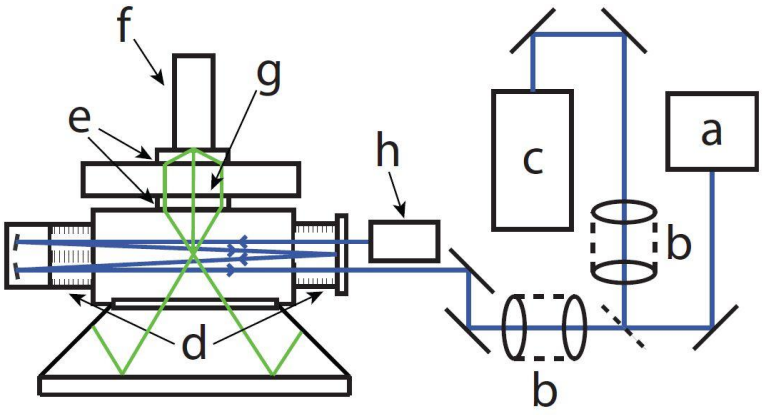
We thank the referee for their constructive questions and comments. The referee's questions and comments are shown in bold with our responses in plain text.

General comments:
This manuscript presents a method for simultaneously measuring glyoxal and methylglyoxal. The two molecules are distinguished by their phosphorescence decay lifetimes. Three-sigma detection limits of 11 pptv and 243 pptv are reported for glyoxal and methylglyoxal, respectively. This work improves upon a previous version of the instrument, reported by Huisman et al 2008, which used a wavelength-tunable Ti:Sapphire laser to determine glyoxal concentration.

This manuscript includes three new and interesting results:
- Measurement of methylglyoxal by phosphorescence.
- Simultaneous retrieval of glyoxal and methylglyoxal by fitting their time-dependent phosphorescent decays.
- Replacement of the Ti:Sapphire laser with a much cheaper CW laser in the phosphorescence instrument.
The paper addresses a relevant scientific question and is within the scope of AMT.

Major comments:
- The primary purpose of this paper is to describe a new instrument, but no schematic is shown. Please add a figure for this.

A figure has been added to the experimental setup section (2.2) describing the instrumental layout.



[Figure Caption: Schematic drawing of the experimental setup. The components are: light source (a), focusing optics (b), cavity ringdown cell (c), light baffles (d), collimating optics (e), single photon counter photomultiplier tube (f), optical bandpass filter (g), laser power meter (h). Components d-g are inside the white type multipass cell. A pump (not shown) draws air through the detection cell at ~20 SLM.]

- The authors refer to LIPGLOS both as an instrument and as a data retrieval technique. The authors compare the Mad-LIP instrument to the LIPGLOS instrument, and they also use the LIPGLOS data retrieval technique with the Mad-LIP instrument.

We thank the referee for pointing out this confusion. It has been clarified that LIPGLOS is the method that is used to extract glyoxal and methylglyoxal contributions from decays taken with an experimental set up similar to the Mad-LIP instrument. It has been further clarified that the Mad-LIP instrument uses the gated photon integration method.

I agree with the first reviewer's concern that the description of the instruments and their configurations is confusing and should be clarified. One improvement would be to give the data retrieval technique a name that is separate from the instrument (such as "multiple phosphorescence decay fitting" or something similar).

Text at the beginning of the light source section (2.2.1) as well as the data acquisition card section (2.2.3) has been added to clarify that either and not both of these components are required for operation.

In Sec. 2.2.1:
"The use of two different light sources was investigated independent of one another as this method only requires a single light source."

In Sec. 2.2.3:
"Even though a single data acquisition (DAQ) card is required to digitize the signal from the PMT, two different cards had to be used in different situations independent of one another due to equipment availability."

An additional section "2.2.5 Instrumental Configurations" has been added describing the usages of the different combinations of cards and light sources during what experiments.

- Pg 6160: The first sentence of the introduction ("glyoxal and methylglyoxal are nearly ubiquitous products of the HOx/NOx cycle") is strange. Glyoxal and methylglyoxal are VOC oxidation products, not products of HOx/NOx.

We thank the referees for this comment. This has been rewritten to read:
"Glyoxal and methylglyoxal are nearly ubiquitous and are generated through volatile organic compound (VOC) oxidation by the HO_x/NO_x cycle…"

- Pg 6161: The introduction should include a list of the major precursors for glyoxal and methylglyoxal. When highlighting the importance of isoprene, it would be useful to add the measured first-generation yield of glyoxal and methylglyoxal from isoprene.

The following text has been added to address this:

"Globally, the majority of glyoxal (47%) and methylglyoxal (79%) comes from isoprene (Fu et al., 2008). Isoprene makes up a large portion (1/3 to 1/2) of globally emitted carbon at an estimated rate of 503 Tg yr^{-1} (Guenther et al., 1995). Glyoxal and methylglyoxal have direct yields from isoprene of 2.1% (Galloway et al., 2011) and 4.2% (Galloway et al., 2011; Paulot et al., 2009), respectively. The remainder of glyoxal comes from acetylene (Fu et al., 2008) and various alkenes (e.g. 2-methyl-3-buten-2-ol, propene, or 2-butene Chan et al. (2009); Volkamer et al. (2007)) and aromatics (e.g. benzene, toluene, and p-xylene) (Volkamer et al., 2005b, 2007). The methylglyoxal that does not come from isoprene is yielded by acetone (Fu et al., 2008), alkenes (e.g. methylvinylketone or 2-Methylprop-2-enal (Galloway et al., 2011)), and aromatics (e.g. toluene, (m/p)-xylene (Tuazon et al., 1984))."

- Is the methylglyoxal measurement sensitive enough for ambient atmospheric measurements?

Spaulding et al. 2003 reported methylglyoxal concentrations as high as 320 pptv with a median of 120 pptv in an MBO dominated forest in the western Sierra Nevada Mountains. In Shenandoah National Park in Virgina, methylglyoxal did not exceed the limit of detection of 50 pptv (Munger 1995). At a rural site in Georgia, methylglyoxal achieved a maximum of 160 pptv with a median of 0.016 pptv (Lee 1995). Since the limits of detection of this method as presented here are within the range of reported values instead of below, this method could at least put a close upper limit to ambient methylglyoxal when it was below the detection limit.

The introduction now details concentrations observed:
"Low tens to low hundreds of pptv for both glyoxal (15 - 190 pptv) and methylglyoxal (<50 - 320 pptv) have been reported in rural, urban, and marine regions in this work (Sect. 4) and others (Lee et al., 1995; Munger et al., 1995; Spaulding et al., 2003; Fu et al., 2008; Huisman et al., 2008; Vrekoussis et al., 2009; Sinreich et al., 2010)."

Additional references for response which have been added to the document:
"Formaldehyde, glyoxal, and methylglyoxal in air and cloudwater at a rural mountain site in central Virginia", J. William Munger, D. J. Jacob, B. C. Daube, L. W. Horowitz, W. C. Keene, B. G. Heikes, JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 100, NO. D5, PAGES 9325-9333, MAY 20, 1995

"Atmospheric carbonyl compounds at a rural southeastern United States site", Yin-Nan Lee, Xianliang Zhou, and Kristen Hallock, JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 100, NO. D12, PAGES 25,933-25,944, DECEMBER 20, 1995

Or is it necessary only to correctly quantify the glyoxal concentration?

If the methylglyoxal concentration is below limit of detection, it would have essentially no contribution to the measured phosphorescent decay so it would not have to be fitted out.

- For Table 1, please add a column with the laser wavelengths and FWHM. The references for the absorption cross-sections are not given here and should be included as a footnote. In Section 2.2.1, the importance of the relative cross sections are discussed, but the cross-

sections are not stated. Add a sentence to this section indicating that the reader can find a summary of the laser wavelengths and absorption cross-sections in Table 1.

We thank the referees for pointing out these oversights. Due to the fact that additional columns in table 1 will increase its width beyond the borders of the page creating formatting issues, another table has been included to summarize the laser wavelength and FWHM, their associated symbols in the document, and their respective optical cross-sections. References for the cross-sections have been added as a foot note to this table. A comment that a cross-section summary can be found in a table has been added during the discussion of light sources in Sec. 2.2.1.

- The cavity ringdown spectrometer described in Section 2.2.4 appears to be identical to that presented by Huisman 2008. If so, that should be stated with a reference to the previous Huisman paper.

The cavity ring down cell described in this document is a slightly altered version of that in Huisman 2008 (the inlet and outlet configuration is changed) which is ultimately based on a NOAA design. For this reason, we feel it is more appropriate to cite the NOAA papers which Huisman 2006 has used. These have been included in the document.

- Pg 6168 line 7: Clarify what "until no more data was recorded" means. Is there a signal-level threshhold?

We thank the referees for pointing out this confusion. During operation with the Ti:Sapphire laser, the time between the laser pulses was 333 μs long, but data was only recorded for the first 45.5 microseconds of each of these periods. So data the data that was fit started at 2.5 microsec and extended to the end of the recorded time period (45.5 microsec).

The document has been changed to read:
"To eliminate the laser scatter and fluorescence, which are both short-lived compared to phosphorescence, the fitting began 2.5 μs after the laser pulse and extended to either the end of the recorded data set, as in the case of the Ti:Sapphire (45 μs, Fig. 2a), or until the laser was turned back on, as with the CrystaLaser (35 μs, Fig. 2b)."

Figure 2 corresponds to Fig. 1 in the original document.

- Pg 6170 line 9: Give the chemical structure for biacetyl. Add a reference for the quoted cross-section.

We thank the referees for pointing out this oversight. We have included the smiles string as well as the reference for the biacetyl cross-section.

- Pg 6171 line 15: Give the actual slope and intercept values here. The 29.8 pptv intercept for the lambda_T:S,L fit is large and significant. Please add a comment about this, and about the significance of the intercept for lambda_T:S,H as well.

While remaking the figure to include error bars, changes to what gated photon integration data was included had to be altered slightly to maintain a consistent integration time (this was not the case in the original figure). This led to a slight change in the numbers in the plot, which altered the intercepts to be slightly smaller. A comment on the intercepts has still been included in the document. The text has been changed to read:

"Figure 6 demonstrates that the measurements were highly correlated with R^2 values of 0.98 and 0.97 and 1-to-1 within error 1.00±0.04 and 1.00±0.07 at $\lambda_{T:S,L}$ and $\lambda_{T:S,H}$, respectively. The correlations do possess a non-zero y axis intercept value (-2±22 at $\lambda_{T:S,L}$ and 2±20 at $\lambda_{T:S,H}$), however they are statistically insignificant."

Figure 6 corresponds to Fig. 4 in the original document.

- Presumably, the reason for using the LIPGLOS fitting method at the Mad-LIP lambda_T:S,L wavelength is to achieve a more sensitive measurement of methylglyoxal. This is unclear until the conclusions, and left me wondering about why you would use this poorer wavelength and show it in Fig. 4.

We thank the referee for this comment. The following text has been included in the light source section (2.2.1) of the document:

"Three different wavelengths are considered in this study to investigate how the measurement sensitivities change with optical cross-sections that vary with wavelength. In addition, the interference between glyoxal and methylglyoxal can be probed by changing their relative sensitivities (see Sect. 3.2). Finally, comparing the CrystaLaser as an alternative to the much more expensive Ti:Sapphire laser requires data taken at λ_{CL}."

- Pg 6172 line 5: Why is the standard deviation extrapolated?

The data collected with the Mad-LIP instrument that is compatible with the LIPGLOS method was collected with an integration time of 1.75 minutes. For purposes of comparison with the data from the gated photon integration method native to the Mad-LIP instrument, the standard deviation of the 1.75 minute data was translated to what it should be at 40 seconds. The wording has been changed to:

"During the morning of the 21st when concentrations are the lowest, the standard deviation of the gated photon integration data is 2.9 pptv in 40 seconds and the LIPGLOS data is 2.5 pptv in 1.75 min (translating to 4.0 pptv in 40 sec)."

- Pg. 6172 line 21: The conclusion about slope of 0.98 and r2 of 0.87 is not consistent with Fig. 5.

We thank the referee for pointing out this oversight. The conclusion has been changed to accurately reflect the data in the appropriate figure.

- Fig. 1: I concur with Referee #1's confusion regarding the use of "histogram" to describe the signal decay as a function of time. A histogram usually represents a frequency distribution.

We thank the referee for pointing out this confusion. As we have written it, it is not clear how we have generated the data displayed in this figure. The following text has been included in the data acquisition section (2.2.3) explicitly explaining how the histogram is generated:

"The single photon counting PMT emits a single 30 ns TTL pulse every time a photon is detected. This output is fed into the data acquisition card which records it as an analog signal of voltage versus time after the falling edge of the laser control signal. The arrival times of the pulses are then extracted from this waveform by finding at what times after the laser pulse the analog data exceeds a specified threshold value for a specified amount of time. Once the period of integration is complete, a histogram of these arrival times is then created (Fig. 2) and saved on the hard disk for later analysis."

Figure 2 in the revised text refers to Fig. 1 in the original text.

In addition to this, the y axis label for the histogram figures have been changed to "Number of Photons per Timestep" with "Timestep in y-axis label corresponding to temporal resolution of Alazar data acquisition card (5.55 ns)" included in the caption to further clarify the matter.

- Fig. 4: A few comments - Please add error bars. What is the source of the outliers? It looks like the scale was intended to be in color.

Error bars have been included. The source of outliers is most likely due to some amount of interference from methylglyoxal as it was being varied (130 – 5400 ppt_v) according to the color bar of the graph. The figure is now in color.

Minor comments:
Pg 6163 line 9: "If there are" should be "If there is".

This has been changed.

Pg 6163 lines 27 - 28: Incomplete sentence.

There are no lines 27-28 on page 6163. Assuming the referee meant page 6164, the sentence:

"During operation, 32 passes are used through the volume of the cell which ambient air is drawn (~1/2 L)."

has been changed to:

"During operation, 32 passes are used in the detection volume of the cell (~1/2 L) that ambient air is drawn through."

SLM and SCCM are not defined.

This has been changed.

Pg 6167 line 4: "is based on to the instrument"

This has been changed.

Pg 6168 line 4: "speices"

This has been changed.

Pg 6171 line 14: Change "blind" to "insensitive to".

This has been changed.

Pg 6172 line 9: "dirunal"

This has been changed.

Pg 6173 line 3: "speicies"

This has been changed.

Fig 5: "respectivly"

This has been changed.