We thank the anonymous referees for their helpful comments. We have fixed all technical comments from both referees. Responses (in bold) to general comments from each referee follow.

Responses to Anonymous Referee #1

1.) The approach is done using the 2 product approach by Odum and colleagues in 1997 although more advanced however reduced chemistry schemes exist (Derwent et al., 2003; Bonn et al., 2004; 2005). This will certainly impact strongly on a) the amount of SOA formed and b) on the detailed regional impact as the aerosol is no permanent sink anymore but acts as a temporal sink with a release in a later stage that influences the gas-phase chemistry again. Please comment much more on this!

We agree with the referee that the Odum two product approach to aerosol modeling is less than ideal and fails to accurately capture the semi-volatile nature of SOA and the subsequent impact on gas phase chemistry. As the referee points out, there are more detailed treatments of the SOA production from monoterpenes elsewhere in the literature. Our focus here, however, was the gas-phase chemistry of monoterpenes and investigating the impact of uncertainties regarding monoterpene nitrates. As such, a full discussion of SOA and the impact of SOA on gas-phase chemistry is beyond the scope of this work. We have added the following to the end of Section 2.1:

> *Although more detailed aerosol chemical mechanisms that track the individual species as they transfer between the gas and particle phases do exist (e.g., Derwent et al., 2003; Bonn et al., 2005), our focus here is on investigating the impacts of the gas-phase representation of the chemistry and a full consideration of the aerosol is beyond the scope of this work.*

2.) There are a couple of different chemical schemes on the market (RACM, STOCHEM, MCM derived ones) that display notable differences. How do these intercompare here with respect to the results shown?

As stated in the manuscript, the backbone of our mechanism comes from RACM2. We chose RACM2 due to its inclusion of two monoterpene species, which is the most of any reduced chemical mechanism suitable for a regional CTM that we are aware of. A full description of the differences between the various chemical mechanisms is beyond the scope of this paper and we point the reviewer to the RACM2 model description paper for further information regarding this mechanism (Goliff et al., 2013).

3.) The authors basically state the major monoterpene species would be α- and β-pinene. This highly matters on the tree species involved and a large number of differently structured monoterpenes will be emitted especially when focusing on the whole region of Canada. This will impact on the reactivities with the ambient oxidants, i.e. ozone, OH and NO3 in detail that are hardly matched by a single scheme used in common CTMs. I am aware of the complexity of the problem and the challenges for modelling. But this will have particular consequences on the oxidation capacity, the SOA contribution and the aerosol as well as the gas phase chemical speciation! Please consider this much more in the discussion. This certainly is difficult in a regional or global modal. However this will modify the chemistry from the emitting needle towards the boundary layer towards the free troposphere. Please consider this at least in the considerations. Please mention this at least in more detail. But let's discuss more because its an online journal that offers the possibility and the tools are available. Please consider this in more detail when elaborating your results although they are certainly interesting, nice and mainly correct.

We do not assume that the major monoterpene species are α- and β-pinene and we even state in section 3.2 that measurements of emissions from black spruce trees indicate that emissions of camphene are larger than those of α- or β-pinene. Furthermore, the chemical mechanism has two different monoterpenes: one that behaves similarly to α-pinene and one that behaves more like limonene. This confusion may have resulted from the fact that α- and β-pinene were the only two monoterpene species measured during the ARCTAS campaign.

We note that the MEGAN biogenic emission scheme used in our model calculates the emission of 134 species, many of which are monoterpenes. The monoterpenes have been partitioned into the model species API (low reactivity, similar to α-pinene) and LIM (higher reactivity, similar to limonene). We agree that consideration of more monoterpene species would allow one to more accurately model the chemistry; however, limited data exists on the oxidation rates and products of other monoterpenes. Coupled with the increased computational costs associated with more species, the inclusion of more monoterpene species is beyond the scope of this work. Given the importance of the gas-phase chemistry of monoterpenes as demonstrated by our work, we hope that this encourages more research into the gas-phase oxidation rates and products of other monoterpenes so that these questions can be investigated and lead to improvements in our atmospheric chemistry models. We have added the following to the discussion:

> *It is worth noting that the speciation of the monoterpenes in these environments is likely different and that a parameterization using two representative monoterpenes may not be sufficient to capture the resulting differences in chemistry. We believe that limitations in our knowledge of monoterpene emissions and their gas-phase chemistry likely overshadow any uncertainties introduced with using two representative species in a chemical transport model. Nonetheless, our work has demonstrated that gas-phase monoterpene chemistry is important and further improvement in emissions and representations of their chemistry appropriate for reduced chemical mechanisms is warranted.*

4.) I actually miss the possibility of trajectory analysis when using aircraft measurements and models. That allows a very nice set-up of detailed investigations in this context. Why is this not done?

Our focus here was to compare the impacts of different assumptions regarding the fate of monoterpene nitrates on the NO_x and HO_x budgets. The ARCTAS measurements are used to show that the model results are realistic on the regional scale and are not intended to be directly compared to the model results.

5.) The water solubility mentioned. I kindly ask to consider and to take into account the huge amount of carbon molecules of monoterpenes and the large functionalities of their oxidation products. Assuming Henry's law coefficients between 10^3 to 10^5 may be a little bit to elevated. Have there been any sensitivity studies with a range of water solubilities?

We agree with the referee that the Henry's law coefficient is likely lower. We use the same Henry's law coefficient as Ito et al., (2007). To our knowledge, there have been no sensitivity studies on this. We note that decreasing the Henry's law coefficient would only have a minor change in our results since deposition only accounts for 16% of the loss in the RECYCLE case.

Responses to Anonymous Referee #2

1.) One area that I think should be discussed further is wrt the yield of MTN's in the reactions of MT-derived peroxy radicals with NO. The authors have assumed a roughly 20% yield, which is probably OK, but it seems to me that there are also measurements that suggest near-zero yields (e.g., Aschmann et al., 2002). Additional discussion on this issue, which directly affects the results obtained, is warranted in my opinion.

In Aschmann et al. (2002), the authors hypothesize that the low yields may be due to losses on chamber walls or on aerosol and that their results may be biased low. To our knowledge, this is the only study reporting such a low yield while numerous other lab and theoretical studies show results more consistent with organic nitrate yields near 20% (e.g., Nozière et al., 1999; Peeters et al., 2001; Capouet et al., 2004; Leungsakul et al., 2005; Rollins et al., 2010; Vereecken and Peeters, 2012). Given these results, we believe our yield represents a reasonable estimate, but acknowledge that the real number may be higher or lower. We have added the following sentence to Section 2.2 to address this uncertainty:

> *The exact yield of monoterpene-nitrates carries some uncertainty since yields have only been directly measured for a few specific monoterpenes and these measurements are subject to biases due to loss of the nitrates to aerosol, chamber walls, and/or hydrolysis. The exact yield may be lower or higher than used here, however, any reasonable changes changes will only impact the exact numerical results and not the overall conclusion that MTNs are important in the* NO_x *and* NO_y *budgets.*

2.) In Figure 3, the modeled altitude profiles seen for the biogenics and their reaction products differ from those measured during ARCTAS (measured profiles peaking at altitude, modeled profiles decreasing with altitude). A more detailed discussion of this qualitative difference is warranted, in my opinion. Is the model not capturing key processes? Is this different behavior likely to impact any of the conclusions reached?

This is the result of the high biogenic plume during flight #20 at 21:51 to 22:28 that WRF predicated in the wrong location. Although we use the WRF-Chem data from where the model predicts the plume to be, there are likely vertical transport issues as well which

would explain the difference in vertical profiles. Removing that plume would result in vertical profiles with the same shape in both the model and the measurements with the peak near the surface as is expected for a reactive trace gas that is emitted from the surface. We have added the following to the caption for Fig. 3:

> *The apparent maximum in the ARCTAS profiles (red) at 800-900 mbar is due to the plume high in biogenics sampled during flight #20 at 21:51 to 22:28 UTC. If this plume is removed, both the model and the measurements show a smoothly decreasing profile with increasing altitude, as expected for reactive species with surface emissions.*

3.) On page 22303, line 7 – It would be more accurate to say that the nitrooxy-peroxy radicals (mostly) return NOx upon further reaction, rather than saying that they decompose.

We have changed it to read: *one which primarily recycles* NO_x.

4.) Page 22306 – Could the differences in the modeled vs. measured MVK/MACR ratios also be related to transport (age of air), since MACR is generally shorter-lived than MVK?

Yes, however that would also imply that the emission also needs to be altered (which is likely) since the isoprene to MVK ratio is more similar (although not perfect) than the isoprene to MACR. We have added this possibility to the text.

5.) Page 22308 – I am confused by the statement that 90-95% of the ANs are biogenic. Aren't the biogenics more like 80% in Figure 5a, for example?

We apologize for the confusion; below ~56oN where most of the flight track occurred and thus most of the points averaged to make Fig. 5 ~80-85% of ΣANs are biogenic. Further north it is more like 90-95% due to a reduced anthropogenic influence. We have clarified this in the text by changing it to read:

> *In the northern part of the domain (north of approximately* 56^{o}*N) ~90–95% of ΣANs are biogenic. This number decreases to ~80-85% in the southern portion (which has better overlap with the ARCTAS flight data), due to more anthropogenic influence (not shown).*

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