Author Response to Anonymous Referee #1

We thank the referee for reviewing our manuscript and providing important feedback. Our responses to questions and suggestions are outlined below.

REFEREE: **1. Abstract. The authors discuss how the model "offers new insights into the forest atmosphere exchange." Yet, these new insights were not put into the abstract in a way that makes them sound unique from other studies. I would like to see a revised abstract that includes clarifies what is novel about their results (e.g., the NOx partitioning and sensitivity to gradients; APN fluxes affected by the in-canopy chemistry) and make some statements that discuss how the model limitations (e.g., turbulence; deposition parameterizations) affect their results.**

RESPONSE: We agree, and we will revise the abstract to better emphasize new insights obtained from the modeling results that include NO_x partitioning and gradients, the sensitivity of APN fluxes to chemistry, the inability of measured sesquiterpene concentrations to explain a chemical ozone flux, among a few others.

We acknowledge that uncertainties in diffusion and non-stomatal deposition limit the quantitative comparison between modeled and measured fluxes. This was also pointed out by Dr. Thomas Karl in a short comment on the present manuscript. In our companion paper and in the body of this paper, we mentioned that our turbulence and deposition parameterizations are known to be oversimplifications that inherently introduce uncertainties. Regardless of the limitation imposed by these uncertainties, we still feel that it is important to highlight the delicate and complex link between chemistry and forest-atmosphere exchange. We will more explicitly mention these caveats in the conclusions of this paper – we feel this type of discussion is not necessarily appropriate for an abstract, though we will defer to the editor in this case.

2. VOC Section 3.1: - Isoprene emissions (page 21802). The lack of local isoprene emissions at Blodgett forest and the inability of the model to produce the local isoprene concentrations makes me question the ability of advection parameterization to accurately add the necessary inputs.

Our advection parameterization is capable of bringing modeled and measured isoprene concentrations into agreement, but not isoprene and its main first generation oxidation products MVK and MACR simultaneously. This result is not terribly surprising given the simplicity of the approach. But, we emphasize here (and in the manuscript), the primary role for the advection parameterization to maintain agreement between modeled and measured concentrations of longer-lived species so that we can more directly compare the measured and modeled fluxes.

I don't agree with the statement on page 21803 line 13-16 "Since modeled mixing ratios of locally emitted BVOC are primarily a function of the ratios of emission and chemical loss (e.g., advection often plays a small role)" – clearly this is not the case for isoprene because of the tuning of the authors to make measured and modeled concentrations match. It seems more realistic to put some

isoprene emissions from the understory than to make the assumptions that it is coming out of the canopy.

In regards to our statement about emissions and chemical loss, we think this is still a valid statement for *locally* emitted VOC (e.g. MBO, MT, SQT.). As the referee notes, this comparison is not particularly useful for isoprene, which is not emitted by ponderosa pine (but perhaps from the understory), and thus we have removed isoprene from that particular discussion.

As for which part of the canopy we choose to emit isoprene, we first note as mentioned in the manuscript, our isoprene emission rate is nearly identical to that used in the 4km x 4km grid cell of CMAQ (Steiner et al., 2007), thus we consider this to be more representative of a regional, rather than a true local (i.e. within the fetch) emission. We also state in the manuscript that previous leaf-level emission studies at this site suggest no isoprene emission from either P. pine or the dominant understory vegetation. Thus it seemed rather arbitrary whether we assigned these emissions to the understory or to the overstory. We feel that it is sufficient to state that this is a known issue with CAFE and to point out when this issue might affects our results.

- Case iii – the likelihood that turbulent mixing is "less efficient than parameterized here, leading to the buildup of BVOC emitted from the understory" is probably rather low. As is, the turbulent mixing at that height is probably modeled as close to zero at that height in the canopy – reducing it even further would hardly make a difference. A figure that shows how the modeled K values from Part I could change would be helpful to support or disclaim this theory. Furthermore, this isn't supported by the stagnation in section 3.5.1– Figure 7a shows that the gradients are stronger when it is colder, which is counter-intuitive (colder conditions likely lead to more stagnation). A sensitivity test could be performed to test this.

Comparison with several gradient observations – monoterpenes, methyl chavicol, NO2 and PAN – suggest that CAFE does not accurately represent all relevant processes near the ground. A stagnant layer near the ground is a unifying explanation for these observations, as it would lead to 1) a buildup of emitted BVOC, 2) a buildup of NO2 from the NO ground emission source, and 3) a decrease in PAN due to higher temperatures in this region. Unidentified emission and/or deposition source from/to the ground are another possible explanation, as we note in the text. It is true that the observed NO2 gradient in Fig. 7a is weaker during the cold period; however, NO emissions are also a function of temperature, and a decrease in these would lead to a weaker NO2 gradient.

It may be more appropriate to call this postulated layer "weakly coupled" than "stagnant." Simply changing K throughout the canopy, as was done in the sensitivity tests of Part I, would not reproduce these phenomena. Following the Referee's suggestion, we have performed a simple sensitivity test where the K-values in the bottom half of the canopy were reduced by a factor of 10. This test shows a large buildup of NO2 and MT and a mild reduction in PAN, as we anticipated. We admit that this is not a complete explanation for our model-measurement discrepancies; for example, the above-canopy NO2 gradient during the hot period is also quite steep and cannot be explained by a weakly-coupled layer in the lower canopy. We will add some text regarding the results of this sensitivity test in the appropriate sections.

3. Section 3.3 Peroxides. The authors' summary of the measured-modeled mismatch in ROx species was very well characterized (page 21807). These results from these sensitivity tests were certainly one of the more interesting aspects of this paper, and it would be helpful to have them highlighted more succinctly in the abstract.

As the referenced page is in Sect. 3.2, we assume the Referee is referring to the discussion of OH, HO2 and RO2 (though the chemistry of peroxides certainly ties into this). We will add some of the key points about our solutions to modeling ROx chemistry in the abstract and conclusions.

4. Section 3.4 Ozone. The conclusions of this section of the paper and their tie to other results (e.g, Kurpius and Goldstein 2003, etc.) were seemingly in contrast with the abstract, which in the last line claims that "CAFE underpredicts ozone fluxes by 20%, which may indicate additional in-canopy chemical losses are missing from the current model" and in the conclusions section (page 21828). My understanding of what the authors are saying is that the uncertainties were more likely due to non-stomatal loss processes, not the chemistry. Please clarify in the abstract and conclusions.

Our statement in the abstract about the source of the "missing" ozone flux is likely too strong, and we will revise it accordingly. The model-measurement comparison of ozone fluxes merely suggests that we are missing an in-canopy ozone sink. This may be due to dry deposition (e.g. cuticles) or in-canopy chemistry, both of which are "non-stomatal." The model does include non-stomatal deposition (e.g. to cuticle surfaces), but the parameterization of this process is highly uncertain and likely overestimates this processes. Ozonolysis of unidentified, highly reactive BVOC has been invoked from previous observations at this site, and we wish to point out that our results are not inconsistent with this hypothesis. We will modify the abstract, conclusions and Sec. 3.4 to better explain our findings.

5. Section 3.5 Reactive Nitrogen. Why do the authors think that NOy during this year is much less than other years (p. 21816, line 17)? Any major differences to highlight?

Unfortunately, data collection at BFRS was much more limited in previous years than during BEARPEX-2007. It may be worth noting that average ozone during BEARPEX-2007 is ~10 ppbv lower relative to earlier years, which is not a cause but is consistent with lower NOy (see e.g. Fig. 18 in Murphy et al. (2006, ACP)). At this time, we do not have the necessary data to assess inter-annual variability in NOy at this location; however, we will add some text in this section to point out that the 2007 period was different from observations in previous years.