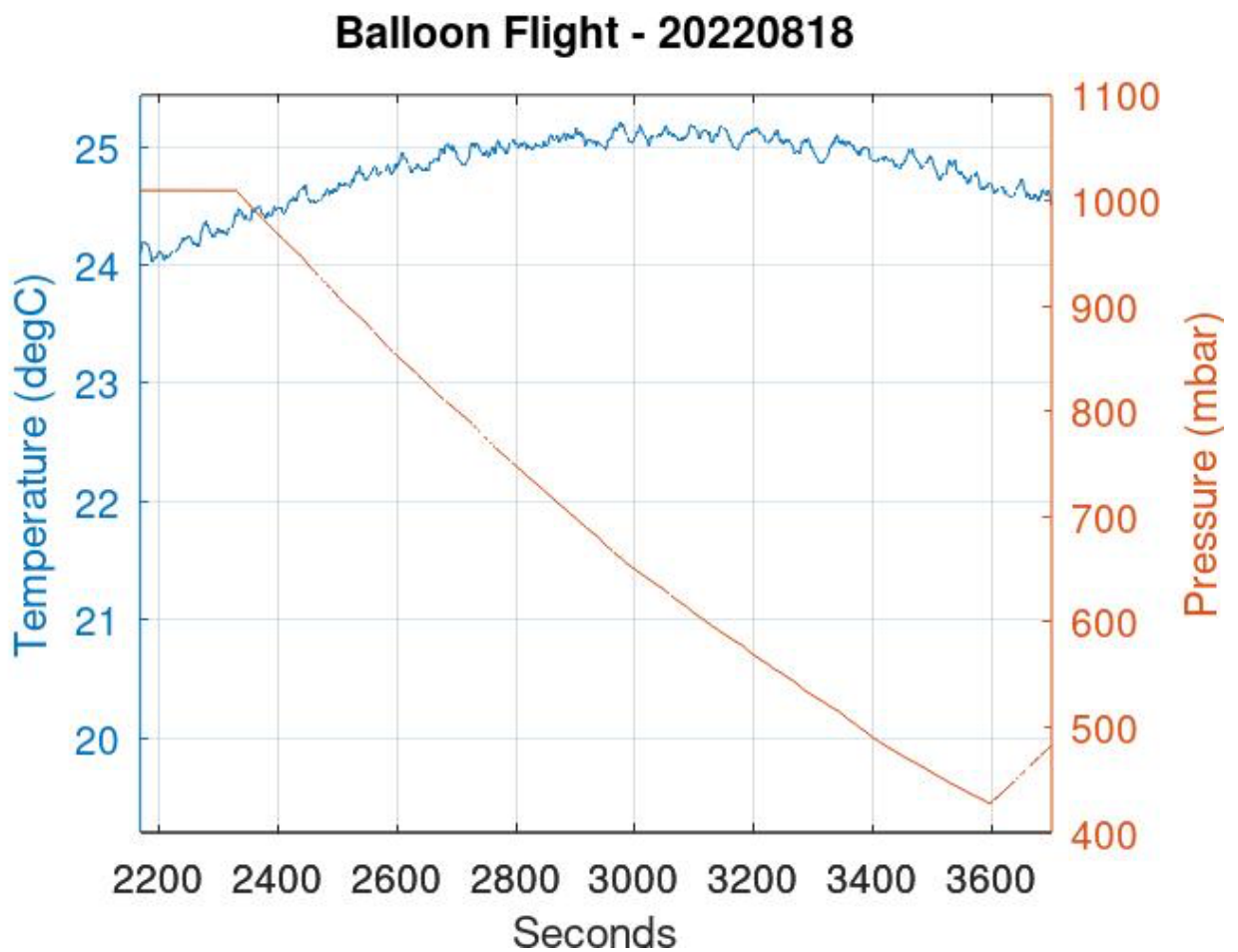


- 1) Comment from reviewer #2: *LED spectrum stability: LED emission spectrum can be very sensitive to its temperature and as you described, the LED here is temperature controlled at 25 degrees. Can you show that this is also the case at high altitudes with environment temperatures well below zero? presumably, that would make life easier for the TEC unless the LED is not generating enough heat.*

Response: Thank you for your comment. Our instrument was designed to fly on a drone up to 400 feet or on a balloon in the boundary layer. Given flight data shown in Figure 7 goes to 7km before balloon cutdown, our temperature in the instrument enclosure (which is insulated) deviated less than 1 deg C between launch (at ~2350 seconds) and cutdown (at ~3600 seconds) as seen in the plot below. I added a sentence to the current paper (line 249) indicating this temperature deviation.



- 2) Comment from reviewer #2: *Comment optical alignment: I can see how fine alignment of the cell is not required. However, the device can experience a significant temperature gradient from ground level to a height of several km. In the altitude profile you presented this gradient can easily reach 40-50 degrees. With thermal expansion, especially when part of the device is kept at 25 C, how confident are you that the device is not too misaligned? can you provide*

data similar to Figure 4 from an actual flight? with temperature profile and LED temperature if you can.

Response: Thank you for your comment. As I stated in my response to comment #1, our instrument is contained in an insulated enclosure with thermal controls for both the detector and LED. Given that and the small temperature deviation in the enclosure during flights through the boundary layer (< 1 deg C), we are not concerned with atmospheric temperature effects for these intended flights.

- 3) *Comment from reviewer #2: section 3.2 is confusing to me. presumably, you are referring to the molecular absorption cross-section which should have cm² or cm² / molecule units. you used molecules / cm². For water vapour, this should be orders of magnitude lower.*

Response: Thank you for your comment. You caught two typos, one of which I caught myself and corrected in the current document which now uses the cross-section units of cm² / molecule for water vapor. However, I did not catch the lower water vapor number. Thank you again. I will correct that number to 3×10^{-27} in the next version of paper.

- 4) *Comment from reviewer #2: in the caption of Figure 8, you refer to the device as a "Sonde" although it was not flown here.*

Response: Thank you for your comment. I will remove the words "NO₂ Sonde" and replaced them with the word "PCAND" in the caption of Figure 8 for the next version of the paper.

- 5) *Comment from reviewer #2: alongside a comparison to a state-of-the-art device at ground level I would like to see a comparison while performing under the intended instrument usage - i.e. during flight, under severe temperature and pressure gradients, and atmosphere composition changes.*

Response: Thank you for your comment. Although we would very much like to compare our instrument (in flight) with "a state-of-art device", there does exist such a device that is small enough to fly on a weather balloon or drone that measures NO₂. Regarding our instrument, we believe we have already flown it "under the intended instrument usage".

- 6) *Comment from reviewer #2: under Acknowledgements, you refer to drone flights. did I miss that in the main text? I didn't see a reference to any drone flights.*

Response: Thank you for your comment. Although we did have a drone flight, you are correct in that I do not mention it in the paper. Thank you for catching that. I will remove the sentence about drone flight in the Acknowledgements.

- 7) *Comment from reviewer #2: I would like to see a discussion about the possible photolysis of NO₂ at 405 nm considering its high quantum yield, or at least an explanation of why it is perhaps insignificant in this case.*

Response: Thank you for your comment. I added a new section 4.4 (Photolysis Effects) to the current paper (line 226) that addresses this comment. Here is that text:

Photolysis Effects

The photolysis quantum yield is 0.22 at 408 nm (*Troe, 2000*), so we expect some fraction of the NO₂ in the cell to photolyze, $\text{NO}_2 + h\nu \rightarrow \text{NO} + \text{O}$. In static cells the photolysis of NO₂ has been shown to be a concern (*Platt et al., 2019*) In the case of our detection, it is unlikely that a significant fraction of NO₂ will be photolyzed because the sample flows through the cell quickly with a flush time of approximately 1 s and the number of photons available for photolysis is small.

We can estimate the number of photons in the cell from the detector signal. The SiPM has a radiant sensitivity of $4 \times 10^5 \text{ A/W}$ and a photon detection efficiency of 50%. Based on our detection signal of $2 \times 10^{-5} \text{ A}$, we estimate the optical power is roughly 10^{-10} W and calculate a photon flux of $2 \times 10^9 \text{ photons/s}$. A typical absorbance with 1 ppb NO₂ in the cell is 10^{-3} , thus we expect that roughly $2 \times 10^6 \text{ photons/s}$ are absorbed by the 1 ppb NO₂ in the cell. At 900 hPa the number density of 1 ppb NO₂ is roughly $2.2 \times 10^{10} \text{ molecules/cm}^3$. The absorption of $2 \times 10^6 \text{ photons}$ would result in the photolysis of $4.4 \times 10^5 \text{ NO}_2 \text{ molecules}$, or 2×10^{-5} of the available NO₂ molecules. While this number is quite low for our conditions, it is worth noting that with slower flows and higher photon fluxes the photolysis could be significant and secondary chemistry could be a concern.