presented in the manuscript (except possibly at the lowest bNOx value). Future versions of the ACCESS model will include more comprehensive parameterizations of this process from Yienger and Levy (1995) and other sources.

Rev#1: p. 24775, l. 17: Is this not originally from Wesely (1989)?
Response: Of course, the foundational work of Wesely (1989) underlies much of our current understanding and modeling formalism for dry deposition; however, the multilayer treatment of dry deposition within vegetative canopies by Meyers et al. (1998) is directly employed by ACCESS.

Rev#1: Section 2.3: Is deposition implemented for all 2,429 species? If so, how are values for f_0 and H* chosen?
Response: Yes, dry deposition is implemented for all integrated species in ACCESS. However, since data is limited for many of the intermediate and product species of MCM, default values for the molecular diffusivity, Henry's Law constant and f_0 were employed when no reliable data could be found in the literature for a particular species. The revised manuscript explains this in more detail and provides the currently used default values for these physical data.

Rev#1: Does the model follow the recommendation of Karl et al. (2010) for fast oVOC deposition, as this will affect conclusions?

Response: The preliminary results presented in this manuscript do not incorporate the recommendations of Karl et al. (2010) for enhanced deposition of oxygenated VOCs. A follow-on investigation to the current work is under way which will incorporate additional measurements of Fuentes et al. (2007) from the Walker Branch Watershed (WBW) canopy during the summer of 1999. One of the goals of this broader study is to investigate whether there is any evidence in the data from WBW which would support the recommendations of Karl et al. (2010). Investigations of this sort are exactly what ACCESS has been designed to probe.

Rev#1: p. 24780, l. 17-19: How are values for v_{si}, k_e and boundary concentrations chosen?
Response: Deposition velocities to the forest floor were computed as recommended by Wesely (1989), again using default values for physical data H^* and f_0 where no reliable data could be obtained. For the results presented in this manuscript, an entrainment rate of 100 cm s^{-1} was used. The revised manuscript includes these additional details in the section where v_{si} and k_e are defined (Section 2.4).

Rev#1: p. 24784, l. 2: Could an average measured isoprene flux be added to Fig. 8 to provide a benchmark?
Response: Yes, available flux measurement data will be added to Figure 8(a) in the revised manuscript.

Rev#1: Section 4: This could be broken into a few subsections to make it more digestible.
Response: The author disagrees with the reviewer on this point and feels that the presentation of Section 4 is fine as is. Thoughtful placement of the Figures in the final formatting of the article will likely help to increase the clarity of the discussion.

Rev#1: p. 24786, l. 21: While isoprene chemistry is fairly well understood at NOx levels above a few hundred ppt, low-NOx chemistry is still hotly debated due to newly-proposed mechanisms (Paulot et al., 2009; Crounse et al., 2011; Peeters and Müller, 2010; Peeters et al., 2009; Wolfe et al., 2012). Some discussion of this distinction is warranted as it can influence results and uncertainty at the low end of the NOx sensitivity study.
Response: The author is not convinced that an extensive discussion of these details is actually warranted for this manuscript. As pointed out by Reviewer #2, and as confirmed by data from Fuentes et al. (2007), the WBW site probably could not be categorized as truly "low-NO$_x$" with typical NO$_x$ measurements ranging in the 2-6 ppbv range. Furthermore, research in this area is progressing so rapidly that any in-depth discussion at this time may be quickly out of date beyond pointing out that there is significant mechanistic uncertainty in this regime. To address the reviewer's concern on this point, however, paragraph 3 in Section 4 has been expanded slightly to make this point.

Rev#1: p. 24787, l. 11: It would be helpful to know what the average NO mixing ratios are for each simulation (rather than background values), as this is what controls the fate of RO2 and thus production of oVOC and it is difficult to read from the figures.
Response: The author agrees with the reviewer on this point. Mean within-canopy mixing ratios of steady-state simulated NO$_x$ have been added to Table 2 in addition to the bNOx value reported previously.

Rev#1: p. 24787, l. 26: What is the "process chain" for this dependence of reactivity on NOx? Is

it just more ozone and therefore more OH?
Response: Yes, within the range of background NO_x values used in the experiments, the increasing reactivity is a result of increased ozone and OH concentrations and thus the increasing rates of chemical loss of isoprene. The revised manuscript contains an additional phrase making this point clear.

Rev#1: p. 24788, second paragraph: a nice way to summarize these results might be to plot isoprene escape efficiency vs. background NOx values.
Response: The author agrees that such a plot would be a useful presentation of this data; however, a more appropriate venue for this would be in publications resulting from the ongoing follow up study where the isoprene escape efficiency can be presented as a function of a wider variety of influences rather than only background NO_x values.

Rev#1: p. 24790, l. 10-12: What is the upshot of this result? Perhaps that SOA precursors will be more locally confined in areas of high urban influence?
Response: Given the preliminary nature of the results presented here, the author is hesitant to speculate too aggressively on broader implications of this result. In the ongoing follow up study, using additional previously unpublished data from Fuentes et al. (2007), these results (and others) will be explored more fully.

Technical Comments
Rev#1: p. 24777, l. 21: define v_{air}.
Response: v_{air} is the kinematic viscosity of air and is defined as such in the revised manuscript.

Rev#1: p. 24782, l. 33: define L.
Response: L is the Obukhov length and will be defined as such in the revised manuscript.

Rev#1: p. 24792, l. 2-8: this is a large sentence and could be broken up.
Response: The author feels that this sentence, although lengthy, is still clear in its communication and should remain as is.