Response to reviews

Reviewer comments are in **bold**. Author responses are in plain text labeled with [R]. Line numbers in the responses correspond to those in the discussing paper. Modifications to the manuscript are in *italics*.

Reviewer #1

The paper is very well written; it's well organized and details are mostly well explained. With experiments at different levels of ozone and b-caryophyllene carried out, the results are relevant. Parameterization of the aerosol yield data is valuable for modelers although in taking such parameterization and applying it to all sesquiterpenes the inherent assumption is that ozonolysis of all sesquiterpenes behaves like b-caryophyllene. I believe the analysis is sound; the only concern I have is about collection efficiency of AMS being 1 (item 3 below). Since the yield calculations are based on AMS measurements, it is fundamental to the findings of the paper that AMS quantification is done properly. Because of this, although I am suggesting publication of the paper after my 'minor' comments and questions are addressed, I would still like to review the response from the authors before publication.

[R0] We thank the reviewer for the valuable feedback and constructive suggestions. Detailed responses to the individual comments are given below. Please see R3 for the response to the reviewer's concern about AMS collection efficiency.

Comments on content:

1. Line 12, p. 30532: polydispersed seed was used to 'minimize' (rather than 'eliminate') contribution of nucleation particles, so was nucleation observed in any of the experiments? If so, how do you think that affects the results?

[R1] No nucleation was observed in the experiments using polydisperse seed particles. The revised text reads, "*polydisperse seed particles were used … to prevent new particle formation …*"
          Nucleation was observed in some experiments that used monodisperse seed and were carried at initial β-caryophyllene concentration of 13.3 ppbv. Because the AMS detects particles having vacuum aerodynamic diameter (d_{va}) of 50 to 1000 nm (Liu et al., 2007), the particles formed by nucleation may not be detected by the AMS if they have smaller size. Figure S4 shows the cumulative particle number-diameter distributions measured by the SMPS. The contribution of particles having mobility diameter smaller than 40 nm (i.e., equivalent to 50 nm d_{va} assuming a spherical morphology and a density of 1.3) to the total particle number concentration is less than 15 %. The contribution of those particles to the total particle mass concentration (i.e., the reported results) is minimal (<<1%). To clarify this point, we added the following statement "…*The particle population largely had diameters within the AMS acceptance window of 50 to 1000 nm vacuum aerodynamic diameter. The error in M_{org} because of undetected particles is estimated to be much smaller than 1% based on the SMPS number-diameter measurements*..." in the revised manuscript.

2. Lines 1-5, p. 30534: as written, it sounds like the only way to get reliable yield data from b-caryophyllene+O3 (in excess) expts is by doing the expt in a CMFR. Isn't it true that if one waits long enough in a batch chamber with excess O3+b-caryophyllene, the final yield is practically what can also be obtained from a CMFR set up? If so, add a sentence to clarify this.

[R2] We thank the reviewer for the need for a clarification. In the revised manuscript, these comparative comments to batch mode are removed.

3. Line 16, p. 30534: Based on results from many lab and ambient studies, I'm very surprised that for lab generated SOA at RH=40%, CE=1 seems appropriate based on the SMPS and AMS closure. Have the authors seen this for any other SOA system? Can there be other errors (for example, errors in IE calibrations, losses in SMPS sampling line, errors in RH correction for SMPS-based mass before comparing to AMS mass (because AMS sees mainly dried particles while SMPS would see the water associated with the particles), differences in size range of two instruments, etc.) that would lead to the apparent CE to be 1?! How is the AMS-SMPS comparison for other systems?

[R3] We observed CE=1 for three SOA systems studied in the Harvard Environmental Chamber (HEC) including α-pinene ozonolysis, isoprene photooxidation, and β-caryophyllene ozonolysis, all at 40% RH with dry ammonium sulfate seed (Shilling et al., 2008; Chen et al., 2011). The AMS and SMPS comparison shows a slope of 1.0-1.2, suggesting CE=1 given the measurement uncertainty of 30% for the AMS and the SMPS. Other factors such as losses in SMPS sampling line (negligible), RH correction for SMPS-based mass (<10%), and differences in size range of two instruments (negligible) are consistent with CE=1. Our continued work in the HEC with additional experiments for α-pinene ozonolysis and isoprene photooxidation shows a good agreement (i.e., <20% difference) between the organic carbon mass concentrations measured by the AMS and those measured by a total organic carbon analyzer (TOC-Vws; SHIMADZU), also suggesting CE=1 for those laboratory-generated biogenic SOA particles. Similar higher values of CE were found in ambient for pristine Amazonian biogenic SOA particles (Chen et al., 2009).

4. Line 25, p. 30535: what resolution for AMS mass distributions was used?

[R4] The resolution for AMS mass distribution is determined by the AMS settings (e.g., number of spectra acquired per chopper cycle and number of spectra to co-add (DeCarlo et al., 2006)). It varies with diameter. For the mode diameters used in the density calculation of this study, the resolution is typically 10 to 20 nm (d_{va}). For clarification, the revised text reads, "The parameters used in the analysis included the uncertainty in the SMPS and AMS mode diameters (i.e., as needed for calculating ρ_{eff}) as well as the standard deviations of the temporal variation of $M_{particle}$, M_{org}, and M_{AS} at steady state." Figure S1 is also updated with markers representing the actual data points, which reflects the resolution.

5. Line 23, p. 30539: how is the lower-end estimate for τ(CMFR)/τ(exo) =0.1 determined? Is that basically just a number <<1.8?

[R5] The lower- and higher-end of $\tau_{CMFR}/\tau_{exo,i}$ for product i are based on one magnitude variation in reactivity while $\tau_{CMFR}/\tau_{exo}(50)$ of 1.8 represents the mean value.

6. Line 25-26, p. 30540: The argument that the RO2 channel wins over the O3 channel at higher O3 is counter intuitive since RO2 is secondarily produced while O3 is inherently higher at higher O3. Can you add some kinetic calculations to support your hypothesis? Also, can there be other losses of p238 at higher O3?

[R6] We thank the reviewer for bringing up this point of confusion. Criegee intermediate can decompose via the "hydroperoxide channel" to form an alkyl radical and subsequently RO_2 in the first-generation ozonolysis step. At higher O_3, the RO_2 concentration is greater (supported by our MCM-SIMPOL model results (Chen et al., 2011)), which may promote the subsequent conversion of the first-to-second generation products. To clarify this point, we revised the manuscript as follows, "*Another exception is P238, showing a negative relative change in mass concentration. This species is produced from the first-generation product P236 by a stabilized Criegee intermediate channel that competes with an RO_2-assisted isomerization channel (Fig. S6). At higher ozone concentrations, the importance of the isomerization channel possibly increases because of higher RO_2 concentrations, providing one possible explanation for the observed decrease in P238 concentration*."

We agree with the reviewer that kinetic calculations would be very helpful. Such theoretical calculation is, however, very difficult given the current knowledge of the reaction mechanism and is beyond our scope of this manuscript. Because of the use of OH scavenger, P238 is not expected to be further oxidized in our chamber experiments. Reaction pathways to remove P238 are not clear yet.

7. Line 28-29, p. 30540: It's not clear why "the nearly completion of P302 from P252-1 at 50 ppbv ozone" can explain lack of a significant change in P252-x??! Please explain better the relation between P302, P252-1 and P252-x. Also, as written, the section of the sentence I've quoted here in "" is not clear- do you mean 'nearly complete formation of P302 from P252-1'?!

[R7] We thank the reviewer for the need for a clarification. We added "*Labels x, y, and z represent a permutation of 1, 2, and 3.*" into the caption of Fig. 4. The text is also revised as follows, "*The relative concentration change of P252-x is negligible given the uncertainty. In this case, we can assign x = 1 because the second-generation product of P252-1 (i.e., P302) also changes negligibly but the products of P252-2 and P252-3 (i.e., P270-y, P270-z, P254-y, and P254-z) all show positive changes (Fig. S6)*."

8. Line 7-8, p. 30541: Is the evidence for P254-1 to be 2nd generation product the fact that its mass fraction goes up with increase in O3? If so, clarify that. If not, what is the evidence then?

[R8] Yes, resigning P254-1 as a second-generation product is based on the observed increase in mass concentration at 200 ppbv ozone. This point is clarified in the revised manuscript.

9. Lines 8-10, p. 30542: If the height of the green bars in Fig. 5 indicates the ultimate particle yield, it means that at Morg=1 ug/m3, yield is 10% regardless of the ozone level while at Morg=10 ug/m3 yield is 25-30%, and at Morg=100 ug/m3 yield is 50-80% depending on the excess ozone level. I think this is important to highlight since most ambient concentrations of Morg are ~1-10 ug/m3 and at these levels, it seems ozonolysis particle yield is not as high as one would think.

[R9] The reviewer brings up a good point. Following the reviewer's suggestion, this point is highlighted in the revised manuscript.

10. Line 23, p. 30542: With quantification of the 15 products identified by UPLC-EST-ToF-MS, can the authors estimate the yield of each product at different ozone levels, rather than speculating that P270-1 has a low mass yield?

[R10] Unfortunately we don't have reliable data to prove that P270-1 has a low mass yield. The 15 products identified by UPLC-EST-ToF-MS were semi-quantified by a surrogate approach, meaning that product concentrations were expressed on pinic-acid or pinonic-acid equivalency basis. Although the comparison of the relative change of each product is scientifically sound, the comparison among different products is risky. To avoid misleading, the speculation about P270-1 is removed in the revised manuscript. This section now reads, "*In this case, the mass yields are not sensitive to the ozone concentration. Product P302 is the lowest volatility product among the 15 identified products and can therefore be supposed as the dominant species to condense to the particle phase at low M_{org} (Li et al., 2011). This conclusion of a fast-forming low-volatility product is supported by the molecular data for P302 showing a negligible change in the mass concentration for an increase in ozone concentration (Fig. 4b).*"

11. Line 26-29, p. 30542: How can the authors be sure that it's 2nd generation products at C*=100 ug/m3 that partition to the aerosol phase and not further generation products? Since contribution of C*-class to particles goes up with increase in ozone, it is quite plausible to think that further oxidation of 2nd generation products leads to increase in C*-class in the particles. Please clarify.

[R11] Because of the use of OH scavenger in the experiments, further oxidation of second-generation products is not expected.

12. Line 4-14, p. 30544: I find the discussion in this section repetitive. Oxidation state and O/C levels give for the most part the same level of information. In page 30543, same conclusions about what type of products exist at different Morg and O3 levels were drawn from O/C measurements. I suggest combining these two sections by just mentioning the corresponding oxidation state for the measured O/C values in page 30543 and removing most of the discussion on page 30544.

[R12] Following the reviewer's suggestion, the discussion of O/C and oxidation state is combined in the revised manuscript.

13. Line 7, p. 30545: For clarity, I would say that organic mass 'inversely' correlates with oxidation state and material density.

[R13] This change is made in the revised manuscript.

14. Lines 14-30, p. 30545: since Griffin et al. chamber data are based on OH reactions and as the authors indicate on p. 30544 OH oxidation may move the products to those at lower volatilities and therefore increase the particle formation yield, I don't think it's appropriate to compare the current results with models that do use yield data of Griffin et al. An additional discussion here regarding this is needed. Also, for comparison, you may want to concentrate more on panel a since that's where most of the ambient Morg range is.

[R14] The yield parameterization based on the data of Griffin et al. (1999) has been widely used as a surrogate for β-caryophyllene oxidation in chemical transport models. Our data provide a new parameterization for representing the β-caryophyllene oxidation in the atmosphere. The purpose of comparing our data to Griffin et al. (1999) is to reveal the potential bias in current estimates because reaction with ozone is the dominant pathway of β-caryophyllene degradation in the atmosphere. To clarify this point, the revised paragraph reads, "*Using an updated ΔH_{vap} of 40 kJ mol^{-1} as well as a material density ρ_{org} of 1300 kg m^{-3} (Bahreini et al., 2005; Offenberg et al., 2006), Carlton et al. (2010) presented the state-of-the-art parameterization. The comparison between our data set and the parameterization of Carlton et al. (2010) suggests a possible underestimate by that parameterization of 100% to 300% for organic particle mass concentrations less than 3 μg m^{-3} given that ozonolysis rather than photooxidation is the dominant degradation pathway of β-caryophyllene. The difference could be in the underlying chemistry, keeping in mind that the parameterization is based on the photooxidation data set of Griffin et al. (1999) whereas the data of this study correspond to ozonolysis experiments.*"

15. Table 2: what is unique about expt 2?! The O/C of this expt is similar to many other expts while the density is significantly higher.

[R15] The cut-off organic mass concentration ((M_{org})$_{outflow}$) for the density analysis is 1 μg m^{-3} in this study. For Exp. #2, (M_{org})$_{outflow}$ of 1.1 μg m^{-3} is close to the cut-off loading. The reported density of 1810 ± 190 kg m^{-3} is associated with large errors. For comparison, Exp. #14 shows another example of low loading ((M_{org})$_{outflow}$ =1.3 μg m^{-3}) but with smaller seed particles, meaning greater change in mode diameter after coating with organic material. The reported density of 1520 ± 100 kg m^{-3} is therefore associated with smaller errors. The two values are nevertheless comparable given the errors.

Editorial Comments:

1. Remove 'nevertheless' in line 22 of abstract and line 8 of p. 30546.

[R16] The word 'nevertheless' is removed in the revised manuscript.

2. Figure 4 caption- it's important to clarify that the fractions are based on the mass of the 15 compounds that were identified (not all the organic mass). I suggest adding the

following: "(a) Mass fraction of first- and second-generation products as identified by UPLC-EST-ToF-MS for increasing ozone concentrations"

[R17] This change is made in the revised manuscript.

3. Figure 7- make the modeling lines thicker or the whole plot bigger- it's hard to distinguish some of them at the current size of the figure. Also, having the oxidation state here is confusing. Since O/C values are already plotted in Figure 6, I suggest adding the points in OS-space as the right-axis in Figure 6.

[R18] Figure 7 is revised based on the reviewer's suggestions.

Reviewer #2

This work investigates aerosol formation by beta-caryophylene ozonolysis, under conditions of excess ozone. By varying the level of excess ozone, distinction can be made between fast-forming low-volatility and slow-forming higher-volatility second-generation products contributing to the total mass yield. This is confirmed by analysis of the O:C ratio and density of the aerosol. A VBS model is derived that could be used in regional or global models.

The paper is well written and organized, scientifically sound and relevant. My main concern is that the large wall-losses could have an affect on the results. I recommend publication in ACP after the following comments are addressed:

[R0] We thank reviewer #1 for the constructive suggestions for improvement of our manuscript. Detailed responses to the individual comments are given below. Please see R2 for the response to the reviewer's concern about potential wall-loss effects on the results.

Remarks:
p. 30538. Given the high wall-losses that are encountered here, should it be recommended then that aged bags are used and/or reneutralization of seed particles is performed, for future experiments?

[R1] Yes, our results suggest that aged bags or neutralization of seed particles lead to smaller particle wall losses, at least for the 5-mil PFA bag that we used.

p. 30538 and Figure S5. The authors have made a thorough effort estimating the amount of SOA lost to the walls and have included this amount in the corrected wall loss yields, which as a consequence increase by a large factor. They show that wall losses are similar (and large) for the series of experiments conducted in the same time period in the same teflon bag. Still, the difference in amount of SOA wall loss between two experiments might also have an impact on the gas phase chemistry or on thermodynamic behaviour of the condensable products, which in turn might impact the SOA concentration. Partitioning to the liquid phase might be increased/decreased in an experiment in which wall losses are high compared to one in which they are low. Could the authors speculate on the possible consequences of such a difference in wall loss? The authors have already partly addressed this problem by indicating in red the two experiments for which wall losses were lower. There does not seem to be a trend obvious from these two experiments. Still, for the experiment 27, in which wall losses were five times lower, the wall loss corrected SOA yield is significantly lower (28.7%+-1.3%) than in the corresponding experiment 18 with higher wall losses (41.4+-3.6%). Also, compared to exp. 18, both the SOA density and the O:C ratio in exp. 27 are lower. This suggests an impact of wall loss on the chemical composition of the SOA. Therefore it seems that, only given the presented data, it is not yet an established fact that the presence of a large wall loss will not have a significant impact on the final corrected SOA mass which is obtained. This should be recognized in the main article.

The discussion on wall-loss focuses on the SOA. What about the wall-loss of gas-phase products? Is there evidence that this can be neglected? The work of Matsunaga and Ziemann (2010) shows that for Teflon walls, gas-wall partitioning can be very important and species-dependent. A discussion on the possible impact of wall-loss of gas-phase products, with reference to Matsunaga and Ziemann (2010), should be inserted.

[R2] We thank the reviewer for bringing up the need for further discussion. Our chamber was operated in a CMFR mode and data were reported for steady-state conditions. One advantage of the continuous-flow mode compared to the batch mode is that the wall-loss effect on the gas-phase chemistry and the absorptive partitioning of condensable products can be largely reduced.

During the spinning-up period of the CMFR, the gas-wall partitioning, the evaporation of the SOA particles deposited on the wall, and the absorption or reactive uptake of gas-phase products by the SOA particles deposited on the wall may affect the gas-phase concentration of condensable species and hence the gas-phase chemistry as well as the gas-particle partitioning. On the one hand, the reactive wall effects, if significant, can lead to continuous changes in the system, meaning no steady state can be achieved. For our experiments, steady-state periods were determined by the AMS, SMPS, and PTR-MS measurements. They were typically achieved 12 h later after the oxidation started and last for a couple days before stopping the experiments (Fig. 2). The measured quantities at steady state typically fluctuated within 5%, suggesting that the reactive wall effects were small. On the other hand, the non-reactive processes are expected in equilibrium at steady state. The work of Matsunaga and Ziemann (2010) shows that the gas-wall partitioning of various organic compounds was nearly independent of bag conditions (untreated or previously exposed to SOA) and reversible, achieving equilibrium with time constants of < 60 min. Our data also show that the steady-state β-caryophyllene concentrations measured by the PTR-MS prior to ozone injection agree with the calculated concentrations based on the injection rate and dilution (Fig. 2 at -1 to 0 h). Considering the wall is saturated at steady state, the gas-wall partitioning does not affect the gas-phase chemistry. And ideally, particles deposited to the wall have the same composition with suspended SOA particles for instantaneous gas-to-particle partitioning. Therefore, the species concentrations in the condensed phase for suspended SOA particles remain the same under different particle wall-loss conditions. Particle wall losses have minimal effects on the partitioning as well as the gas-phase chemistry at steady state.

Although most of our data correspond to large particle wall losses, the wall-loss rate was carefully determined as described in Sect. 2.4. There are two experiments carried with lower particle wall losses (Fig. S5). For Exp. #10, there is a good agreement between the data corrected by different wall losses. For Exp. #27, as the reviewer pointed out, the organic mass concentrations corrected by different wall losses differ by 30% (but by 100% for uncorrected data). The O:C ratio also differ by 17%. The differences are, however, within the measurement uncertainty. The IE calibration of the AMS and the O:C calculation are both associated with ~30% uncertainty. Repeated experiments under the same wall-loss condition also show differences of 0 to 30% (e.g., Exp. #2 and #12). Given all the facts, we must conclude that the presented data do not suggest the impact of particle wall losses on the SOA formation at steady state of the CMFR.

To reflect the above discussions, the work of Matsunaga and Ziemann (2010) is cited. The following sentences are added in the revised manuscript: "*Experiments carried out at repeated conditions show differences no more than 30%...*"; "*The measured quantities at steady state typically fluctuated within 5%...*"; *and* "*By establishing a steady state and thereby saturating the*

surface layers of the Teflon bag material (Matsunaga and Ziemann, 2010), CMFR operation also reduced the effects of wall interactions on reactive and non-reactive exchanges with gas-phase molecules, including the re-partitioning of condensable products (cf. further discussion in the Sect. A of the supplementary material)." Also, section A is added into the supplementary material for detailed information.

p. 30541, line 5-6. "Our data are consistent with the plausible suggestion that P254-1 has been incorrectly assigned as..." Which data? Simply the fact that P254-1 increases at higher O3 levels, or is there other data that suggest that P254-1 is second-generation?

[R3] We thank the reviewer for the need for a clarification. "*Our data*" is replaced by "*The increase of P254-1 in mass concentration at 200 ppbv ozone…*" in the revised manuscript.

p. 30542, line 17. "This conclusion is supported by...". This is only true if P302 has a dominant contribution and P270-1 a negligible contribution. Otherwise it does not prove that. Do you have an estimate of the contribution of the different compounds?

[R4] Unfortunately we don't have reliable data on the relative contribution of different compounds. Please see R10 in the response to reviewer #1 for more details.

end of p.30545: "Nevertheless, the comparison between our data set and the parametrization of Carlton et al. (2010) suggests a possible underestimate by that parametrization of 100% to 300% for organic particle mass concentrations less than 3 µg/m3." Availability of data in this lower SOA mass range is indeed a great improvement over an extrapolation from data at higher masses, as this might be responsible for a large uncertainty. However, when comparing SOA yields for beta-caryophyllene between the present study and the study of Griffin et al. (1999), it should be recognized in the article that the different photochemical conditions in these experiments might lead to different SOA masses. Although in the photo-oxidation experiment of Griffin et al. (1999) beta-caryophylene will probably react with ozone too, the primary oxidation products might also undergo reactions with OH instead of with ozone. The presence of NOx might also have a significant impact on SOA yields, although it is not clear whether for sesquiterpenes this might be a reducing influence, as for monoterpenes, or an increasing one (Ng et al., 2007). In the real atmosphere the secondary products might also partly react with OH instead of with ozone. Photolysis might also play a significant role. Therefore it is not yet certain whether the current parametrization for dark ozonolysis ageing experiments including an OH scavenger will exactly represent all aspects of beta-caryophyllene aging in the real atmosphere. The reader should probably be warned about this.

[R5] We agree with the reviewer that our parameterization might not be a perfect surrogate representing the degradation of β-caryophyllene in the real atmosphere because factors such as NO_x, OH pathways, and photolysis were not considered in our experiments. The potential impacts of these factors and the difference between Griffin et al. (1999) and our study are highlighted in the revised manuscript (see the last paragraph of Sect. 3 and the last two paragraph of Sect. 4).

Fig. 7, comparison to literature. There is a comparison with Lee (2006) and Griffin (1999), but not with Jaoui (2003), Grosjean (1993), or Hoffmann (1997), present in Table 1. Include the points or motivate why this is not appropriate.

[R6] Following the reviewer's suggestion, the discussion on those literature results is added in the last paragraph of section 3 in the revised manuscript.

Technical remarks:

p. 30539, line 15. Put spaces between "and" and inequalities. line 16-17: strange sentence construct. Probably "show" must be "showing". line 23: second "tau_exo(50)" should be "tau_exo(200)"

[R7] Those changes are made in the revised manuscript.

Table 1. "Huffmann" should be "Hoffmann"

[R8] This change is made in the revised manuscript.

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