We thank the reviewers for their comments on the manuscript *Primary to Secondary Organic Aerosol: Evolution of organic emissions from mobile combustion sources*. While all three reviewers found the research described in the manuscript to be topically relevant for publication in *Atmospheric Chemistry and Physics*, two of the three reviewers expressed significant concerns over the scope of the manuscript, noting that it was perhaps too narrowly focused and/or more suitable as an additional piece to another manuscript rather than as a stand-alone publication. In addition, all three reviewers were critical of the basis set model that was used to discuss SOA-POA mixing behavior.

As a result of the comments, we have overhauled the manuscript and made significant additions and changes. In this Author Comment, we first outline the proposed changes for a revised manuscript, followed by specific responses to the reviewer comments.

Outline of revised manuscript

In response to comments by Reviewers #2 and #3 that the original submission did not contain enough new insights or in-depth analysis to serve as a stand-alone manuscript, we have made substantial additions to the manuscript. The largest addition is the inclusion of data from 10 experiments with gas-turbine engines. One engine is a CFM56 that was mounted on a military aircraft, and the other is a T63 helicopter engine mounted in a test stand. As with the gasoline and diesel vehicles presented in the original submission, we have previously reported SOA formation from these engines, focusing specifically on SOA mass enhancement due to photo-oxidation of the dilute emissions (Miracolo et al., 2012;Miracolo et al., 2011). However, the AMS mass spectra of the SOA formed from these experiments have not been presented in detail before, and AMS data for the POA or SOA for the T63 engine have not been published previously.

The addition of these data enhance the manuscript in 3 specific ways: (1) we now consider a larger data set representing a more diverse group of emissions sources, (2) AMS data from the T63 engine are presented for the first time, and (3) the experiments now cover a wider range of VOC/NOx conditions, as many of the gas-turbine engine experiments were conducted under very high VOC (VOC/NOx > 10 ppbC/ppb-NOx).

The original manuscript made several qualitative comparisons between the mass concentration and mass spectrum of POA determined by PMF and other methods of determining the POA-SOA split such as the residual method. Reviewer #2 was critical of this qualitative treatment. As a result, the revised manuscript presents a more robust comparison of the PMF-derived POA mass spectrum to the observed mass spectrum of the POA prior to photo-oxidation as well as a direct, quantitative comparison between the mass concentration of POA determined by PMF and the residual method.

In response to comments from Reviewer #2, we have also expanded the comparison between the mass spectra of the PMF-derived POA and SOA factors to ambient HOA, SV-OOA, and LV-OOA factors. The original manuscript relied primarily on comparisons via scatter plots. The revised manuscript includes full mass spectral comparisons up to *m/z* 100 and discussion of specific *m/z* where there are major similarities or differences between the laboratory and ambient mass spectra.

In response to comments from Reviewer #2 and #3, we have included data from other instruments where appropriate. While this manuscript focuses primarily on analysis of AMS data, the Reviewers are correct in their criticism that omitting data from other instruments narrows the scope of the manuscript significantly, subtracts from the overall message of the paper, and essentially requires readers to read other papers from our group in order to fully understand these experiments.

In response to comments from all three reviewers, we have removed the basis set modeling and accompanying discussion regarding POA-SOA mixing. While the mixing behavior of different types of OA is an important and relevant topic, we agree with the reviewers that the analysis we presented in the original manuscript was ambiguous. Investigating POA-SOA mixing behavior is better studied through specifically designed experiments.

Response to comments

The original reviewer comments are in plain text. Our responses are in *italics*. In cases of long comments, we have parsed the comments and our answers to hopefully improve readability.

REFEREE #1

The discussion about the mixing behavior of POA and SOA seems a bit overreached. The wall loss corrected POA time trend shown in Fig 6 is very noisy.

We have removed the modeling section from the revised manuscript, as all three reviewers were skeptical of the conclusions drawn from it. In its place we have expanded the discussion of the mass spectra of SOA and POA factors, and expanded the comparisons between the laboratory-derived SOA and POA factors and ambient HOA, SV-, and LV-OOA factors.

Were the POA increases during the 1st hour statistically significant? What's the performance of the model for predicting SOA formation?

The model was not designed to predict SOA formation. The model was intended to investigate POA evaporation due to chemistry, changes in OA concentration (C_{OA}), and changes in temperature. This discussion has been removed from the revised manuscript.

In terms of wall loss correction, how was black carbon measured, what are the measurement uncertainties? It might be useful to provide error estimates for the wall-corrected POA data.

BC was measured using a 7-wavelength aethalometer. The text in the revised manuscript has been edited to indentify the aethalometer and other supporting instrumentation.

We have added an example of wall-loss corrected data to the revised manuscript. Figure 1c below (which is a revised version of Figure 1 from the manuscript) shows wall-loss corrected data to help clarify the discussion about wall loss corrections.

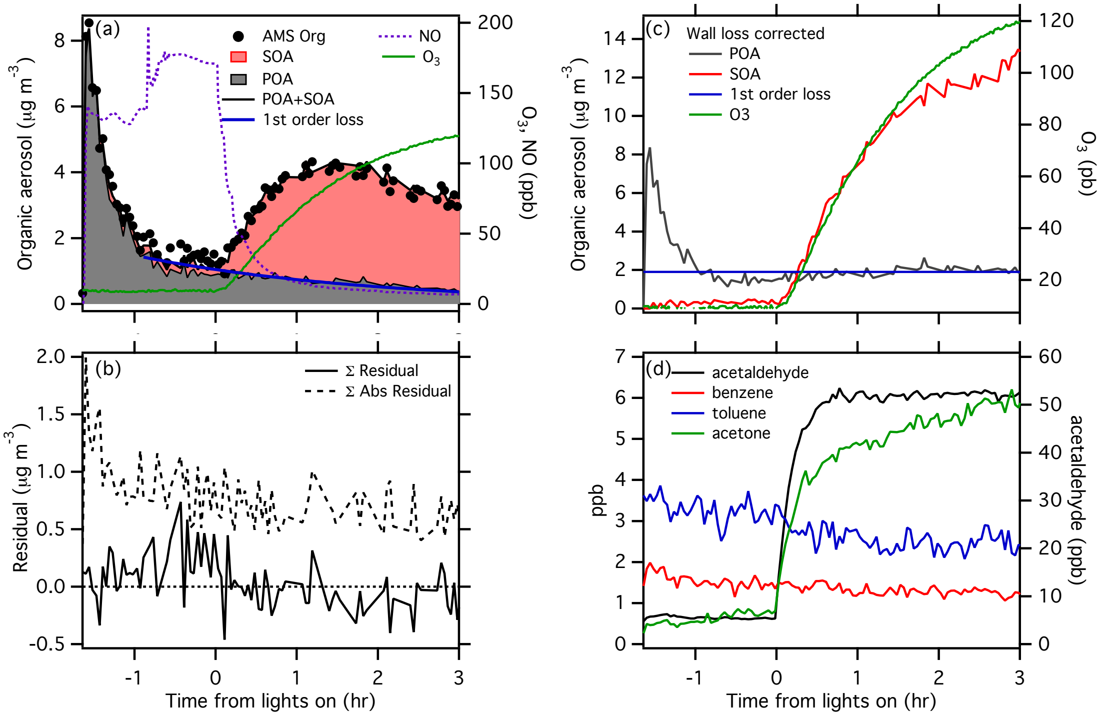

Figure 1. Revised version of Figure 1, showing a time series of a typical experiment (LEV1-5.2). Panel (a) shows data without correction for wall losses, and panel (c) shows data after wall loss correction. Diluted exhaust was introduced to the chamber starting at t=-1.4 hours (1.4 hours before the start of photo-oxidation). Chamber filling ended at t = -0.74 hrs. The POA was characterized for approximately 45 minutes prior to the start of photo-oxidation at t=0. The observed OA concentration increased after the start of oxidation, indicating production of SOA. The colors in (a) and (c) indicate the POA and SOA factors determined from PMF analysis. The blue line in (a) and (c) shows a predicted first-order wall loss rate for POA based on black carbon wall loss. Panel (b) shows both the residual and the absolute residual of the PMF solution. Panel (d) shows characteristic traces of VOCs as measured with a PTR-MS.

Some thermodenuder data are shown in Fig. 1, but no additional information is given. Nothing is known about TD temperature, residence time, etc. According to the TD data, roughly ¼ of the POA remained under heating (Fig 1). Yet, POA was found to evaporate completely according to some experiments. Are these results consistent?

The reviewer raises an excellent question about the TD data, especially for vehicle D4, where the POA completely evaporated during photo-oxidation. The complete evaporation of the POA is consistent with data recently published by May et al (May et al., 2013), who determined the volatility distribution of the diesel POA from these vehicles. Our thermodenuder model indicates that the POA should completely evaporate under relatively mild heating – 80^{o} C at a residence time of 10-20 seconds – suggesting that the POA is indeed semivolatile.

We have removed the TD-influenced data points in the revised manuscript (see Figure 1 above), as the TD data were not discussed in detail in either the original or revised manuscript.

Was the residual shown in Fig. 1b the simple sum of the residuals at all m/z’s? If so, the small values may not necessarily indicate good fit; they could arise simply because of the cancelling effect of adding up positive and negative values. It may be more appropriate to show the sum of the absolute values of the residuals or show the Q/Qexp time trend. In fact, I recommend the authors provide the diagnostic plots for the PMF work performed in this study.

The residual in Fig 1b was the simple sum of residuals. We have revised this figure to include both the simple sum of the residuals and the sum of the absolute values of the residuals, as per the reviewer's suggestion. We will also include the diagnostic plots in the online supporting information for the revised manuscript.

The *m/z* 44 signal in the D4 POA factor appears too high to be realistic. Is it possible that gas phase CO_2 subtraction was not done perfectly? It is particularly suspicious since the rest of the D4 MS after excluding m/z 44 and 18 peaks look very similar to the normal-looking POA MS.

We were also surprised by the high m/z 44 in the D4 POA, and investigated the CO_2 *subtraction. However, we observed the same high* f_{44} *in both experiments with D4. We also used the same* CO_2 *correction in all of the experiments. Specifically, we replaced 370 ppm* CO_2 *in the frag table with a time-dependent wave of measured* CO_2 *concentrations in the smog chamber. Given that the* CO_2 *concentration in the chamber was similar for all experiments after filling with dilute exhaust (600-800 ppm), it seems unlikely that there would be a systematic bias in the* CO_2 *correction for the two experiments with vehicle D4.*

The method used here to correct for CO_2 *was recently validated by Qi Zhang's group (Collier and Zhang, 2013). While there are some differences in the specific method – we replace the assumed* $CO_2(g)$ *concentration of 370 ppm in the frag table with the measured* CO_2*, whereas Collier and Zhang developed a relationship between the* CO_2^+/N_2^+ *ratio as a function of measured* CO_2 *– both methods serve to correct for particle-phase* CO_2^+ *by adjusting the fragmentation table to reflect elevated gas-phase* CO_2*.*

While the high f_{44} *in the D4 POA is surprising, it is not the first time that we or other groups have observed oxidized signal in POA. Aircraft emission POA (Presto et al., 2011) has* f_{44} *of a few percent; we show this in the revised manuscript with emissions from the T63 engine. Chirico et al (Chirico et al., 2010) observed the presence of oxidized fragments in POA from a diesel vehicle, that like D4, was equipped with a DOC. Collier and Zhang (2013) observed* f_{44} *of 0.09 for vehicles tested in the same facility as these experiments when the organic PM loading was 1-2* $\mu g\ m^{-3}$*. Therefore, while D4 seems to be an extreme case of POA containing m/z 44, we are confident in our result.*

The SOA spectra extracted from PMF in this study looks more different than ambient SOA spectra. But the SOA spectra of vehicle exhaust reported in Sage et al. based on an earlier work conducted in the same group are more similar to ambient SOA. Sage et al. also found the SOA MS in vehicle exhaust changed over the course of photo-oxidation, yet this study used PMF to extract the SOA spectrum, thus assumed that the SOA spectrum is fixed over time. The issue whether the SOA spectrum is indeed constant over time should be more thoroughly investigated. Was the approach of Sage et al. applied to the data of this study? Since Sage et al. assigned all m/z 57 to POA, which may not be valid, another approach to check this issue is to examine the difference matrix: diff_mx = mx_org – mx_POA = mx_SOA + mx_residual

The revised manuscript directly addresses the differences between POA and SOA factors determined from PMF and the residual method of Sage et al. Briefly, we find that for most experiments using 2 PMF factors (POA and SOA), the residual method also identifies an SOA mass spectrum with time-invariant f_{44}. However, when 3 PMF factors are required (POA and 2 SOA factors), which is the case for many of the gas-turbine engine experiments that we have added to our analysis, the f_{44} in the residual SOA changes with time, as in Sage et al (Sage et al., 2008).

Unfortunately we no longer have the Sage et al data and experimental details available for analysis in PMF, and therefore cannot make the direct comparison and verify that the Sage experiments would require multiple SOA factors. The revised manuscript investigates this issue for an experiment from a gas-turbine engine.

Two other potentially confounding factors between the diesel experiments presented here and those of Sage et al are (1) differences in engine-out emissions between the diesel generator used by Sage et al and the vehicles tested here and (2) a difference in SOA formation chemistry due to changes in the VOC/NOx ratio, which dictates radical reaction pathways. The diesel generator used by Sage et al has organic-rich emissions (OC/EC > 1), whereas most diesel engines have OC/EC <1. Additionally, we are unsure of the SOA chemistry in the Sage experiments. For example, nearly all vehicle experiments in this manuscript were tuned to VOC/NOx ~3. We do not have similar information for the Sage et al experiments, however based on the emissions from the diesel generator those experiments may have been conducted under high-VOC (VOC/NOx > 10) conditions unless additional NOx was added to the chamber. Photo-oxidation chemistry under these different VOC/NOx regimes would produce different mixes of product species, which in turn could alter the time trend of f_{44}.

Last paragraph, P24266, placing the in-text citations after "," is confusing.

This will be fixed in the revised manuscript.

P24270, 2nd paragraph, why heat to 47 C? What's the significance of this temperature?

Vehicle testing procedures in the U.S. are codified under CFR-1065 (CFR = Code of Federal Regulations). These regulations require that filter sampling be conducted with the filter media held at 47^o C. Therefore, we maintained the transfer line at this temperature as well, to maintain the same gas-particle partitioning between the dilute emissions sampled onto the filters and the dilute emissions entering the smog chamber. The revised manuscript has been edited to clarify this point.

P24270, L21, how much of the mass is BC?

We have revised this sentence as follows: "In some diesel tests, primary PM concentrations were as high as 80 $\mu g\ m^{-3}$, though 80-90% of this mass was black carbon."

P24271, L18, in addition to citing the paper, it is necessary to briefly describe the method used to apportion the nitrate signal.

We have made this change in the revised manuscript.

Fig1, why does the blue dashed line start around -1? Wall losses should be happening all the time when there are particles in the chamber.

The blue dashed line does not start until after the completion of the driving cycle. Most of the emissions occur during the "cold start" of the vehicle, which accounts for the first few minutes of the driving cycle. Therefore we observe an initial burst of emissions and then a gradual decay. During chamber filling, the observed decay in BC and OA concentrations is the result of both wall losses and dilution, with dilution dominating.

Therefore in Figure 1 we do not start the blue line indicating first-order loss until the end of the chamber filling period, because that is the point in time when wall losses, and not dilution, begin to dominate the observed decrease in OA concentration. We have made additions to the text of the revised manuscript to clarify this.

P24275, 2nd paragraph, the discussion of quantification limit needs clarification. How was the quantifiable limit determined, under what experimental condition? What's the averaging time for the quoted 0.3-0.5 value? The observation of 0.3 ug m^{-3} background OA signal in filtered air could be an indication that the AMS fragmentation table was not adjusted properly. Has this issue been examined closely? What does the mass spectrum of this background signal look like? Does it look like the MS of the SOA factor? If not, what's the base of estimating the detection limit of SOA according to this value? As shown in Fig. 1, small SOA signals began to appear at t = -0.5h, but the signal was zero earlier. If SOA is not expected to occur before t=0, have the authors tried to use fpeak to tune away the SOA signal before t =0?

We realize that this passage was confusing and have removed it from the revised manuscript. A more appropriate statement would have been that OA measured with a HEPA filter upstream of the AMS inlet ranged from -0.5 to 0.5 μg m^{-3}*, though the average OA concentration during the HEPA period was 0 μg* m^{-3}*, as one would expect.*

P24275, 3rd paragraph, it is strange that a 7% reduction of Q/Qexp led to almost no change in the residuals. It may be worthwhile to show in the supplementary material some PMF diagnostic plots. Showing the results from factor 3 may also be useful to compare with the 2 factor solution.

We have added diagnostic plots to the Supplementary Material. We will also add the results for the 3-factor solution to the Supplement.

P24275, line 27, "reduced peak" is vague. Please revise.

We have edited the revised manuscript to ensure that every reference to a "reduced" peak includes a note that this refers to peaks of composition C_xH_y*, with no oxygen present.*

P24277, L9, it is said "The number of PMF factors determined by Craven et al. is broadly consistent with the PMF analysis presented here.." What is consistent, the factor spectra?

The consistency is that Craven et al observe a single SOA factor for the first 5 hours of oxidation, and we observe a single SOA factor for the gasoline and diesel vehicle exhaust experiments. We have edited the revised manuscript to clarify this passage.

Fig. 4, how do the SOA spectra look like?

In response to this and other comments, we have revised the Figures to include more mass spectra of the SOA factors.

REFEREE #2

General Comments

In this manuscript, the authors report results of chamber experiments examining SOA formation from a fleet of both gasoline and diesel-engine vehicles. The authors apply PMF to their quadrupole AMS data to partition OA into a POA factor and an SOA factor. The authors compare the smog chamber PMF factors to ambient HOA and OOA data and they evaluate the mixing of their chamber OOA and POA using the PMF factors and a simple model. The authors reach 3 main conclusions. First, they argue that the PMF analysis improves upon previously described methods to separate SOA and POA in these types of chamber experiments. Second, they argue that the POA factor derived in their experiments strongly resembles the ambient HOA while the SOA factor derived in their experiments does not resemble either ambient SV- or LV-OOA factors. Finally, they argue that the experiments provide evidence of mixing between SOA and POA.

The general topics of SOA formation from diesel or gasoline vehicle emissions and conversion of POA to SOA are certainly relevant and of interest to the readership of ACP. In this manuscript, the authors focus exclusively on analysis of quadrupole AMS data, particularly on PMF analysis of the quadrupole data. No other data is shown. It appears that at least 5 other manuscripts have been written detailing other aspects of the same set of experiments (Gordon et al 2013 a, b; May et al 2013 a,b,c). Therefore, the additional data that one would typically expect to find in a manuscript describing these types of experiments are missing, presumably because they are presented in the other papers (most of which are also in the review process).

We appreciate the reviewer's sentiment that our exclusive focus on AMS data made the scope of the manuscript too narrow. As noted above, the revised manuscript includes data from these supporting instruments. We feel that the inclusion of this data improves the manuscript, and helps to strengthen it as a stand-alone journal paper.

The general topics of SOA formation from diesel or gasoline vehicle emissions and conversion of POA to SOA are certainly relevant and of interest to the readership of ACP. In this manuscript, the authors focus exclusively on analysis of quadrupole AMS data, particularly on PMF analysis of the quadrupole data. No other data is shown. It appears that at least 5 other manuscripts have been written detailing other aspects of the same set of experiments (Gordon et al 2013 a, b; May et al 2013 a,b,c). Therefore, the additional data that one would typically expect to find in a manuscript describing these types of experiments are missing, presumably because they are presented in the other papers (most of which are also in the review process).

The results described in this manuscript would potentially make a nice addition to a larger paper. However, as it reads now, I don't think this manuscript meets the criteria for publication as a stand-alone paper. The scope of the work is very narrow and the conclusions are not sufficiently supported by the data, scientifically novel, or well-developed for this work to stand on its own. As the authors point out, a number of studies have previously reported on SOA formation from dilute vehicle exhaust and it isn't clear what new insights this manuscript provides. I encourage the authors to consider combining the results presented here with some of the other data to make a more robust, novel, and substantive manuscripts. Alternatively, the present manuscript should be substantially revised before publication to 1) further substantiate their conclusions and 2) expand on the presented material and discussion so that is more clear what this study adds to the scientific understanding of the topic and how this study improves upon or is different from previous studies. More specifics are below.

As noted above in our outline for the revised manuscript, we have expanded the original manuscript by including data from experiments conducted with gas-turbine engines. These additional experiments give us a larger data set to present, and cover a wider range of sources and VOC/NOx ratios. Additionally, POA and SOA mass spectra for the experiments with the T63 engine had not been published previously, and are therefore a unique contribution of the revised manuscript.

Major Specific Comments

As the authors point out, there have been a large number of chamber studies of SOA formation from vehicle emissions, many of them from their group. There is certainly novel and exciting science that has and will continue to come out of this type of work. In this manuscript, the authors present a narrow slice (PMF analysis of quadrupole AMS data) of data from what appear to be well-conducted experiments. The difficulty I have with this paper is identifying what new scientific insights this paper reports. The authors have 3 major points, but I don't find any of these sections sufficiently well developed to warrant a stand-alone publication.

First, the authors make the argument that the PMF method is an improvement over past approaches to separate POA and SOA in these types of experiments. However, they don't show a comparison of their method with previous methods. Furthermore, the majority of the figures show data for which POA does not appear to evaporate and is well described by a simple first-order wall loss. To improve this section, the authors could present some comparisons of the PMF-derived POA and SOA time traces and spectra with similar data using previous methods to show that this method is better and how/why. They could also extend their analysis to data where the POA was observed to evaporate, though the experiment where this was observed appears to be an outlier. In short, the benefits of the PMF analysis are not clear. More comments follow below specific to the figures.

We have followed this suggestion and now make a direct comparison between the mass spectra of POA determined by PMF and the mass spectrum of the POA measured prior to photo-oxidation. One advantage of our experimental design is that we observe POA prior to SOA formation, and can therefore directly compare the measured POA mass spectrum to the mass spectrum of the PMF POA factor. In the revised manuscript we use this mass spectral agreement as a screening tool for determining the number of factors used in the PMF solution. For example, in experiments conducted with the

CFM56 engine, a 2-factor solution is selected over a 3-factor solution because of better agreement between the observed and PMF-derived POA mass spectra for the 2-factor solution, as shown below.

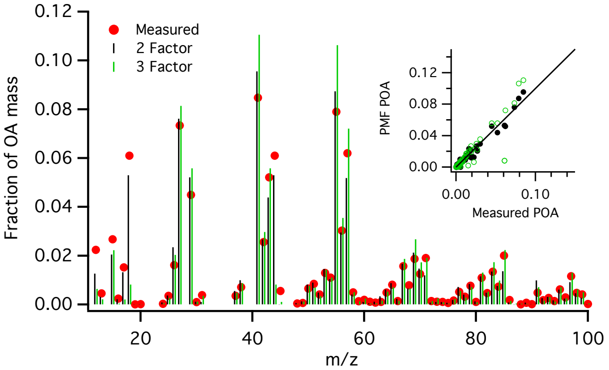

Figure 2. Figure 5 in the revised manuscript. Mass spectra of the measured POA (red circles) and PMF POA factors for 2- and 3-factor solutions for an experiment with the CFM-56 engine operating at idle load. The POA factor for the 2-factor solution clearly outperforms the 3-factor solution in matching the observed POA mass spectrum, and therefore was chosen. The inset shows a scatter plot of the measured versus PMF-derived POA mass spectra; the black line is the 1:1 line.

Second, the authors make comparisons of the SOA and POA factors from their chamber experiment to ambient data. They conclude that the diesel SOA does not resemble either ambient SV- or LV-OOA. This contrasts earlier work from the same group (for example Sage et al 2008). The authors offer a few short explanations of why this might be the case, but quickly rule them out. It would improve the paper if the authors could offer some reasonable explanation for the difference. I have further technical questions about this comparison detailed below. However, the actual differences between the ambient OOA spectra and the chamber-derived SOA spectra are never presented or discussed, beyond the correlations. I suspect significant insight could be gained from examining the differences in the spectra.

We have followed this suggestion and significantly expanded the comparisons between the laboratory and ambient PMF factors in the revised manuscript. This expansion includes direct comparisons in the text of m/z that are similar or different between the laboratory and ambient factors as well as figures that directly compare the factor mass spectra (rather than just scatter plots). One example of a mass spectral comparison is shown in Figure 2 above for different POA factors; the revised manuscript includes similar comparisons of SOA factors against SV- and LV-OOA and POA factors against ambient HOA.

We have also added the ranges of ambient HOA, SV-, and LV-OOA factors defined in Ng et al (Ng et al., 2011) to the triangle plots to facilitate comparisons. Ambient SV- and LV-OOA factors are highly variable from location to location. An example is shown in Figure 3 below.

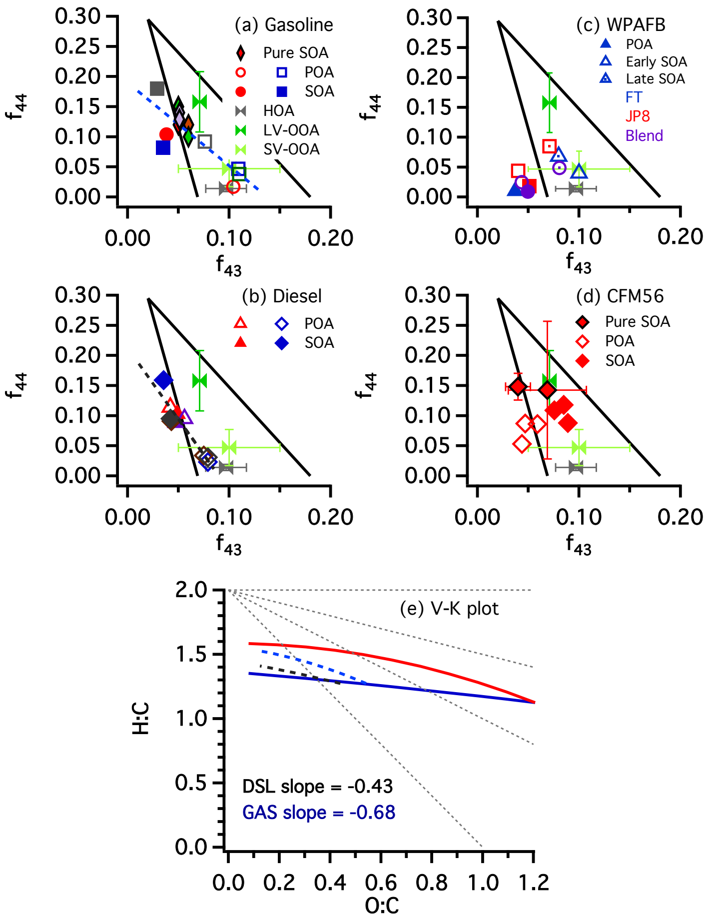

Figure 3. Figure 9 in the revised manuscript. Triangle plots are presented for POA and SOA factors for all of the sources considered (gasoline vehicles, diesel vehicles, CFM56 engine, and T63 engine) in panels a-d. Also shown are the mean (points) and standard deviations (error bars) for ambient HOA, SV-OOA, and LV-OOA factors reported by Ng et al (2011). Panel e shows the gasoline and diesel data translated into a van Krevelen plot.

Finally, the authors use the PMF factors and a simple model to infer the miscibility of SOA and POA. The analysis here doesn't seem very robust. First the data are very noisy and appear to agree with either model assumption within the measurement noise. Second, the model is very simple and has many assumptions that aren't well constrained but could change the predictions (for example the POA heat of vaporization and/or mass transfer limitations induced by low-viscosity SOA (Vaden et al., 2011)).

Finally, I'm not clear on how the PMF, which is the main data reported in this work, improves the quality of the POA/SOA apportionment. Asa-Awuka et al (2009) provide a much more thorough, robust, and convincing demonstration of POA/SOA mixing state (Asa-Awuku et al., 2009). It isn't clear that the present work is an improvement upon Asa-Awuku or presents new or different results. More specific comments on figure 6 are below.

As noted above, we have removed the modeling and POA/SOA mixing discussion. POA/SOA mixing is an important question, and should be addressed more fully with specifically designed experiments.

P24279,L16-30. The relationship between m/z 44 and O:C described in Aiken et al (2008) is based on ambient data. If the chamber SOA spectra really aren't similar to ambient OOA spectra the Aiken parameterization may not work well. Similar to additional comments elsewhere, it would improve the paper if the authors expand the discussion of the differences between the ambient and chamber SOA spectra.

We disagree with the characterization that the Aiken et al relationship between f_{44} *and O:C is based entirely on ambient data. Figure 1 in Aiken et al (Aiken et al., 2008) shows that the elemental analysis method was calibrated using laboratory-generated OA (fulvic and amino acids).*

The resulting elemental analysis was then applied to data collected in Mexico City. This ambient data was then used to develop the relationship between f_{44} *and O:C (Figure 4b of Aiken et al). Therefore the* f_{44}*-to-O/C relationship is more of a hybrid of laboratory and ambient data than purely derived from ambient data.*

In addition, there is significant scatter in the O/C vs f_{44} *plot shown in Figure 4b of Aiken et al (2008). While the SOA observed in these experiments does not look like "canonical" SV- or LV-OOA, it is unlikely that the experimental data fall entirely out of the bounds of the* f_{44}*-to-O/C relationship presented by Aiken et al. We use this conversion as a screening tool to evaluate whether the OA chemistry observed in these experiments is similar to the OA chemistry observed in the atmosphere, and conclude that for the gasoline and diesel vehicles, it is. We think that this is a reasonable and responsible use of the* f_{44}*-to-O/C conversion, and stand by our result.*

P24275, L3-9. The fact that the AMS measures 0.3-0.5 ug/m3 of organic aerosol when behind a filter suggests the frag table wasn't adjusted properly. Were adjustments made to the frag table based on the filter measurements? If the frag table isn't adjusted properly, it could bias the SOA spectra. It could also explain why the SOA factor doesn't appear to agree with the ambient data.

Please see our response to Reviewer #1 on this issue. The original passage was poorly worded and has been changed in the revised manuscript. The average OA mass measured by the AMS when a HEPA filter is placed in line is 0 $\mu g\ m^{-3}$*, as expected.*

Figure 1: The POA concentration appears to be well-modeled by simple wall loss in this experiment. Will the PMF work as well when POA is lost by evaporation? How does the residual change when the data is more complicated?

The revised manuscript includes a direct comparison of POA determined by PMF to both the residual method and first-order wall loss, including cases when POA evaporates and when POA mass increases due to partitioning effects at high C_{OA}*.*

PMF is used to identify the experiments when the POA evaporates. This phenomenon is obviously not predicted by first-order loss of POA or by the residual method, which relies on a single m/z as the POA tracer.

The revised version of Table 1 in the manuscript includes the difference between PMF-derived and residual POA mass concentrations at the conclusion of photo-oxidation. For every experiment considered here, the residual method predicts a higher POA concentration. This is a result of assuming that all m/z 57 is in the POA, with none in the SOA.

Figure 2: Please provide a short description of what exactly is plotted in the correlation plots. The scatter in the SOA correlation plot looks to be dominated by 2 m/z values. What are those m/z's? Is it possible these differences are a result of different frag tables that were used to process the data at m/z 28, 18, etc? It is clear that m/z 28 is not set equal to m/z 44 in the chamber SOA spectrum, which is a common practice in ambient data sets. The manuscript would benefit from a more thorough discussion of the difference between the chamber and ambient OOA factors.

These scatter plots have been changed to complete mass spectral comparisons in the revised manuscript. As the Reviewer noted, there is too little detail when only scatter plots are presented.

In response to this comment we have expanded the explanation of our choice of fragmentation table in the Experimental section of the revised manuscript. As discussed in the experimental section of the original manuscript, there is no evidence of m/z 28 in the OA in these experiments, and therefore we use the fragmentation table of Allan et al (Allan et al., 2004) to apportion OA mass. We verify this through analysis of the pTOF data as described by Grieshop et al (Grieshop et al., 2009) and by examining the time series of the airbeam strength. When OA is formed with m/z 28 in these types of smog chamber experiments, there is an unambiguous increase in the airbeam strength that coincides with an event in the experiment (e.g., introduction of exhaust or the start of photo-oxidation) and/or particle signal at m/z 28 in the pTOF data.

The HOA and OOA reference spectra used in this manuscript also set m/z 28 equal to zero for unit mass resolution data. This is true for both the Zhang et al (Zhang et al., 2005) OOA and HOA and the Ng et al (Ng et al., 2011) HOA, SV-OOA, and LV-OOA. We refer the reviewer to the online AMS spectra database for confirmation: http://cires.colorado.edu/jimenez-group/AMSsd/#AmbientDecon

Figure 4: The authors state there is high correlation between gas SOA and diesel SOA (m= 1.11, R2 = 0.94) and gas POA and diesel POA (m= 1.11, R2 = 0.94) yet poor correlation between gas SOA and ambient LV-OOA (m=0.74, R2=.93). The R2 is similar in all cases and the deviation of the slopes from 1 is reasonable in all cases, especially considering the uncertainties. Please expand on why you conclude the correlation is good in two cases, but poor in the third. The high R2 but non-zero slope implies to me that differences in frag tables such that f_m/z is changed equally across the entire spectrum. This could happen for instance by setting m/z 28 = m/z 44 in one case but not the other.

As noted above, we have amended the figures to provide direct spectral comparisons between laboratory and ambient factors. We have also added discussion of specific m/z in the text. However, as noted in the comment on Figure 3, any differences between spectra are not a result of differences in the frag table.

Regarding Figure 6: 1) I don't understand the use of data from test D5.2 in this figure. Figure 5a shows that the POA doesn't evaporate and is well-modeled by a simple first order wall loss. If this is the case, what is the added value of the PMF? It seems PMF is not needed to separate SOA from POA. If instead, the point is to validate the PMF method, shouldn't PMF be challenged with a more difficult experiment? I suggest adding a second panel to Figure 6 showing the same techniques applied to the data from case D4.1or similar.

2) The POA data are very noisy, particularly near the end of the experiments where the models diverge do a significant degree. Why are the data so noisy? The noise appears to be are on the order of 2-4 ug/m3. Based on the noise, it is easy to imagine either model fitting the data adequately, especially considering all the uncertainties with the model. Please add errors bars to the POA trace. Are the experiments sensitive enough to the effect of mixing to detect a different in the models? Are there other cases where the models diverge to a larger degree relative to the noise the authors can show?

3) The "no chemistry" case suggests that the temperature increase had a substantial effect on the observed POA, relative to the difference between the models. If a larger or smaller temperature correction is applied, how does the data compare to the models? What is the estimated uncertainty in the temperature correction? Can the authors show a simple sensitivity analysis of the temperature correction?

We have removed Figure 6 and associated discussion.

Minor Comments and Technical Corrections

P24286, L20: I suggest changing this to "the answer to the first question may be yes" or something similar. The author points out several conditions where the factors are not physically interpretable.

This paragraph refers to PMF factors determined from ambient data sets, but perhaps this was unclear. It has been clarified in the revised manuscript.

P24268, L14-17. Please clarify where the results of Asa-Awuka et al (2009) and Song et al (2007) are similar and where they contrast (Song et al., 2007;Asa-Awuku et al., 2009). Both came to similar conclusions about the mixing of SOA with lubricating oil particles. Asa-Awuka et al (2009) observe that diesel POA and SOA do mix while Song et al. do not investigate diesel POA. Both papers are in agreement when comparing similar systems.

This discussion has been removed along with the model, Figure 6, and associated discussion.

P24270, L27. How was the total VOC concentration measured?

Total VOCs in the chamber were calculated from the non-methane VOC measurements conducted in the CVS, correcting for dilution based on CO_2 measurements in the CVS and smog chamber. We have amended the text accordingly.

P24272, L11. Please clarify what you mean when saying there was no evidence of organic particle signal at m/z 28. The quadrupole doesn't have sufficient resolution to separate CO+ and N2+. The N2+ signal will be so large that it will completely overwhelm any signal at m/z 28 from the modest amounts of SOA formed in these experiments. Most experiments with SOA find significant particulate CO+, to the extent that the "default" frag table has been modified such that org_28 = org_44 (Aiken et al., 2008). Furthermore, the authors argue that oxidation chemistry is proceeding via a combination of carboxylic acid and alcohol/peroxide formation. Studies have shown that organic acids produce signal at m/z 28. So what is different about this SOA?

We use the methods presented in Grieship et al (2009) to investigate particle signal at m/z 28. While the reviewer is correct in noting that the quadrupole cannot resolve CO^+ *and* N_2^+*, it is possible to determine increases in particle-phase signal at m/z 28 by monitoring the airbeam strength. This is especially true in these sorts of experiments where we have defined time points that would be expected to correlate with particle signal at m/z 28, e.g., introduction of emissions to the smog chamber or the start of photo-oxidation.*

Particle signal at m/z 28 would also be apparent in pTOF mode. In the experiments presented here, we have demonstrable pTOF signal at m/z 44, 43, 57, etc., but none at m/z 28, consistent with our choice of fragmentation table.

The CMU group has conducted numerous SOA formation experiments using a variety of sources that are relevant to this manuscript:

- *Gasoline and diesel vehicle emissions (e.g., this manuscript)*
- *Gas-turbine engine emissions (included in the revision to this manuscript) (Miracolo et al., 2011)*
- *Biomass burning emissions (Hennigan et al., 2011)*
- *Low volatility SOA precursors (Miracolo et al., 2010;Presto et al., 2010)*

In the majority of these cases, there is no evidence of particle-phase m/z 28 in the SOA; the lack of particle-phase m/z 28 is not an unusual finding. In addition, as noted above, the ambient SV-OOA and LV-OOA factors (Ng et al., 2011) do not apportion mass at m/z 28 either.

P24271, L13- 14. If a minor fraction of the nitrate signal is organic nitrate, how is the inorganic nitrate condensing on the particles? The vapor pressure of nitric acid is much too high for condensation (Duisman and Stern, 1969) and it is unlikely that a few monolayers of adsorbed nitric acid would be detectable by the quadrupole AMS. Do the authors have excess ammonia in the chamber? Do you see evidence of ammonium nitrate formation?

We do not have direct ammonia measurements in the smog chamber, but there is clear evidence of ammonium nitrate formation during photo-oxidation. We can confirm the nitrate as inorganic in two ways: via the ammonium/nitrate ratio and the ratio of m/z 30/46. The thermodeunder also provides anecdotal evidence that the nitrate is inorganic, as it rapidly evaporates at low temperatures in the TD.

P24284, L 9-10. PMF wasn't performed on the SOA only experiments, so please be specific on what you mean when you say they "produced a single SOA". The wording implies to me that you only extracted one PMF factor.

We have clarified this passage.

Figure 1: I suggest removing the thermodenuder data to make the figure less confusing, since it isn't used or discussed in the manuscript.

We have done this. A more complete description is provided above in responses to reviewer #1 and in Figure 1.

Figure 4. The small panels are very difficult to read and are likely to be even smaller in

the final printed version. I suggest splitting F4 into two figures to improve legibility.

Figure 4 has been reformatted, and the small panels have been removed.

REFEREE #3

This work used PMF analysis to determine separate factors from the organics formed in the photooxidation of diluted gasoline and diesel exhaust in chamber experiments.
In most cases, two factors were obtained: POA and SOA. It was found that the POA mass spectra are similar to ambient HOA, while the SOA mass spectra are markedly different from ambient OOA factors. The authors did not observe substantial change in f44 with aging and suggested that only the first generation oxidation chemistry was achieved in these experiments. The authors also attempted to use the PMF factors and a basis set model to infer mixing of POA/SOA, where they concluded that in most cases the POA and SOA seem to mix and form a single phase.

I think the manuscript is well-written overall, but I found it lacking substantial new insights.

As noted above in our outline for a revised paper, we have included additional data and analysis in the revised manuscript. This includes AMS analysis of experiments using gas-turbine engines, which has not been previously published.

This work is similar to a previous publication by the same group (Sage et al.), one of the main differences was that PMF was used in this analysis while residual spectrum method was used in Sage et al. The authors pointed out that the assumption in Sage et al. where all the OA mass at m/z 57 was attributed to POA could lead to an erroneous split between the POA and SOA mass in that study. However, it appears in this work that all the OA mass at m/z 57 is associated with the POA factor (e.g., Fig. 2) and the assumption made in the residual method in Sage et al could be appropriate.

The revised manuscript includes a quantitative comparison between PMF and the residual method, and we directly discuss the implications of using m/z 57 as the sole POA tracer. The SOA factor being devoid of m/z 57 was not a feature of all experiments presented in the manuscript, and this is apparent in the figures and mass spectra used in the revised manuscript.

Further, the authors observed some differences in this work and Sage et al. but did not provide sound explanations regarding these differences. Hence, it is not clear what the causes of such differences are.

The revised manuscript addresses these differences directly.

The authors used the PMF factors and a basis set model to infer the mixing of POA/SOA. The analysis could potentially be interesting. However, I do not think that their conclusions are well-supported by the data. Neither results from "ideal mixing" nor "no mixing" seem to fit the data particularly well given the noisiness in the data.
This work belongs to a large body of publications from the same set of experiments.

As described above, we have removed this section from the revised manuscript. The issue of POA/SOA mixing is important, and warrants more careful study in experiments specifically designed to probe this mixing behavior.

While the analysis was carefully done and well-presented in general, this work does not appear to offer substantial new insights to warrant a separate publication. I do not recommend publication at this stage. I suggest the authors either incorporate the results from this work into another manuscript to provide a more detailed, in depth analysis of the results from these experiments, or, they will have to expand this work substantially, and at the same time offer and emphasize the new scientific insights in addition to what are known from their previous publications.

We feel that the additional data and analysis, as described above in the outline for a revised manuscript and in the responses to Reviewer 2, address this comment.

Specific comments:

1. Page 24266, line 17. It should be f44 vs. f43.

This has been fixed.

2. Page 24267, lines 6-7. The authors stated that Sage et al. demonstrated that the mass spectra of SOA formed from photooxidation of dilute diesel engine exhaust are similar to ambient OOA. In this regard, it seems that the results from the current study are not consistent with Sage et al. The authors need to comment on this, given both the Sage et al. study and the current study are from the same group.

We have revised the manuscript to address this comment. Please see our response to Reviewer #1 above for additional detail. Two possibilities for the differences between the experiments presented here and the Sage et al experiments are (1) differences in emissions from the diesel engines, with the laboratory-scale engine used by Sage et al being more organic-rich, and (2) differences in chemistry due to the VOC/NOx ratio, with presumably higher VOC/NOx in the experiments of Sage et al.

3. Page 24270, line 22. How are the VOCs measured and what are the specific VOCs identified?

Total VOCs in the chamber were calculated from the non-methane VOC measurements conducted in the CVS, correcting for dilution based on CO_2 *measurements in the CVS and smog chamber. The manuscript has been edited to explain how VOCs were measured in the chamber.*

4. Page 24270, line 27-28. The authors wrote that VOC:NOx ratio was adjusted by adding propene to investigate the effect of VOC/NOx ratio on aerosol formation. I do not understand how adding propene can provide useful information regarding the effect of VOC/NOx ratio on aerosol formation from the primary vapors. Please explain further.

Propene was added to adjust the VOC/NOx ratio to 3 ppbC/ppb-NOx in nearly all experiments with gas and diesel vehicles to ensure similar radical chemistry. We will clarify the discussion of VOC/NOx in the revised manuscript. However, the effect of VOC/NOx on SOA mass formation (or yield) is beyond the scope of this work.

The authors noted that individual VOCs in the chamber were typically less than 1ppb. This would seem to suggest that propene will be the dominant precursor hydrocarbon. The radical chemistry in the system will then be dominated by peroxy radicals from the photooxidation of propene. How can this provide any insights into the aerosol formation from the vehicle exhaust (either with or without POA)? Further, the authors did not actually discuss the effects of VOC/NOx in the subsequent parts of the manuscript.

Propene is commonly added for VOC/NOx conditioning (Chirico et al., 2010) because it does not form SOA itself. The goal of adding propene to the chamber is to ensure a similar VOC/NOx ratio for nearly all experiments, so that any differences in SOA formation between vehicles or vehicle classes is the result of changes in emissions rather than a result of different radical chemistry (e.g., nitrate versus peroxide formation).

5. Page 24275, line 6. The authors noted that AMS still has 0.3 ug/m3 of organics with a HEPA filter upstream. This would seem to suggest that either the HEPA filter was not working or the authors did not adjust the fragmentation table correctly, or there was something else that was wrong with the instrument. If it is the fragmentation table, it could potentially affect all the mass spectra presented in the manuscript as well as the subsequent discussions/conclusions.

Please see our response to Reviewer #1 and #2 above on this question.

6. Page 24276, line 19. The authors should also show the mass spectra of the "pure SOA" systems. Are those mass spectra similar to the SOA factor shown in Fig. 2?

The revised manuscript will include this mass spectrum.

7. Page 24276, line 27. The authors should show the mass spectrum of the nitrate factor. It is unusual that almost half of the OA mass is at m/z 30. What is the source of this factor?

We are unsure of the source of this factor, and it is difficult to investigate further because it only occurs in one experiment. The revised manuscript will include the mass spectrum of the nitrate SOA factor in the Supplemental Information.

8. Page 24277. The authors offered two explanations regarding the differences in the change of the mass spectra with further oxidation in Sage et al. and the current study. Neither explanation is sound. The authors discarded their first explanation and offered an alternative explanation that perhaps the difference could arise from the apportionment of all OA mass at m/z 57 to POA in Sage et al. However, from Fig. 2a it appears that all the OA mass at m/z 57 is assigned to POA in the current study as well. As noted in point #3, the authors need to reconcile the differences between this work and their previous study.

The revised manuscript includes a more quantitative comparison between PMF and the residual method. Briefly, we find that for most experiments using 2 PMF factors (POA and SOA), the residual method also identifies an SOA mass spectrum with time-invariant f_{44}. *In these cases the two methods are in agreement. However, when 3 PMF factors are required (POA and 2 SOA factors), which is the case for many of the gas-turbine engine experiments that we have added to our analysis, the* f_{44} *in the residual SOA changes with time, as in Sage et al (Sage et al., 2008).*

Unfortunately we no longer have the Sage et al data and experimental notes available for analysis in PMF, and therefore cannot make the direct comparison and *verify that the Sage experiments would require multiple SOA factors. The revised manuscript investigates this issue for an experiment from a gas-turbine engine.*

Two other potentially confounding factors between the diesel experiments presented here and those of Sage et al are (1) differences in engine-out emissions between the diesel generator used by Sage et al and the vehicles tested here and (2) a difference in SOA formation chemistry due to changes in the VOC/NOx ratio, which dictates radical reaction pathways. The diesel generator used by Sage et al has organic-rich emissions (OC/EC > 1), whereas most diesel engines have OC/EC <1. Additionally, we are unsure of the SOA chemistry in the Sage experiments. For example, nearly all vehicle experiments here were tuned to VOC/NOx ~3. We do not have similar information for the Sage et al experiments, however based on the emissions from the diesel generator those experiments may have been conducted under high-VOC (VOC/NOx > 10) conditions unless additional NOx was added to the chamber. Photo-oxidation chemistry under these different VOC/NOx regimes would produce different mixes of product species, which in turn could alter the time trend of f_{44}.

9. Page 24278, line 6. It should be f44 vs. f43.

We have made this change.

10. Page 24279, line 24 The slope of -1 in Heald et al. is for all ambient data (including different OA types) while the slope of -0.5 in Ng et al. corresponds to OOA components from the ambient data. This should be clearly stated when citing these two publications.

We have made this change in the revised manuscript.

11. Page 24279, line 28. The authors wrote that the Van Krevelen diagram results suggested that the SOA chemistry observed in gasoline vehicle experiments is atmospherically relevant. However, earlier they emphasized that the SOA mass spectra for the vehicles are markedly different from ambient OOA factors. These two statements seem contradictory. If the oxidation chemistry in their experiments is similar to ambient environments, shouldn't one expect the mass spectra to be similar as well?

We disagree – these statements are not contradictory. The van Krevelen diagram provides information about the overall chemical processing of the emissions during photo-oxidation. In the gasoline and diesel experiments, the van Krevelen slopes suggest a combination of acid and alcohol/peroxide formation.

The SOA mass spectrum, on the other hand, is a function of several potential factors, including chemical processing, the composition of the SOA precursors, and potential instrument-to-instrument variations in AMS fragmentation patterns. A valid interpretation of the SOA in these experiments not "looking like" ambient SV- or LV-OOA is that the mixture of SOA precursors in these experiments does not match the exact mix of SOA precursors in the ambient atmosphere.

Furthermore, both Reviewers #2 and #3 appear to be of the opinion that SV-OOA and LV-OOA have fixed, static mass spectra that are consistent across many locations. This is hardly the case. Ng et al (2011) show that there is substantial variability in the SV-OOA and LV-OOA mass spectra determined from ambient data sets; this is shown in Figure 3 above. Indeed, there is overlap between the SV-OOA and HOA factors determined from ambient data sets (Figure 3).

The revised manuscript includes the ranges of ambient SV- and LV-OOA factors, and gives increased discussion to the comparison between the experimental data and the ambient factors.

12. Page 24280. The authors should also show the mass spectra of the SOA from diesel so readers can compare those with the mass spectra from gasoline SOA directly.

We have made this change in the revised manuscript.

13. Page 24281, section 3.3. I think the use of the basis set model and PMF factors to infer the POA/SOA mixing in the experiments is not as informative as the authors suggested.

The data points in Figure 6 are extremely noisy. What are the uncertainties? It is difficult to tell from Fig. 6 whether ideal mixing or no mixing would represent the data better.

We have removed Figure 6 and associated discussion.

14. Supplementary info. I do not understand the context of Figure S5. More explanations of the figure and how it relates to the present work are needed.

We have removed this figure, and now discuss the gas-turbine engine experiments in detail in the main text of the revised manuscript.

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