First, we would like to thank the reviewer #2 for his valuable comments on the manuscript. We did our best to address all the comments and summarized the changes made to the revised manuscript below.

Response to referee #2:

>The authors present a framework for utilizing ToF-PTR-MS to measure methylglyoxal in the ambient atmosphere. The sampling set up and methodology for the PTR-MS are well developed. What is generally unclear is why the DNPH cartridge sampling method used to compare didn't have a better developed sampling system that would minimize and account for loss mechanisms in the inlet and other bias. If it is known in the literature that MGLY is considered "sticky" for inlets, then steps should be taken to mitigate or quantify the issue, rather than have to use inferences from the field campaign to explain differences later. It would have been a logical next step to apply the calibration procedure used post field campaign with the PTR-MS with the DNPH set up as well to understand and quantify the loss processes of the MGLY in the DNPH set up such as the sampling line and the KI ozone scrubber. As the paper stands the authors have done a good job of sorting out what they can based on the ambient data and the post campaign calibrations with regard to the interference of water clusters and other factors on the MGLY measurements.

The PTR-ToFMS sampling system was optimized to get the best transmission of "very sticky" compounds such as carboxylic acids for this campaign, which also helped for the measurement of other species such as MGLY.
The DNPH method was initially used in this experiment to focus on major carbonyls such as formaldehyde, acetone, acetaldehyde, MEK, MVK and MACR. However MGLY is among the species measurable and thus DNPH method offered this possibility for comparison with the PTRMS. The method has been used with sampling line and ozone scrubber as recommended within TO-11 and EMEP sampling and analysis manual. Indeed, the present study points out some issues to be considered and since then additional studies were performed to improve this method 1) a European study (ENV56/Key-VOCs) tested different materials for sampling lines and sulfinert is now currently used; 2) the collection efficiency of cartridges remains around 100% for aldehydes whatever the relative humidity while a decrease has been observed for ketones above 60% (consistent with Ho et al 2014) leading to the systematic use of two cartridges back to back.. These tests have been done using gaseous standard for carbonyls not including MGLY since it is more challenging to generate mixture with stable concentration of MGLY at a flowrate of 1,5 L min^{-1}. These tests should be performed in a mid-term.

>In the review of other techniques used to measure MGLY (pages 3-4), the authors rightly point out many of the advantages and disadvantages of other analytical techniques including derivatization and measurement with either GC or HPLC, highly reflective cavity instruments (CE-DOAS and IBBCEAS), and Laser-Induced Phosphorescence. The authors point out the fragile equipment and the need for highly skilled operators as a draw back for these techniques, implying that PTR-MS does not have these as an issue. It could definitely be argued that PTR-MS which comes with a much larger price tag and can certainly be described as temperamental (as any field instrument involving a mass spec and ionization source could be described) certainly need the attention of highly skilled operators and a knowledgeable analyst doing the data work up. The PTR-MS definitely beats out the other techniques in terms of the possible low detection limits, as even in isoprene rich environments such as the LA basin (Washenfelder et al., 2011), while glyoxal is reported by multiple instruments, MGLY is not.

We agree with the reviewer that operating PTR-ToFMS instruments and processing data also requires highly skilled operators. However, this type of instruments is used more extensively than LIP and cavity instruments during field campaigns. We have added the following statement at the end of the introduction section to clarify these points:
"While PTR-ToFMS instruments also require highly skilled operators and are more expensive than other techniques allowing MGLY measurements, a growing number of research groups is deploying this type of instrumentation during intensive field campaigns, making it of great interest for MGLY measurements. It is expected that PTR-MS should allow reaching a lower LOD than any other techniques reported in the literature so far."

>Page 5 line 21: Methyl glyoxal should also be considered as a 1st generation oxidation product of isoprene oxidation. This was shown mechanistically by Paulot et al. (2009), inferred by Galloway et al. (2011) and measured by Thalman (2013) as a 1.8(0.4)%.

This has been modified in the revised manuscript as follows:
"Since MGLY is an oxidation product of isoprene (1st, 2nd & 3rd generations)…"

>Page 9 line 10: The authors state that the calibration range was higher than the actual ambient range, why not just dilute further to reach the ambient range?

We agree with the referee that we should have further diluted the calibration gas to reach, at least, the lowest ambient concentrations. Nevertheless, as stated in the manuscript, we always observed a linear response for the calibrated VOCs on this PTR-ToFMS. There is therefore no indication that the calibration performed for MGLY in this study is not usable for ambient quantification.

>Page 10 Line9: Use "concurrent" rather than "concomitant" which implies that the accompanying of the two measurements was 'naturally occurring'.

This modification has been made in the revised manuscript.

>Page 11 line 2: As Reviewer 1 has pointed out, an R2 of 0.48 for two techniques measuring the same compound is much worse than expected, though Thalman et al. (2015) reported R2 values for MGLY in mixed compound experiments in the range of 0.65 between the CE-DOAS and SPME techniques.

This statement has been modified in the revised manuscript. Please see the answer provided to the first reviewer.

>Page 14 line 34: It is generally considered that methylglyoxal has a much shorter atmospheric lifetime than glyoxal (which is one of the reasons that it is much harder to measure in ambient air), where MGLY has both a larger rate constant for reaction with OH and a 5x larger quantum yield from photolysis (Plum et al., 1983).

Methylglyoxal has indeed a shorter lifetime than glyoxal. However, their global average lifetimes are of the same order of magnitude: 1.6 and 2.9 h for methylglyoxal and glyoxal respectively (Fu et al., 2008). Even if the lifetime of MGLY is shorter by approximately a factor 2 than glyoxal, we cannot rule out a breakdown of the photostationnary state balance and we prefer to be cautious. The manuscript has been modified as follows:

“However, Washenfelder et al. (2011) showed a breakdown of the photostationary state when applied to glyoxal, a dicarbonyl compound exhibiting a lifetime of the same order of magnitude than MGLY, and as a consequence the calculated loss rate reported in this study only provides a rough estimation of the local production rate.”

References:

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