We thank Referee #2 for his/her comments on our paper, which helped us to improve the quality of the paper. Below, we answer the reviewer's question point by point.

Liu et al. describe a new incoherent broadband cavity enhanced absorption spectrometer (IBBCEAS) for atmospheric measurements of NO_2, glyoxal, and methyl glyoxal. The instrument has been thoroughly characterized and performs well compared to existing methods. As a new feature, the instrument used a photolytic converter (NPC) to selectively reduce the amount of NO_2 in their sample stream.
Major revisions will be required before this paper is acceptable for publication.
(1) There are too many figures in the current draft. Many are not necessary.
Reply:
We have revised the manuscript and the number of figures have been shorten to 10. Some figures which we think could be helpful for illustrating the performance of our IBBCEAS system are shown in the Supplement.

(2) Some figures are unclear as the captions are too short.
Reply:
Captions of all figures in the manuscript are revised in the way that clearly describing the figure content.

(3) I was not convinced of the merits of the NPC. Is the NPC even necessary? It seems that (valid) data could be obtained without it, though at a slightly higher noise level.
Reply:
At the beginning of the experiments, we have the same doubt as you about how much NPC can improve the quality of measurements. Subsequent experiments indicate that NPC not only reduces noise level, but also weakens the measuring interference of high NO_2 concentrations. As shown in Fig. 8(a) and (b) in the revised manuscript, the NO_2 concentrations reduced by 76% and GLY concentrations reduced by 5% when the NPC was turned on. With respect to well-mixed NO_2 and GLY, the concentration of NO_2 dropped rapidly while the NPC was running and the concentration of GLY increased. After the NPC turned off, the concentrations of the two compounds returned to the same level as before the NPC was turned on (Fig. 8c). So the concentration of GLY determined by IBBCEAS can be underestimated in the presence of high NO_2 concentrations and by utilizing NPC to reduce the NO_2 concentration, the measurement accuracy of GLY is improved.

(4) Does the NPC generate glyoxal and methyl glyoxal when ambient air containing organic compounds is sampled? The MCM lists 127 precursors for glyoxal and 143 precursors for methyl glyoxal. I haven't gone through these lists in detail, but it seems that more characterization experiments are warranted in my opinion. It seems to me that the NPC may generate more issues than it solves.
Reply:
Given the size of the photolysis cell (i.e., a cylinder with 60 mm length and 13.4 mm

inner diameter) and the sampling flow rate of 2 L/min, the residence time of the sampled air in the cell is about 0.25 s. In such a short period, the production of GLY and MGLY in the cell is negligible which can be verified by model simulations. The model includes the full MCM chemistry (version 3.3.1, http://mcm.leeds.ac.uk/MCM/) for all NMHCs and their oxidation products. The initial concentrations of OH, HO_2, O_3, NO, HONO, CO, CH_4, C_2-C_{12} NMHCs are set to the average values obtained during a field observation campaign in 2018 in Yangtze River Delta, China (Table S1). The total OH reactivity due to NMHCs is about 20 s^{-1}. The relative humidity, temperature, and pressure are constrained by the campaign averages, i.e., 60%, 300 K, 1007.65 hPa, respectively. The photolysis frequencies are constrained by values calculated from the spectral actinic flux inside the cell (Fig. S1). The spectral actinic flux is estimated by the LED emission spectrum and by the concentration change of NO, NO_2, and O_3 when NO_2 standard (100 ppb) is running through the system. The initial values of NO_2, GLY, MGLY are set to 60 ppb, 100 ppt, and 100 ppt, respectively. Fig. S2 shows the calculated concentrations of NO_2, O_3, GLY, and MGLY for the period of 0.25 s residence time. While the NO_2 concentration drops from 60 ppb to 15 ppb leading to an increase of O_3 concentration by approximately 45 ppb, the change of GLY and MGLY concentrations is only around 1%. As shown in Fig. S3, the production rate of GLY and MGLY increases to around 5 ppb h^{-1} which could only result in maximumly 0.34 ppt increase of GLY and MGLY. Even if we increase the initial NMHCs concentration in the model by a factor of 10, the maximum production of GLY and MGLY within 250 ms is less than 2 ppt which is only 2% of their initial concentration (Fig. S4). Since the NMHCs concentration in the model represents typical atmospheric condition influenced by urban air pollution, our model simulation clearly shows that the NPC can be used in the field observations for removing NO_2 without additional production of GLY and MGLY.
The above discussions are added in the Supplement.

(5) The convolved spectrum of MGLY (Figure 4, thick purple line) does not look correct.

Reply:

Thank you for pointing out this mistake. We tested two high resolution cross sections of MGLY at the very beginning of our experiments, one is from Meller et al. (1991) and the other is from Staffelbach et al.(1995). In the manuscript, we mistook convolution results based on Staffelbach's spectrum instead of that based on Meller's spectrum. We revised Fig. 4 accordingly and checked the whole manuscript to ensure that the correct cross sections are used in every part involving MGLY spectral fitting.

Specific comments

pg 2, line 28 "Thalman et al. first encountered the challenge of fitting GLY and MGLY absorption spectra in the presence of high NO_2 concentrations (Thalman et al., 2015). To our knowledge, an effective method has not yet to solve this problem." This is a bit of mischaracterization - Thalman et al. [2015] stated that "For both CE-DOAS and BBCEAS ... we do not find significant bias; i.e., an upper-limit change in glyoxal due to

NO_2 is derived as +/-200 pptv glyoxal in the presence of 200 ppbv NO_2 (or bias of 1 pptv glyoxal/1 ppbv NO_2)."- see also major comment #3.

Reply:

According to Fig. 4 in Thalman et al. (2015), we can find that the fluctuation of the measured glyoxal concentration increased at least twice when NO_2 concentration reached 150 ppb. In addition, even though the upper-limit change in glyoxal due to NO_2 is derived as ±200 ppt glyoxal in the presence of 200 ppb NO_2 on the whole, the measurement interference on glyoxal already reached around 150 ppt when 50 ppb NO_2 was added. Since the ambient mixing ratios of glyoxal typically range between 10 ppt and 100 ppt, and NO_2 concentration in polluted area can reach 50 ppb or even higher, accurate quantification of glyoxal could be a challenge under this condition.

pg 7 line 31. Consider adding a digit to each uncertainty as otherwise 4% + 2% = 4%.

Reply:

Revised accordingly in the manuscript.

pg 9 line 23. "The presence of high NO_2 concentrations may affect the spectral fitting of GLY and MGLY". Perhaps, but this is not obvious from Figure 9 (which seem OK to me). To make their point, it seems like the authors could perform a rather easy experiment: Deliver a constant concentration of GLY (or MGLY) and add increasing amounts of NO_2 to this blend. Are the correct GLY (or MGLY) concentrations retrieved?

Reply:

Similar experiment as you suggested was performed in the Sect. 4.2. We delivered a constant concentration of GLY and add decreasing amounts of NO_2 to this blend for four times. The concentration of GLY measured by IBBCEAS increased as that of NO_2 decreased, although only the NO_2 concentration was reduced manually. The experimental phenomenon indicates that the presence of NO_2 will affect the retrieved concentration of GLY.

Figure 1 (a) Consider adding dimensions to this sketch, such as the inter-mirror distance

Reply:

Revised accordingly in the manuscript.

Figure 2 is not necessary in my opinion. Simply state the result in the text.

Reply:

Thank you for your advice. Figure 2 is deleted and corresponding results are stated in the text of the revised manuscript.

Figure 3. You may want to state the dimensions of the mixing chamber, temperature of the cold trap, and flow rates.

Reply:

Detailed information such as the dimensions of the mixing chamber is added to the caption of Figure 3. This figure is moved to the Supplement.

Figure 4 The convolved methylglyoxal spectrum does not look right.

Reply:

Revised accordingly in the manuscript.

Figure 6. This factor will be flow rate dependent, and the sample flow rate should be stated in the caption. Please zoom in on the ratio more (0.85 to 0.95) and state the standard deviation of this measurement. The effective path was determined from a comparison of measured to known concentration; the figure is misleading as it is suggested that it was determined by turning the purge gases on and off (filling the purge flow volumes would take much more time than shown here). Consider clarifying this in the text.

Reply:

Thank you for your advice. Figure 6 is re-plotted as you suggested and its caption is rewritten including sample flow rate and standard deviation of this measurement. This figure is moved to Supplement. The method of determining effective path that we used in the manuscript is measuring NO_2 concentration with or without purging flows. This kind of method has been approved by peers. For example, Duan et al. (2018) calculated the reduction of the effective cavity length successfully by measuring O_2-O_2 collision pair with or without purging flows.

Figure 11 The figure caption should describe the experiment conducted here in more detail. Figure 11 (c) The figure caption should indicate what is meant by the yellow overlay. Are the times displayed correct? It seems that the NO_2 mixing ratios stay at the high levels well after the NPC was turned on, but increase again before the NPC is turned off?

Reply:

The figure caption is rewritten as following and Figure 11 (c) is re-plotted.

"(a) NO_2 only test: A concentration gradient of NO_2 gas samples were measured twice, with the NPC on (yellow overlay) and off. The removal efficiency is constant at 76 % for different NO_2 concentrations. (b) GLY only test: The constant concentration of GLY produced by the standard gas generator was measured with the NPC on (yellow overlay) and off. A small fraction of the GLY (5%) was photolyzed by the NPC. (c) NO_2 and GLY mixture test: Well-mixed NO_2 and GLY was measured with the NPC on (yellow overlay) and off. The concentration of NO_2 dropped while the NPC was running and that of GLY increased. After the NPC was turned off, their concentrations returned to the same level as before the NPC was turned on."

Figure 12 - what does 'fixing' the concentration mean? What is 'first', 'second', 'third', and 'fourth'?

Reply:

The figure caption is rewritten as following.

"(a) Keep the concentration of GLY constant and mix it evenly with different concentrations of NO_2 for four times. The concentration of GLY measured by IBBCEAS

increased as that of NO_2 decreased, although only the NO_2 concentration was reduced manually. (b) Simulate a series of spectra with constant GLY concentration and increasing NO_2 concentration and then calculate concentrations of GLY and NO_2 by retrieving these simulated spectra. The retrieved GLY concentration decreased with the increasing NO_2 concentration, although the set value of GLY concentration in the simulation was kept constant."

Figure 13 (b). There are two sets of concentrations displayed which one is methylglyoxal, and what is the other one?

Reply:

Both sets of concentrations displayed are methylglyoxal. The figure caption is rewritten as following. This figure is moved to Supplement.

"(a) Four spectral fit ranges (440-451 nm, 445-453 nm, 440-453 nm, and 430-453 nm) for MGLY. (b) Each spectral fit range was used to fit two experimental sets of MGLY produced by the standard gas generator. Experimental results of both sets indicated that fit range has a great influence on the fitted concentrations of MGLY."

Figure 14. The figure caption should indicate what is meant by the yellow overlay. (b) Are the times displayed correct? It seems that the NO_2 mixing ratios stay at the high levels well after the NPC was turned on, but increase again before the NPC is turned off? The photo-oxidation of certain organic molecules (e.g., benzene, toluene etc.) can yield glyoxal or methylglyoxal. It looks like this is happening here. Consider evaluating the NPC while sampling an atmospheric background of those VOCs.

Reply:

The figure caption is rewritten as following and Figure 14 (b) is re-plotted in the manuscript.

"(a) The constant concentration of MGLY produced by the standard gas generator was measured with the NPC on (yellow overlay) and off. The effect of NPC on MGLY concentration is negligible. (b) Well-mixed NO_2 and MGLY was measured with the NPC on (yellow overlay) and off. The concentration of NO_2 dropped while the NPC was running and that of MGLY increased. After the NPC was turned off, their concentrations returned to the same level as before the NPC was turned on."

The experimental results shown in Figure 14(b) are the concentration of measuring the mixture of methylglyoxal and NO_2. Because the gas mixture only contained methylglyoxal, NO_2, and N_2, the increase of methylglyoxal concentration cannot be caused by the photooxidation of certain organic molecules. So the methylglyoxal concentration determined by IBBCEAS could be underestimated and we used NPC to reduce the interference.

The advice of evaluating the NPC while sampling an atmospheric background of VOCs is reasonable. The results of model simulation shows that the production of GLY and MGLY in the NPC is negligible. For more information please refer to the reply of major comment (4).

Fig 14 (c) The caption does not adequately describe what is shown in the figure.

Reply:

The caption of Fig. 14(c) is rewritten as following.

“(c) Simulate a series of spectra with constant MGLY concentration and increasing NO_2 concentration and then calculate concentrations of MGLY and NO_2 by retrieving these simulated spectra. The retrieved MGLY concentration decreased with the increasing NO_2 concentration, although the set value of MGLY concentration in the simulation was kept constant.”

Reference

Duan, J., Qin, M., Ouyang, B., Fang, W., Li, X., Lu, K., Tang, K., Liang, S., Meng, F., Hu, Z., Xie, P., Liu, W., and Häsler, R.: Development of an incoherent broadband cavity-enhanced absorption spectrometer for in situ measurements of HONO and NO2, Atmos Meas Tech, 11, 4531-4543, 2018.

Meller, R., Raber, W., Crowley, J. N., Jenkin, M. E., and Moortgat, G. K.: THE UV-VISIBLE ABSORPTION-SPECTRUM OF METHYLGLYOXAL, Journal of Photochemistry and Photobiology a-Chemistry, 62, 163-171, 1991.

Staffelbach, T. A., Orlando, J. J., Tyndall, G. S., and Calvert, J. G.: THE UV-VISIBLE ABSORPTION-SPECTRUM AND PHOTOLYSIS QUANTUM YIELDS OF METHYLGLYOXAL, J Geophys Res-Atmos, 100, 14189-14198, 1995.

Thalman, R., Baeza-Romero, M., Ball, S., Borrás, E., Daniels, M., Goodall, I., Henry, S., Karl, T., Keutsch, F., and Kim, S.: Instrument intercomparison of glyoxal, methyl glyoxal and NO2 under simulated atmospheric conditions, 2015. 2015.

Table S1. Initial concentration of species included in the model simulation.

Species	Concentration	Species	Concentration
OH	10^7 cm^{-3}	NO	0.60 ppb
HO2	10^9 cm^{-3}	HONO	0.45 ppb
CH4	1.9 ppm	CO	0.33 ppm
O3	70.00 ppb	SO2	1.33 ppb
NO2	60 ppb	MGLY	100.0 ppt
GLY	100.0 ppt	TOLUENE	0.515 ppb
C2H2	1.000 ppb	NC8H18	0.034 ppb
CBUT2ENE	0.050 ppb	EBENZ	0.140 ppb
C2H4	0.900 ppb	MXYL	0.045 ppb
C2H6	2.130 ppb	NC9H20	0.019 ppb
IC4H10	0.380 ppb	OXYL	0.076 ppb
IC5H12	0.330 ppb	STYRENE	0.017 ppb
NC4H10	0.650 ppb	IPBENZ	0.012 ppb
NC5H12	0.240 ppb	PBENZ	0.014 ppb
PENT1ENE	0.004 ppb	METHTOL	0.016 ppb
TPENT2ENE	0.002 ppb	PETHTOL	0.015 ppb
C5H8	0.233 ppb	NC10H22	0.017 ppb
CPENT2ENE	0.003 ppb	TM135B	0.014 ppb
M22C4	0.019 ppb	OETHTOL	0.036 ppb
M23C4	0.020 ppb	TM123B	0.014 ppb
M2PE	0.100 ppb	PXYL	0.045 ppb
M3PE	0.076 ppb	NC11H24	0.019 ppb
HEX1ENE	0.009 ppb	C3H8	2.010 ppb
NC6H14	0.130 ppb	C3H6	0.120 ppb
M2HEX	0.030 ppb	C4H6	0.005 ppb
CHEX	0.044 ppb	TM124B	0.016 ppb
M3HEX	0.041 ppb	TBUT2ENE	0.002 ppb
BENZENE	0.364 ppb	BUT1ENE	0.040 ppb
NC7H16	0.055 ppb		

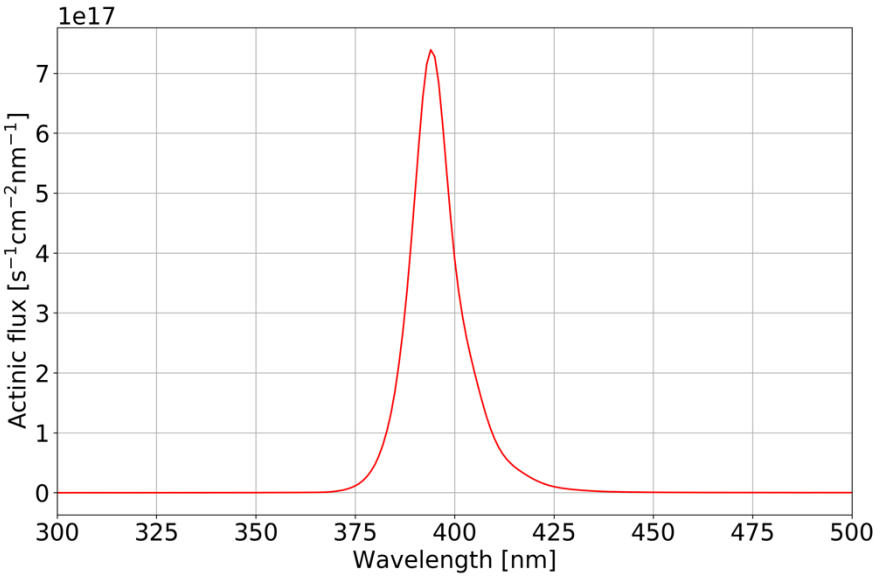

Figure S1. Spectral actinic flux inside the photolysis cell of the NO_2 convertor.

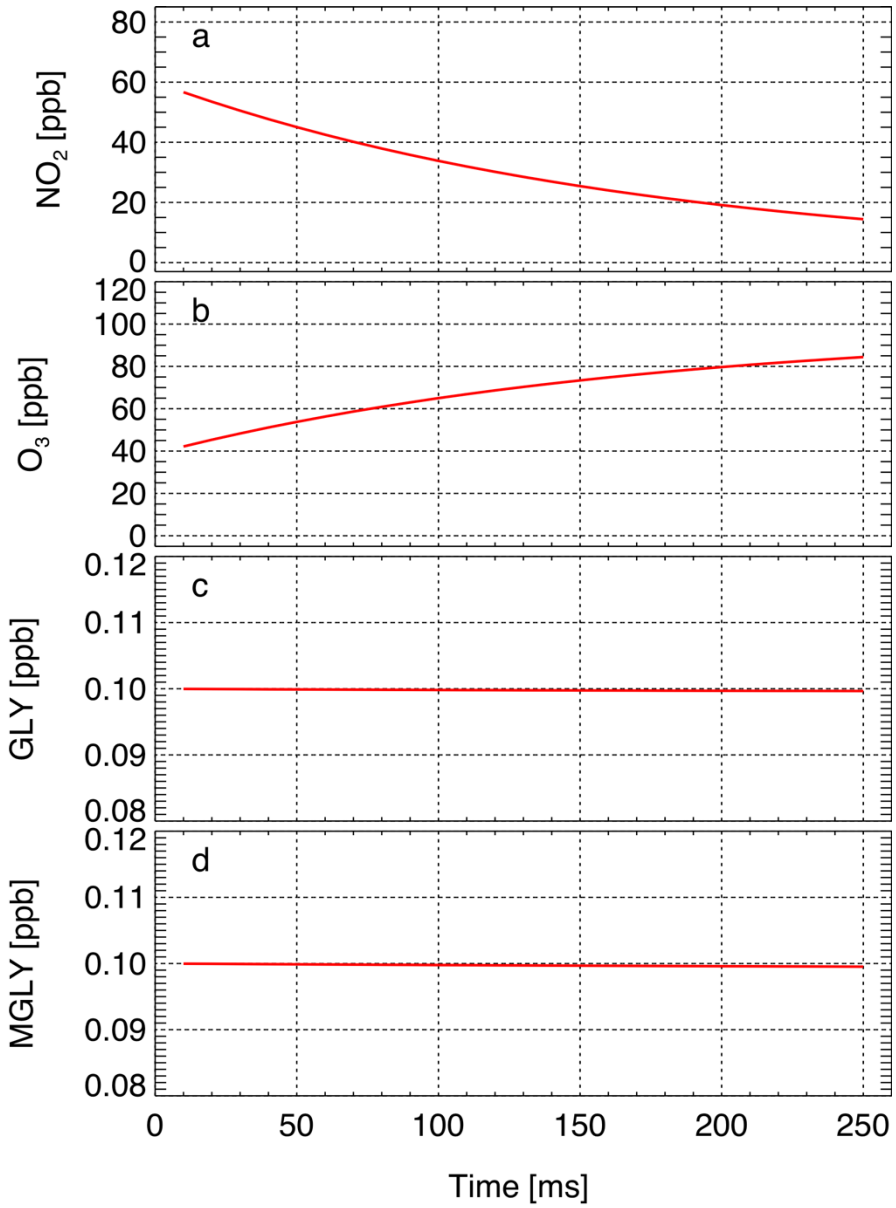

Figure S2. Model calculated concentrations of NO_2 (a), O_3 (b), GLY (c), and MGLY (d) in the photolysis cell of the NO_2 convertor. Note that the concentrations at 250 ms represent the condition of the sampled air exits the cell, since the residence time in the cell is about 250 ms.

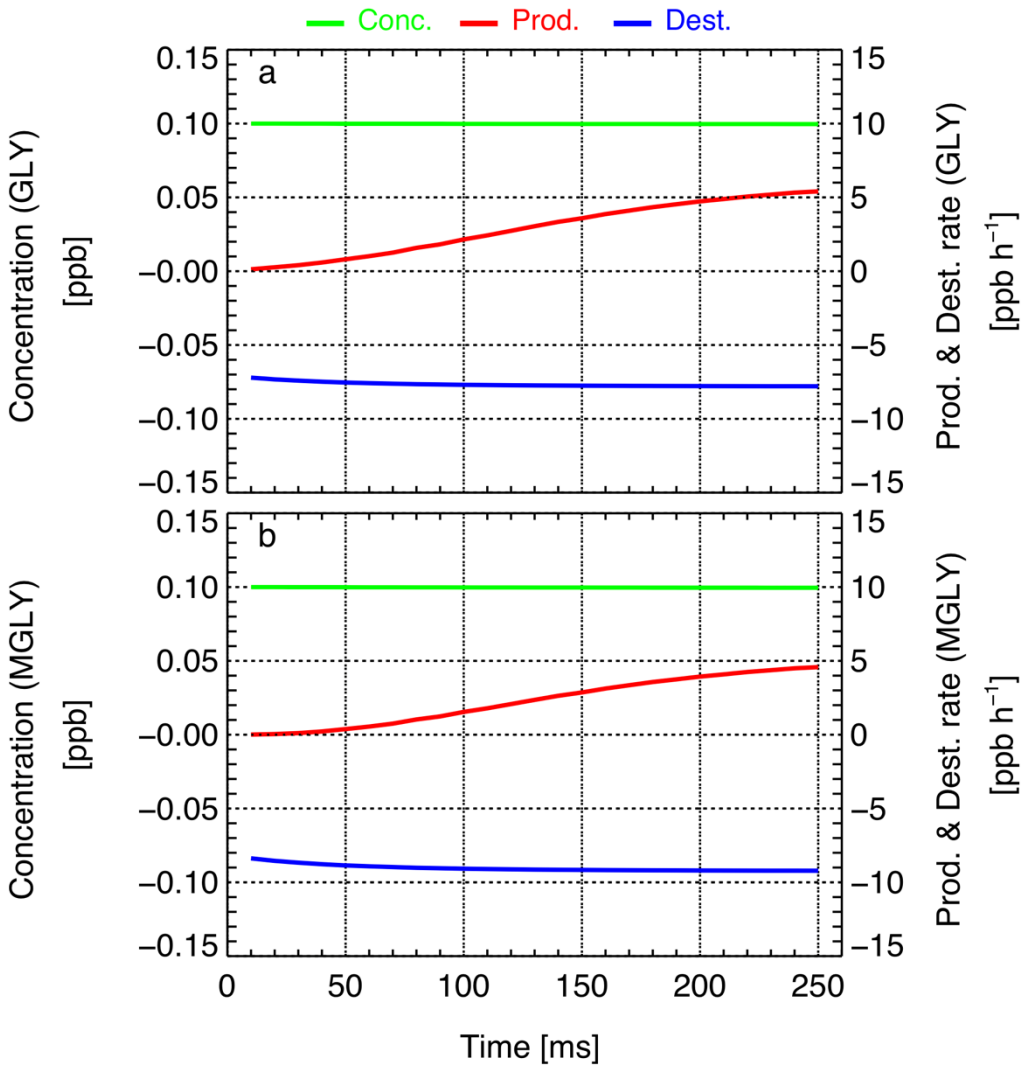

Figure S3. Model calculated concentrations (green), production rates (red), and destruction rates (blue) of GLY (a) and MGLY (b) in the photolysis cell of the NO_2 convertor. Note that the concentrations at 250 ms represent the condition of the sampled air exits the cell, since the residence time in the cell is about 250 ms.

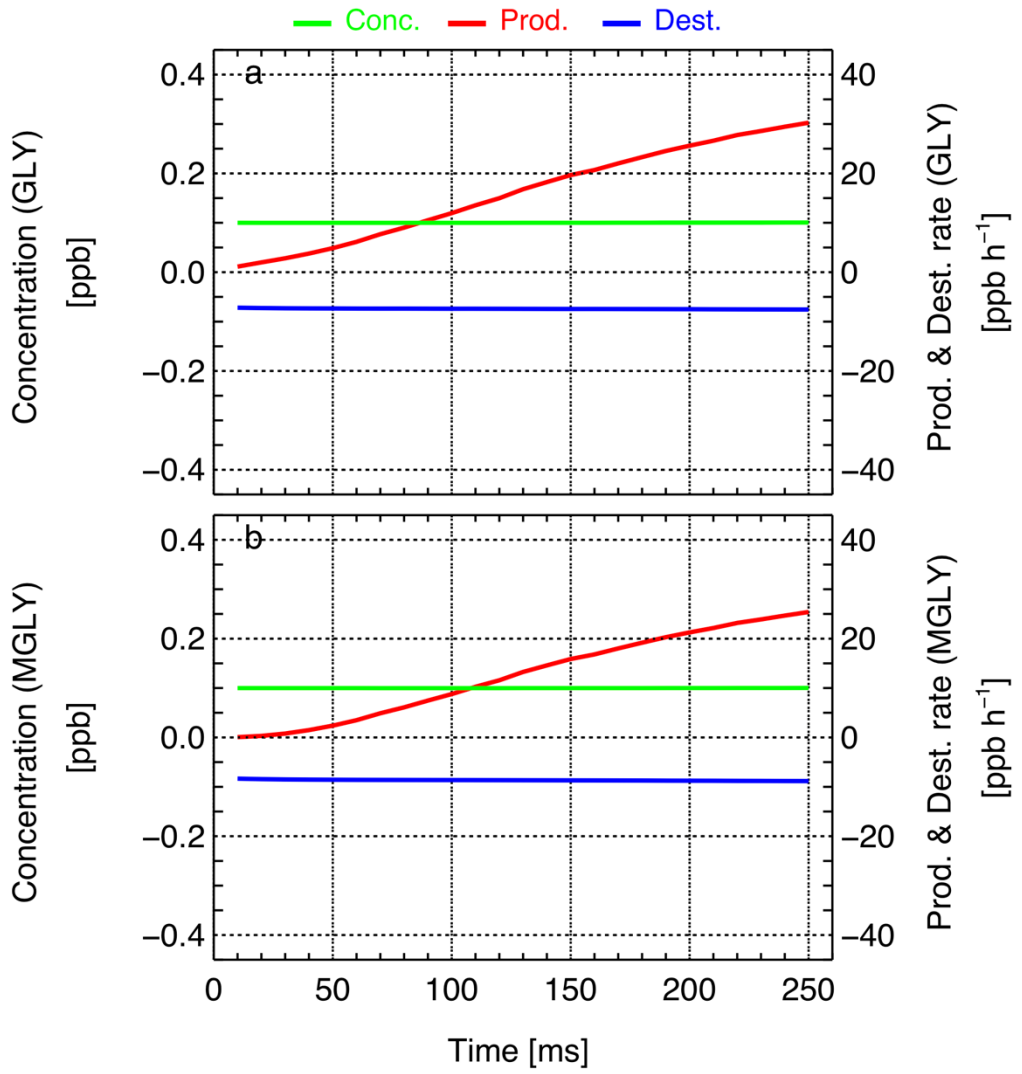

Figure S4. Model calculated concentration (green), production rate (red), and destruction rate (blue) of GLY (a) and MGLY (b) in the photolysis cell of the NO_2 converter. The initial concentrations of NMHCs in the model are set to 10 times of the values listed in Table 1. Note that the concentrations at 250 ms represent the condition of the sampled air exits the cell, since the residence time in the cell is about 250 ms.