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:
In the submitted manuscript, Henry et al., describe the development of a compact, inexpensive, laser-induced phosphorescence instrument for the simultaneous detection of glyoxal and methylglyoxal. The authors exploit the different phosphorescence lifetimes (12.75 and 7.26us, for glyoxal and methylglyoxal respectively) of the two structurally similar compounds and achieve an instrument that has a detection threshold of 11 ppt (glyoxal, 3sigma, 5min averaging) and 243 ppt (methylglyoxal, 3sigma, 5min averaging). The paper represents a novel advance in the field and should be published in AMT, although the description of the instrument and the discussion of the retrieved concentrations need to be revised, following the specific comments below.

Specific Comments:
1. The description of the instrument is complicated by discussion of multiple DAQ cards used (Alazar Tech vs. GaGe Applied Tech.) and multiple lasers employed (CW diode vs. Ti:Sapphire). It was not immediately clear that all of these components were not required simultaneously. As I understand, the authors are describing one instrument that can be operated in a number of different configurations using either laser A or laser B (or DAQ card A or B). This should be made clearer.

We thank the referee for pointing out this confusion. 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.

Further, the LODs reported in the abstract are for different lasers, this presumably means that the instrument as described needs to have both lasers on board to achieve these detection limits (for example, if only the CW laser is used the methylglyoxal LOD is high and dependent on the glyoxal abundance). This also, should be clarified.

We thank the referee for pointing out this oversight. Since this paper is primarily discussing a method that will allow inexpensive instrumentation, but not the instrumentation itself, it has been

clarified that the two limits of detection are with different light sources. The limit of detection yielded by the ambient studies has also been added to the abstract. It now reads:

"With two different light sources at different wavelengths, the lowest 3σ limits of detection observed during calibration with this method are 11 pptv in 5 min for glyoxal and 243 pptv in 5 min for methylglyoxal. During ambient measurements of glyoxal, a 3σ limit of detection of < 4.4 pptv in 5 min was observed."

I might suggest describing one system, then adding a separate section discussing the use if an alternative laser to improve the detection of methyl glyoxal.

See response to first comment.

2. In many places, the authors comment on the feasibility of these devices for inexpensive routine measurements. Could the authors provide some definition of inexpensive, with the optics and DAQ cards used, it is hard to imagine this device is much cheaper than $25k (for the CW diode based instrument). Again I might separate the manuscript into a discussion of the Ti:sapphire based instrument, then a discussion of a less expensive, less precise (for methylglyoxal anyways) CW diode based instrument.

With the CrystaLaser and the faster data acquisition card, it would cost ~$40k in components. With alternate light sources (e.g. high power LEDs) it could realistically be reduced to ~$30k. This has been included in the abstract as well as the conclusion.

3. On page 6161, it would be nice to have more specifics on the observed concentration ranges for glyoxal and methylglyoxal. It would be nice to have some confidence that a detection limit of 243 pptv in 5min (methylglyoxal), is this good enough to constrain chemical mechanisms using atmospheric observations. Outside of major cities, has this level ever been recorded?

We thank the referee for this comment. While there have been observations of methylglyoxal exceeding the reported LoD as determined with the setup described in the experimental section of the paper (Spaulding et al. 2003, high of 320 pptv, median 120 pptv), there is also work showing that methylglyoxal may not achieve a value detectable by this method in other areas (Munger et al. 2005, high <50 pptv or Lee et al. 1995, high 160 pptv, median 16 pptv). Clearly, in those areas where methylglyoxal is detectable this method could quantify ambient methylglyoxal. While the LoD may not be sufficient to quantify methylglyoxal in other areas, it still will provide a potentially close upper limit.

The introduction now reads:
"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)."

References for this response that have been added to this section:

"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

4. I am confused by the use of the histogram (in Figure 1) and in the analysis section. I generally think of a histogram as a probability distribution of a continuous variable.

We thank the referee for pointing out this confusion. Figure 1 is a decay taken in the laboratory. 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:

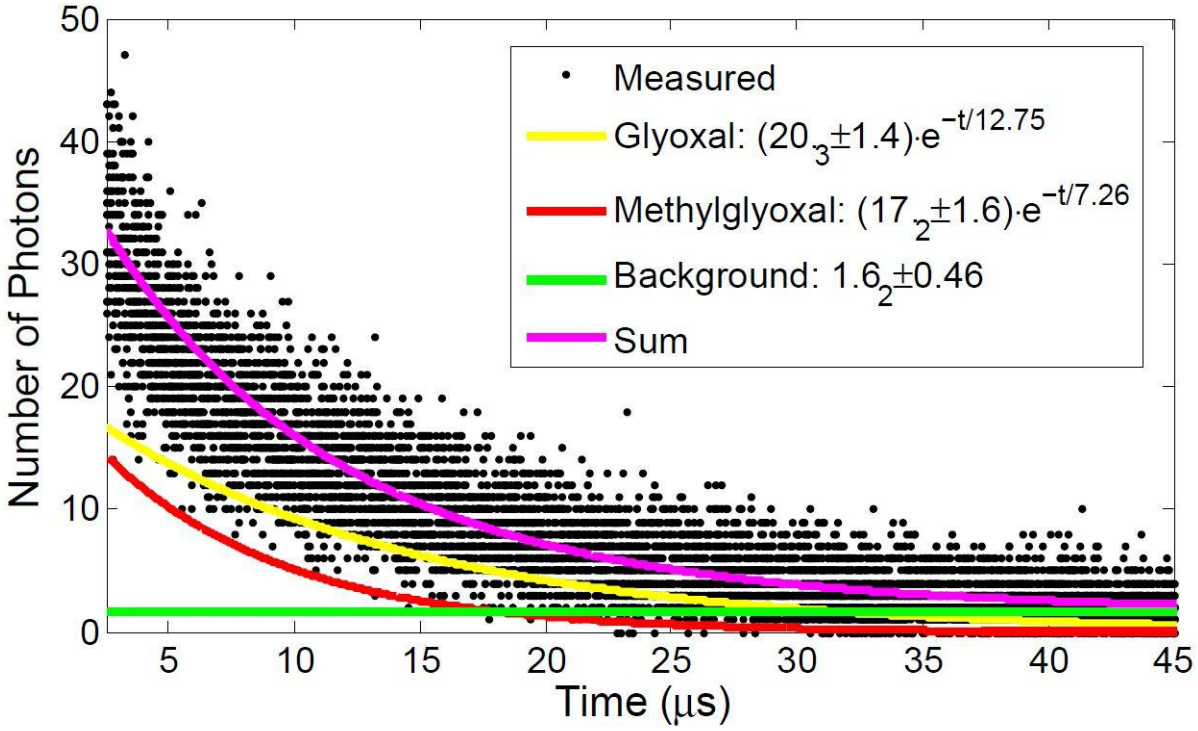
"The single photon counting PMT emits a single 30 ns TTL pulse every time a photon is detected. This output is fed into the data acquisition card which records it as an analog signal of voltage versus time after the falling edge of the laser control signal. The arrival times of the pulses are then extracted 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.

Figure 1, looks to me like an example decay. I would like to see an example of an atmospheric (or laboratory decay) that is fit with Eq4 to determine concentration.

Another figure illustrating the fitting method of a decay corresponding to a mixture of glyoxal and methylglyoxal to Eq. 4 has been added to the document.



[Figure Caption: Fig. 4. Timestep in y-axis label corresponds to temporal resolution of Alazar data acquisition card (5.55 ns). Decay of 5 minute integration for a mixture of glyoxal (180 ppt_v) and methylglyoxal (2500 ppt_v) taken with the Ti:Sapphire laser at $\lambda_{T:S;H}$ with line of best fit to Eq. (4) and its individual components.]

I think that the analysis section could benefit from a few additional paragraphs that describe exactly how the "histogram" or Eq4 is used to calculate concentration. Finally, I would also be interested to see a short few lines describing how the LOD was calculated.

We thank the referee for this comment. There is now text in the calibration section (3.2) that reads:

"After the standard gas was characterized via CRDS (see Sect. 2.2.4), independent phosphorescent calibrations were performed for glyoxal and methylglyoxal at concentrations between low pptv to low ppbv. Calibrant mixing ratios in the LIP detection cell were determined by diluting different calibrant gas flow rates that were controlled with a 10 SCCM flow controller (1779A, MKS Instruments) with ~20 SLM zero air that was controlled by a 100 SLM flow controller (1559A, MKS Instruments).

Once the analyte concentration inside the cell was stabilized, a histogram was taken. The resulting decay was fit to Eq. 4 to retrieve the contribution of glyoxal and methylglyoxal. A calibration curve was generated from the known concentrations in the cell and their given responses as determined from the fit. Table 2 summarizes the resulting calibration curves from these experiments. The LoDs included are three times the fit error of the blank in units of pptv."

Is this assuming that the uncertainty in the background is time independent and Gaussian over 5 minutes?

The background is assumed to be independent of time and Gaussian as it results from two things: stray ambient light in the cell and dark counts from the PMT which are not determined.

5. The glyoxal comparisons with Mad-LIP are excellent, as is the insensitivity of the instrument to the fraction of methylglyoxal. (Fig. 4 and 5). Is it possible to show a something similar for methylglyoxal (e.g. and x-y plot of glyoxal added vs. methylglyoxal measured for a lab experiment of constant methylglyoxal and varying glyoxal). This has been discussed a bit in Table 2.

The purpose of figure 4 is to demonstrate that the LIPGLOS method agrees with an already established glyoxal measurement especially under conditions where a possible interferent of glyoxal as measured by LIPGLOS (namely methylglyoxal) is significantly varied. Since we did not possess an established means of measuring methylglyoxal, a similar graph cannot be made. We feel adding a figure will not include any information not already contained in table 2.

Page 6171, lines 1-9 describe the results of experiments where glyoxal was held constant while methylglyoxal was varied. The analogous experiments of holding methylglyoxal constant and varying glyoxal would yield the same result as they are being determined is the same manner.

6. In table 2, what were the mixing ratios of glyoxal and methylglyoxal used in the "mixed" scenario?

We thank the referee for this comment. The following has been included in the caption for the appropriate table:

"During experiments at $\lambda_{T:S;L}$, glyoxal ranged from 140 to 1000 pptv and methylglyoxal ranged from 130 to 5400 ppt_v. During experiments at $\lambda_{T:S;H}$, glyoxal ranged from 140 to 750 ppt_v and methylglyoxal ranged from 130 to 5400 ppt_v. During experiments at λ_{CL}, glyoxal varied from 720 to 4200 ppt_v while methylglyoxal varied 730 to 6800 ppt_v."