

**Review of “Airborne measurement of peroxy radicals using chemical amplification couple with cavity ring down spectroscopy: the PerCEAS instrument” by Midhun George et al.**

This paper describes a configuration of a peroxy radical chemical amplification (PerCA) inlet system and a cavity ring down spectrometer (CRDS) detector in a flight ready package for measurement of the sum of peroxy radicals ( $\text{HO}_2 + \text{RO}_2$ ) in the troposphere. The authors describe a series of instrument development experiments and modelling exercises used for optimization of instrument parameters. Example data from a single flight is shown as an example of flight performance.

This paper is a worthwhile addition to the literature as an update to a previously developed instrument from this group. Considerable improvements have been made in the detection of  $\text{NO}_2$  from Horstjann et al 2014 ( $\text{LOD}_{\text{NO}_2} = 300$  pptv) in the current configuration ( $\text{LOD}_{\text{NO}_2} = 60$  pptv), averaging time (120 s to 60 s) and altitude pressure limitations of the inlet. The reviewer believes this paper should be published but recommends a more focused approach on the novel improvements to the instrument rather than discussing well established and previously published methods (ie CRDS calibration, PerCA calibration, etc).

**General comments:**

Overall the reviewer believes 23 figures is too many for an instrument development paper of this nature. An instrument schematic could replace the first three figures (photos of the instrument, inlet and inside of aircraft) similar to Horstjann et al 2014 figure 1. The authors referred to an improved inlet design (DUALER vs DUALER II) by modifying the pre-chamber design and reducing wall interaction in the inlet. This modification seems significant and likely affects the instrument performance more than discussed in this paper. A figure comparing the two inlet designs or the changes in inlet design would be useful.

The general description of how the inlet operates (alternating measurement modes) is somewhat confusing and is evident in the Reviewer #1's comments. A time series figure of the operation of each channel would make this clearer (ie switching from amplification mode to background mode and showing how each channels mode switching is out of phase with each other). Furthermore, a detailed description of how the mixing ratio of  $\text{NO}$  was decided on (30 ppmv) would be useful, as it differs significantly from the DUALER I (6ppmv) inlet and other groups PerCA inlets (0.9 to 7.7 ppmv).

Generally speaking the flight data section of the paper should be focused on the improved performance of the instrument rather than flight tracks and mixing ratio figures. A comparison of DUALER I and DUALER II flight data is recommended. Considering how to show improvements between DUALER II deployments is recommended.

**Specific comments:**

Page 4, line 120, “The optical cavity remains similar to that described in Horstjann et al...” It is useful to include mirror specifications (substrate, coating, reflectivity, diameter, etc) for a CRDS instrument, as they are critical part of theoretical instrument performance. Does the piezo optical alignment system run in a closed loop control with beam profile as a feedback parameter? If so describe this, as it seems novel.

Page 5, line 146, “mode and modulation times...” it is not clear what mode and modulation times refer to, might be useful to define them discretely

Page 5, line 160, “...detector temperature. For this different detector temperature gradients,  $\Delta T$ , where applied to modulated signals generated by varying the sampled  $\text{NO}_2$  concentration...” it's not clear why the investigators modulated  $\text{NO}_2$  while applying a temperature gradient to the detector. Would it not be easier to interpret if a constant mixing ratio gas was sampled while applying a temperature gradient? It is not clear from the text where this temperature gradient is and how it was applied. It would be useful to readers that are not familiar with optics, on why a temperature gradient of 7 degC would cause detector instability. It is also not clear from the text, what was done to address this flaw in the detector design, as the authors state earlier detector stability is paramount in overall instrument performance.

Page 6, line 187, “...of the sampled  $\text{O}_3$  by  $\text{NO}$  to form  $\text{NO}_2$  also depends on the concentration of  $\text{NO}$  added to the sample flow and the time for reaction before reaching the detector.”, This would be a good place to discuss how 30 ppmv  $\text{NO}$  was decided on for a reagent mixing ratio and discuss flow rate choices for both  $\text{NO}$  and  $\text{CO}$ .

Page 6, line 192, “3.2.1 Effective Chain Length...”, This section seems to describe a well-established method documented in literature. The reviewer recommends shorting the description of the method and explain better the difference in DUALER I and DUALER II eCL.

Page 7, line 216, “The model was initialized with 9%  $\text{CO}$ , 3 ppb  $\text{O}_3$ , 50 pptv  $\text{HO}_2$ ...” why was 3 ppb  $\text{O}_3$  determined to be a representative mixing ratio for ozone? I may be misunderstanding the inlet chemistry, but it seems like missing 30 ppmv of  $\text{NO}$  would significantly affect the modeled CL. Assuming the box model initialization is correct, would it not be useful to vary the wall loss rate constants to match the eCL and determine if this wall loss is reasonable? It would also be useful to experimentally determine the wall loss of the inlet.

Page 7, line 222, “figure 8 shows eCL vs CL”, the authors should include error bars on these data.

Page 7, line 228, “Figure 9 depicts the  $\text{O}_3$  decay simulated for 100 to 200 ppb...” these figures are somewhat confusing to the reviewer. One could take the 99% conversion time for each  $\text{NO}$  mixing ratio curve and plot all 4 conditions (ie pressure and  $\text{O}_3$  mixing ratio) on 1 figure for varying  $\text{NO}$  mixing ratio. Additionally, adding an inlet residence time reference line would be useful for helping the reader visualize what time limit you have on this reaction.

Page 7, line 234, “PAN and PPN thermal decomposition”, the reviewer believes that experimental work is justified to confirm ‘this source of radicals is considered to be negligible’. The box modelling done for CL prediction was shown to not capture the actual inlet system, so it's not clear why it would do a better job with modelling PAN and PPN. Figure 11 shows up to 10 pptv interference, this does not seem negligible to the reviewer.

Page 8, line 252, “Figure 12 shows the variation of the eCL for 45 ppm  $\text{NO}$  within a pressure range...”, The reviewer does not understand why this experiment was done with 45 ppm  $\text{NO}$  when the decided upon mixing ratio of  $\text{NO}$  addition seems to be 30 ppm  $\text{NO}$  for the rest of the paper. If this is a typo, it should be corrected, if not the experiment should be done at the actual mixing ratio the instrument is operated at.

Page 9, line 284, “Figure 15 depicts exemplary a comparison of spectra...”, the reviewer does not believe including NO<sub>2</sub> absorption cross section and detector spectra is a useful figure for the main text of this paper. Remove or include in the SI.

Page 9, line 289, “In addition, the effective  $\sigma_{\text{NO}_2}$  can be calculated by sampling known mixtures...”, the reviewer does not believe including a time series of calibration gas addition to instrument is a useful figure for the main text of this paper. Remove or include in the SI.

Page 9, line 294, “The result of apply Eq. 4 to the PeRCEAS detectors at 200 mbar is depicted in Figure 17.” It is more common to plot NO<sub>2</sub> number density [molecules/cm<sup>3</sup>] vs.  $\alpha$ , as the slope has the physical meaning of the absorption cross section of NO<sub>2</sub>.

Page 9, line 302, “The main source of uncertainty...” the authors previously mention detector drift due to temperature changes (figure 5), is this not a significant source of uncertainty as well?

Page 10, line 309, “Figure 18 shows the calculated eCL from 14 radical calibrations....” why were radical calibrations done with a NO mixing ratio of 45 ppm when the instrument is run at 30 ppm?

Page 10, line 330, “As can be seen in Figure 19...”, it would be useful to plot actual O<sub>3</sub> mixing ratio rather than set point in the top panel of this figure. It would also be useful to plot chamber pressure or channel pressure, as it seems like when the O<sub>3</sub> mixing ratio is changed the NO<sub>2</sub> signal displays a large amount of noise (50 ppb). It is also unclear what D1, D2, SG and BG stand for in this figure.

Page 11, line 364, “The pressure regulation in PeRCA based airborne instruments results in lower eCL than ground based ones.” This statement should be substantiated. The reviewer sees a range of eCL for aircraft instruments from 45 to 322 and ground based instruments a range of 91 to 1010. A description of why lower pressure or pressure regulation in general affects eCL would be useful.

Page 11, line 367, “the detection limit and uncertainty of PeRCA based instruments are strongly depending on the variation of O<sub>3</sub> and NO<sub>2</sub> in the sampled air mass...” The reviewer believes this statement should be substantiated with uncertainty and/or detection limit analysis to show the reader the magnitude of this affect. If the changes in O<sub>3</sub> and NO<sub>2</sub> are measured (as often are in aircraft field campaigns) can a correction method not be proposed and evaluated?

Page 12, line 379, “As can be seen in figure 20...” this reviewer believes it would be useful to add the detector temperature (or deltaT used earlier) to this figure, as this was determined to be a large effect on  $\Delta\text{NO}_2$  earlier in the manuscript. If NO<sub>2</sub> and O<sub>3</sub> mixing ratio data is available from the flight, this would be useful to include as well.

Page 12, line 383, “Figure 22...” the reviewer does not believe this or figure 21 add considerable value to this paper. Recommend removing or moving to SI.

Page 12, line 387, “...illustrate the improvement in the dynamical stability achieved in successive airborne deployments...” the reviewer finds it difficult to see the improvements made in the measurement by looking at time series data. Suggest thinking of a different way of presenting this conclusion. It would also be very useful to add a comparison to the previous generation of instrument somewhere in the paper.