We would like to thank the reviewers for their time taken to review the manuscript and for their helpful comments which will improve the manuscript.

RC1

The paper describes a new set-up combining laser-photolysis with time-resolved absorption spectroscopy using cw-QCL lasers. For validation, a part of the spectrum of the simplest Criegee intermediate, CH_2OO, has been measured as well as the rate constant of the reaction between CH_2OO and SO_2. The paper shows the future potential of the experimental set-up, without showing any new data. I have a few minor remarks that could improve the paper.

Figure 3: it would be interesting to see the signal on a longer time scale to have an idea about the influence of diffusion.

We have replaced the plot in the manuscript with one showing the full time series, which was measured between -1.5 ms and 3 ms and so shows limited impacts of diffusion.

Figure 4: I guess CH_2I_2 does not absorb in this wavelength range, because otherwise it would influence the measured spectrum?

Yes, we do not observe any effects of CH_2I_2 absorption in this region and the HITRAN database indicates there is no appreciable absorption by CH_2I_2 ~1286 cm^{-1}. We have added a comment on this to the caption.

Even though possible to calculate by everybody, it would be good to indicate the time it took to measure the spectrum to give an idea to the reader. I calculated that it has taken more than 60 hours to measure the spectrum, which is rather long given the quality of the obtained spectrum compared to the spectrum available in the literature. Why is the quality so low? Given the S/N ratio of the kinetic decay in Figure 5, which has been obtained also from averaging over 1000 laser pulses and with even lower concentration of CH_2I_2, I would have expected a much better S/N ratio in the spectrum.

The reviewer is correct that the method used in this paper required a lengthy series of experiments. We agree that the S/N ratio is lower than that achieved in the previous work, but we dispute that the quality of the spectrum is low. The high CH_2OO reactivity and concentrations on the order of 10^{12} cm^{-3} make high resolution spectroscopy challenging. For the experiments reported in this work, the spectrum was obtained using the temporal profiles recorded at each wavenumber, rather than an FT-IR method whereby the whole spectrum is obtained for each photolysis shot. Slight variations in the photolysis laser fluence from shot to shot in this work thus lead to slight differences in the initial CH_2OO concentration between experiments which impacts the S/N for the spectrum. For future work we intend to measure the laser fluence for each photolysis shot so that results can be normalised for this.

We also note that the technique used in this work is not proposed for detailed spectroscopic studies, but rather for the identification of species to be monitored in kinetics experiments. The full band for CH_2OO is included in this work to provide confidence in the capability of the technique, but in most cases only a partial band will be required for unambiguous identification.

Line 274: I don't understand the sentence: "The pre-photolysis region was defined as -4000 μs to -500 μs, owing to detection of some radiofrequency noise associated with the Q-switch delay of the photolysis laser which was set to 280 μs, and the post-photolysis region as 500 μs to 6000 μs, where t = 0 is the time at which the photolysis laser is fired." Is the Q-switch at -280 μs, and does the noise still influence even after the laser pulse, or why is there such a long post-photolysis delay? The signal in Figure 5 looks perfect from the first μs on. Maybe show an example of a typical signal with pre-trigger to clarify?

Yes, the Q-switch delay of 280 μs results in the Q-switch firing 280 μs before the 266 nm pulse (i.e. at -280 μs). We have clarified this in the main text as follows:

"The pre-photolysis region was defined as -4000 μs to -500 μs, owing to detection of some radiofrequency noise associated with the Q-switch delay of the photolysis laser which was set to 280 μs (i.e. the Q-switch fires at t = -280 μs), and the post-photolysis region as 500 μs to 6000 μs, where t = 0 is the time at which the photolysis laser is fired."

The radiofrequency noise generated by the Q-switch has a small impact on the measurement for a period that lasts beyond $t = 0$. In order to avoid this completely, we analyse data for the spectral measurements from $t = 500\ \mu s$ onwards, although using data from t = 0 would make only a small difference to the overall results.

Why do the data in Figure 5 show that the diode does not drift? Because Figure 5 is the average of 1000 photolysis shots, the average could still be a good quality decay, even if the wavelength changed during the measurement, no?

This is a good point, and should have referred more clearly to repeated measurements taken in separate experiments rather than the averaged measurements shown in Figure 5. We have re-worded the text as follows:

"The QCL can be tuned to a particular spectral feature and can remain tuned to that feature for prolonged periods of time during which kinetics experiments, and repeat measurements, can be performed without any drift in spectral position."

It would make it easier for the reader if Table 1 would also contain the corresponding reaction numbers.

We have added these to the table and modified the text at the start of Section 4.2 to include loss by diffusion labelled as R6.

The difference in pathlength is somewhat strange: would it not have been possible to measure both kinetics with the same alignment to verify if this is the reason for the change? Also I'm wondering if the k_{phys} has changed a lot between both experiments: with CH_2OO it is 500 s^{-1}, which should leave to a visible increase of the CH_3I kinetic in Figure 3, but Figure 3 looks like a k_{phys} well below 100 s^{-1}.

The difference in path length is relatively small and likely arises from small changes in alignment, or potential uncertainties in the absorption cross-sections or concentrations of CH_3I or CH_2OO used in the analysis. Unfortunately, there was a significant time period between the measurements involving CH_3I and those involving CH_2OO where access to the laboratory was restricted, with the result that the system had to be realigned between the two sets of experiments. The realignment also likely impacted k_{phys}, since this represents the diffusion of the species under investigation out of the probe region, and the extent of diffusion may depend, in part, on the overlap between the photolysis and probe lasers. However, we would like to note that for measurements of first-order and pseudo-first-order kinetics, knowledge of the path length is not required and is primarily included in this work to demonstrate the sensitivity of the instrument.

The error bars in Figure 6 are very small: I guess they are statistical from the exponential fitting? Please show a few examples of decays at the highest pseudo-first order rates, it would be interesting to see the quality of the data, especially with respect to the noise from the photolysis laser described above. Maybe this could be done as supplementary material.

Yes, the uncertainties are the statistical uncertainties obtained from the fits to the exponential decays. We have added comments to the captions to Figures 6 and 8 to clarify this:

"Error bars are 1σ from the fits to Equation 2/3".

We have also added some more example decays as an inset to Figure 6.

Because you can know the initial CH_2OO concentration as well as the formed SO_3, you should be able to measure the yield of SO_3. Did you try?

Measurement of the SO_3 yield requires knowledge of the SO_3 absorption cross-section at the spectral position used in these experiments, which is subject to large uncertainties, and so we did not attempt to characterise the yield in this work but this should be possible with the instrument described in the manuscript.