A Cavity-Enhanced UV Absorption Instrument for High Precision, Fast Time Response Ozone Measurements

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General comments:

This well-written and well-structured manuscript describes the development, validation and application of an instrument based on incoherent broadband cavity-enhanced absorption spectroscopy (IBBCEAS) for the direct detection and quantification of ozone in the UV-C region (~265 nm). The pulsed instrument (Rapid Ozone Experiment, ROZE) is designed for O_3 Eddie covariance measurements from airborne platforms and thus requires high time resolution. The authors give an excellent motivation for their work in the introduction. They briefly review the measurement principle before outlining the instruments operation, where sufficient attention to detail is provided to understand the function of the instrument and how the key objectives in instrument performance were met. The lab characterization of the instrument's performance addresses all relevant aspects. Finally the first application in airborne operation also shows the quality of the instrument for Eddie covariance measurements in comparison with a more established chemiluminescence (reference) approach. The authors pay sufficient attention to detail, error discussion, and relevant critical measurement parameters.
The manuscript is basically publishable in its current form, however subject to some improvement and amendments concerning more explanations as outlined below.

Specific comments:

L49: The UV region has the drawback that in this region many other trace species also have significant absorption bands and this puts substantial constraints on the selectivity of the method in that region. See also comment below (L216-220).

L70-74: The use of an optical filter simplifies the approach and is adequate because the spectrum is unstructured and the absorption has not much or very little "fingerprint character". However, it also illustrates the low selectivity for ozone in this region (see L216-220).

L101: The divergence of the LED is quite large – the surface area of LED is also large 1x1 mm^2 (extended light source). Light collection at a distance of 79 mm with a 1" diameter optic causes substantial light loss. Using a beam expander in revers increases the divergence after partial collimation again by a factor of 2. Can the authors give an estimate what fraction of the overall emission from the LED is actually being imaged onto the cavity – in other words, what is the light collection efficiency for the LED?

L103: "*…direct the beam 180° into the cell.*" This sounds strange. Please rephrase or at least refer to Figure 3 here.

L105-114 (section 3.1.2): There is no mention of a purge system. Were the cavity mirrors purged? If not – why not? There is also no mention of an aerosol filter in the inlet at this point, but later the authors point this out (since scattering in the UV is substantial). This section would benefit from mentioning these elements explicitly here.

L125/126: At this point I was wondering whether there was a flow controller? The flow seems to be only controlled by pump power? How accurate is the flow and how does it vary with pressure

variation? (Relevant for aircraft measurements on the ascent and descent). What are the three flows according to the 3 flow speed adjustments?
L145: Here it would be nice to explicitly learn what on- and off-times a 90% duty cycle refers to? Are there are delay times. Inherently pulsed IBBCEAS is not commonly applied and in the literature the large majority of instruments is indeed continuous wave. Pulsed IBBCEAS can in principle be biased by offsets depending on the measurement timing, which in turn may lead to systematic deviations in the measured absorption signal (see e.g. Keary & Ruth, Opt. Express, 2019). The authors may want to make an argument that these offsets are not observed here based on the electronics' timing. See also comment on Fig. 1 below.

L182/183: "*…and differential scatter or absorption due to non-uniform flow within the sample cell at high flow rates.*" As in the comment above, more on flow variation and or flow control would be helpful. The instrument might benefit from a pressure controlling flow controller.

L205: "*During this experiment, the pump maintained a sample flow rate of 18 SLM.*" Why was this not done at a lower mass flow of 11 SLM relevant for the airborne measurements with particle filter?

L206: I am a bit confused here: Please explain "3e fold flush rate". Also compare with caption of Fig 6 which says 1/3e is 9.5 Hz. The 3e-folding time is 150 ms (6.6 Hz)? A few more words would help.

L216-221: "*In fresh, concentrated smoke plumes, UV active species such as SO_2 and aromatic hydrocarbons can give rise to positive artifacts in the O_3 absorption measurement (Birks, 2015). … However, such UV-active absorbers are generally not abundant enough in the background atmosphere to be of concern.*"
This paragraph, which is a consequence of the low selectivity of the method for Ozone in the UV region, leaves a lot of open questions for the reader and is a real weakness of the manuscript. With a detection limit of tens of pptv the average abundance of species with absorption in the UV region, like e.g. BTX, formaldehyle, ketones, … can be in the low ppbv range. The selectivity of the current experimental approach is of clear concern here. In the sentence in line 221 this issue is simply discarded and not enough consideration is given to this issue in this manuscript. The implication is that, if the sample air composition is completely unknown, the interference of other species may render this approach inadequate, if it is applied on its own. A balanced discussion of the selectivity aspects, or potential ways to improve it by combination with other techniques, must be included in the manuscript. Finally, in line 220 the campaign data are said to be "*quality filtered*". If quality filtering means excluding data when formaldehyde mixing ratios were above 5 ppbv, then this is a rather crude way of doing this. Again, there must be a way of also identifying formaldehyde at the same time by other means.

L230: Cross plot -> Correlation plot

L232: Data from 15 extra flights could go into the supplementary material.

L253: …not shown in Figure 6.

L257/258: "*Scalar data processing included detrending the scalar mixing ratios by subtracting a 20 second running mean and synchronizing the data with the vertical winds.*" On what basis was that decided. Some more background explanation would be useful here.

Generally, the manuscript would benefit from a brief and compact comparison with other techniques for ozone detection and how the new ROZE compares in performance with those approaches. This could take the form of a small table. If this turns out to be too formidable, the authors may restrict this comparison to cavity enhanced absorption approaches.

Technical corrections (small improvements and formal/formatting errors):

Many superscripts (in units) did not come out correctly in print, see lines:
19, 45, 189, 190, 244, 266, 283

L43: Rephrase: … cylinders containing compressed toxic gases and the use of dangerous chemical dyes.

References:

Aubinet et al. – relevant pages might be missing
Birks – typo in titles and reference incomplete
Bourgeois et al. – volume seems to be missing
Ryerson et al. – NO 2 -> NO_2 (insert proper index)
Serdyuchenko et al. – ill-defined symbols in my copy
Young et al. – Journal name should not be abbreviated

Table:

Size – unit should read cm^3, or X cm x Y cm x Z cm
Precision – typo in value and unit

Figures:

Figure 1
- Even though this figure is only schematic, the way the LED light pulses are drawn is confusing. Pulses arrive at cavity with a 10 Hz repetition rate. At 10 Hz only one pulse is in the cavity at one time. The pulse duration is long enough to draw this figure in CW mode and explain the pulsed nature of ROZE in the text.
- Fig. 1 caption – replace *"long optical pathlength"* by *" long effective optical pathlength*"

Figure 2
Axes titles and colour code could be improved. Axis title for the LED spectrum (normalized intensity) is okay, for the cross-section the chosen (right) axis title is also okay. The figure however also contains the reflectivity spectrum, which is unitless, and thus the title should occur somewhere. The relative intensity axis can of course be used, but the title should be changed to make clear that 3 different quantities are shown. Moreover, the two shades of blue are difficult to distinguish, and do not work in my opinion; modify the color code.

Figure 6
- O3 on the axis title should be indexed properly; i.e. O_3.
- In panel (a) it would be better to show an individual pulse and a proper fit rather than 5 pulses, where the fitted data point can hardly be made out. The sentence: "*Individual pulses were fit to an exponential decay using the selected data points in red*" is not clear. An exponential function is fitted to the experimental data and not vice versa. This should be rephrased.
- What is meant by an "e/3 flush rate"? This is phrased casually.

Figure 7
- O3 on both axis titles should be indexed properly; i.e. O_3.
- Scatter plot -> Correlation plot
- How does an intercept of "0.17 +/- 0.02 ppbv" agree with the detection limit? Comment.