Response to RC1

General comments and suggestions

(1) Comments from Referee: The researchers appear to accept the basic characteristics of the inlet/reactor as given. The paper does not justify the selection of flow rates, inlet/reactor volume and composition, and thus the corresponding reaction time. This reviewer sees this approach as flawed. Perhaps this has been well thought out, and previously published, but not included in this paper. Suggest that the authors include some discussion of why these parameters were selected, and what the compromises and advantages in their selection are.

(2)Response to RC1: The objective of the present publication is to explain the dominant factors affecting the overall performance and accuracy of PeRCEAS for the determination of the hydroperoxyl, HO_2, and organic peroxy radicals, RO_2, which react with NO to form NO_2, when deployed on the the HALO aircraft. The operating conditions of PeRCEAS are optimised for the specific sampling position used, cabin location of the instruments, safety requirements and the type of flight tracks and altitude profiles which were flown by HALO. Generally, these limitations are often different in different campaigns. For this reason, this manuscript does not aim at describing a unique universal set of PeRCEAS operating conditions. In practice the mechanical constraints and the safety requirements of the HALO (e.g. the size and weight of the pylon for the inlet, the amount of CO permitted on board) determine the volume and shape of the reactors and partly the range of flows of the gases and the residence time within the PeRCEAS. More detailed information about the inlets DUALER I and DUALER II and their differences are now provided in section 2 (see answers to specific comments).

(1) Comments from Referee: One aspect is that higher NO levels used in the studies reported in this paper (30-45 ppmv) result in lower sensitivity to CH_3O_2 (and other RO_2) compared to previous values because of a faster rate of $CH_3O+NO+M$. High NO also converts a greater fraction of ambient ozone to NO_2, although complete conversion is not necessary. Most other chemical amplifiers used NO reagent mixing ratios of 2 to 6 ppmv.

(2)Response to RC1: The selection of the concentration rather than the mixing ratio of NO is indeed a critical issue for the PERCA approach. High concentrations of NO are required to guaranty the full titration of O_3 in NO_2 to capture fast variations of O_3 within a measurement cycle. The total conversion of O_3 to NO_2 in the system enables the quantification of radical data at a 60s temporal resolution, as explained now in figure 4. In this way, the horizontal resolution of the PeRCEAS airborne measurements, which depends on the speed and altitude of HALO, is typically between 7 and 15 km. Longer modulation cycles than 120 s result in noisy and unrepresentative averages for ambient measurements in air masses having significant short term variability of O_3 and NO_2. Provided the partial conversion is stable and identical for both reactor-detector lines of the PeRCEAS, we agree with RC1 that the partial conversion of O_3 to NO_2 is sufficient to determine RO_2^*. Malfunctioning of one of the detectors yields the RO_2^* to be determined at 120 s. In this case, if O_3 is completely converted and the simultaneous O_3 and NO_2 measurements on board are of sufficient accuracy, a RO_2^* time resolution of 60 s may still be feasible.

The majority of PERCA measurements in the literature using NO concentration in the range 3-6 ppm were made at 1 atmosphere i.e. concentrations of (7.3-14.6) x 10^{13} molecule cm^{-3}. At 300 mbar these correspond to the mixing ratio

range 10-20 ppm. We expect that at concentrations of NO of 14.6 x 10^{13} molecule cm^{-3}, i.e. a mixing ratio of 20 ppm, the ratio of eCL(CH_3O_2)/eCL(HO_2) is 65 % and 40 % for 45 ppm NO.

As now explained in the section 3.3. and in the response to the corresponding specific question below, the water vapour dependence of eCL decreases significantly from 10 ppm to 45 ppm NO ([NO] 7.29 x 10^{13} to 3.28 x 10^{14} molecules cm^{-3} at 300 mbar). The results in figure 14 for 45 ppm NO indicate that variations in the sample humidity do not lead to additional uncertainty in the RO_2^* retrieval as the PeRCEAS eCL remains invariable within the experimental error up to [H_2O] ~ 1.4 x 10^{17} molec cm^{-3}.

The final selection of [NO] will be a balance between having stable eCL with respect to water vapour concentrations and having smaller eCL for RO_2 measurements. As now shown in Table 2 the eCL of RO_2 for 300mbar and NO 45 ppm, assuming to be CH_3O_2 the dominant atmospheric RO_2, would be 40% of the eCL for HO_2. The sum of HO_2 + 40% RO_2 can then be compared with atmospheric model values to test our understanding of the production of HO_2 an RO_2 in air masses sampled in flight.

(1) Comments from Referee: It is not apparent why the response time of the system is so slow. For example, the Figure 5 caption states that 20 seconds is eliminated after change in NO_2. Given a reactor transit time of 3 seconds, this seems extreme. There may be delays, which can be accounted for in data analysis, that are different than transition to the correct value after perturbations.

(2) Response to RC1: The PeRCEAS operating conditions have now been explained more in detail in the text and supplementary information (see answers to specific comments). During calibration measurements, different NO_2 mixing ratios are generated by the dilution of a mixture of NO_2 in synthetic air from a commercially certified gas cylinder (Airliquid 10 ppmv NO_2 in N_2). As illustrated in the Figure RC1_I below, the flow controller used for adding different NO_2 mixing ratios to the detector requires approximately 10s second to reach the set value after a change. If this time is added to the time required for the probe to reach the detector (see Table 3; 5.27 s), at least 16 seconds of data should be ignored after a change in the NO_2 mixing ratio.

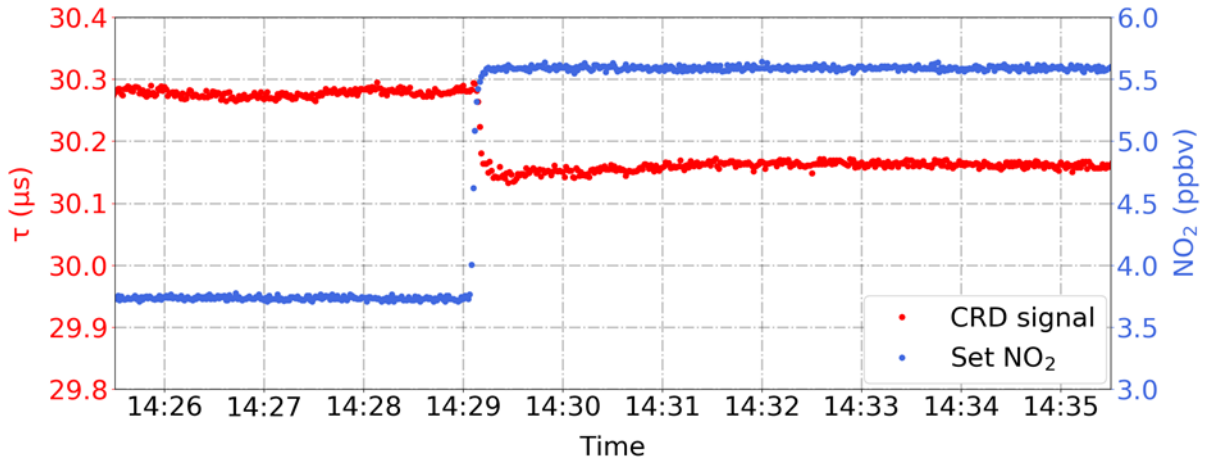

Figure RC1_I: PeRCEAS response to a step change in sampled NO_2 mixing ratio

(1) Comments from Referee: Part of the answer may be in Figure 19, where step functions in the O_3 concentration result in perturbations lasting about 40 seconds.

(2)Response to RC1: The O_3 concentration and mixing ratios were changed by adjusting the speed of the flow passing through a Hg lamp, which photolyses O_2. When the O_3 concentration is changed, it takes a time for the ozone generator to stabilise the O_3 concentration in its flow and a time lag required for the probe to reach the detector. This is the reason for the changes in figure 19 (now figure 17).

(1) Comments from Referee: Also, in Figure 20, the pressure variations last for a long time during and after altitude changes. There are also pressure fluctuations even when the aircraft is not changing altitude. This implies need for better pressure control. Perhaps the PID parameters of the pressure controller have not been adjusted properly. This is very important to get correct. Though improved, such fluctuations are still apparent in Figure 23. They add unnecessary noise to the measurements. Suggest adding more discussion of the pressure control system (manufacturer, model, adjustment procedures) and the response time of the system to step changes in NO_2 concentrations to allow the reader to better understand these issues.

(2)Response to RC1: To avoid misunderstanding the figure 20 (new figure 18) has been improved. In this figure the dynamic pressure changes are depicted which are the cause of fluctuation in the inlet. The changes in the dynamic pressure arise from altitude changes and from changes of the aircraft velocity (i.e., including turning) and air turbulence. Under laboratory condition the time constant for the pressure system to stabilise after a perturbation in PeRCEAS is 15s. The error induced for the RO_2^* measurements is shown in figure 20 (new figure 18) to be small or negligible as a result of turbulence or HALO velocity changes. After identifying the measurements influenced by changes in the dynamic pressure, only RO_2^* which have pressure fluctuations of less than 2 mbar in 60 s, the modulation time, are primarily used for analysis. The relevant paragraph now reads as follows (Lines 481- 487)

(3) Author's changes in manuscript: "As can be seen in the figure 18, the dynamic pressure variations experienced by the aircraft influence the stability of the inlet pressure. These changes are attributed to altitude changes, air turbulence, and changes in aircraft velocity, including turning, of the aircraft. The effect of inlet pressure instabilities on the retrieved ΔNO_2 is not exactly identical for both detector signals. This leads to additional uncertainty in the RO_2^* determination when using the procedure discussed in section 4.3. For the data analysis, pressure spikes within 1 minute standard deviation higher than 2 mbar are identified and flagged. This approach enables data with large error due to dynamic pressure changes to be identified. Overall the error in the retrieved RO_2^* is around 20 % in the measurement period shown in figure 18."

(1) Comments from Referee: Finally, why are the chain lengths reported in this paper so much lower that previous publications (Table 3)? In fact, values reported in this paper (28-38, Figure 18) are much lower than what is reported under "This Work" in Table 3 and would be the lowest values for the CO/NO chemical amplifier in the table (there are lower values for the ethane/NO chemical amplifier). This compromises the potential quality of the measurements. Perhaps this bears on the question about optimization of the instrument earlier. It seems that chain lengths of 100 or more are possible (at lower reagent NO mixing ratios). Some explanation in the paper is needed to explain this.

(2)Response to RC1: The text has been extensively rewritten to address this issue. We hope that we have removed any misunderstandings with respect to the values reported in the text and in the old table 3 (new table 4).

Specific comments and suggestions

(1) Comments from Referee: Page 1, line 16. "…for the airborne measurements in the…"

(2)Response to RC1: It has been corrected as suggested by the referee.

(1) Comments from Referee: Page 1, line 18. "…instrumental channels successfully captures short term…".

(2)**Response to RC1:** It has been corrected as suggested by the referee.

(1) Comments from Referee: Page 1, line 20. Not sure why the word "gradients" is used here. How about "…range of atmospheric pressures and temperatures expected…"?

(2)Response to RC1: It has been changed as suggested by the referee.

(1) Comments from Referee: Page 1, line 24. The phrase "…collectively known at RO_2*…". Is it true that HO_2+RO_2 is "known" as RO_2*? Is this the term accepted by the community? This reviewer suggests just using "HO_2+RO_2" instead.

(2)Response to RC1: RO_2^* is the term defined as $RO_2^* = HO_2 + \sum RO_2 + OH + \sum RO$ and used specifically for the PeRCA measurements. The text has been accordingly modified.

(1) Comments from Referee: Page 1, line 29. Suggest removing the summation symbol, since there is a plus sign used.

(2)**Response to RC1:** It has been corrected as suggested by the referee.

(1) Comments from Referee: Page 1, line 32-33. Suggest "…photolyzed to ultimately produce…"

Response to RC1: It has been changed as suggested by the referee.

(1) Comments from Referee: Page 2, line 34. Suggest "Overall, HO_2+RO_2 influences the…"

(2)Response to RC1: It has been changed as suggested by the referee.

(1) Comments from Referee: Page 2, line 41. Not sure what is meant by "those RO_2".

(2)Response to RC1: The text has been extended for clarification. Lines 41-50:

(3) Author's changes in manuscript: "The chemical amplification technique (Cantrell and Stedman, 1982; Hastie et al., 1991) has been used to measure the sum of peroxy radicals. The Peroxy Radical Chemical Amplification (PeRCA) converts by addition of NO and CO, HO_2 and most atmospherically significant RO_2 to NO_2. The OH formed in the reaction cell reacts with CO to reform HO_2 in a chain reaction. Oxy, alkoxy, hydroxy and alkylperoxy radicals ($OH + \sum RO + HO_2 + \sum RO_2$) are converted into NO_2. As the RO and OH abundances in the troposphere are much lower than those of HO_2 and RO_2, PeRCA measures to a good approximation the sum of peroxy radicals collectively known as RO_2^*, ($RO_2^* = HO_2 + \Sigma\ RO_2$; being R any organic chain), which convert NO to NO_2. The rate coefficients of the HO_2 and RO_2 reactions with NO are very similar (Lightfoot et al, 1993). Large RO_2 which do not react with NO to form NO_2 are not detected, and are assumed to be negligibly small compared to the sum of $HO_2 + \sum RO_2$ concentrations. HO_2 and CH_3O_2 are the dominant peroxy radicals present in an air mass in most conditions."

(1) Comments from Referee: Page 2, line 43. Suggest "…compared to the total amount of HO_2+RO_2, with HO_2…"

(2)Response to RC1: This has been changed as suggested by the referee.

(1) Comments from Referee: Page 2, line 53. Should be "Kanaya".

(2)Response to RC1: This has been corrected as suggested by the referee.

(1) Comments from Referee: Page 2, line 54. Suggest a different word than "largely". Suggest "The interference by some RO_2…"

(2)Response to RC1: This has been changed as suggested by the referee.

(1) Comments from Referee: Page 3, line 72. Add space "Peroxy Radical…".

(2)Response to RC1: This has been corrected as suggested by the referee.

(1) Comments from Referee: Page 3, line 74. Suggest "…in a previous publication…".

(2)Response to RC1: This has been changed as suggested by the referee.

(1) Comments from Referee: Page 3, line 79. Suggest "…where ΔNO_2 is the NO_2 formed…"

(2)Response to RC1: This has been changed as suggested by the referee.

(1) Comments from Referee: Page 3, line 84. Suggest "…spectroscopic measurement technique…"

(2)Response to RC1: This has been changed as suggested by the referee.

(1) Comments from Referee: Page 3, line 87. NO_2 comes from radical amplification and from the background (O_3 conversion). Suggest changing the sentence to reflect this.

(2)Response to RC1: The sentence has been reworded. Line 102:

(3) Author's changes in manuscript: "In PeRCEAS the absorber of interest is NO_2 which is formed in both the amplification and the background modes. "

(1) Comments from Referee: Page 3, line 92-94. Suggest also saying that c_0 is the speed of light in a vacuum (stated later in the paper).

(2)Response to RC1: The text has been extended as suggested by the referee.

(1) Comments from Referee: Page 3, line 92. Suggest "…are, respectively, the absorption…"

(2)Response to RC1: This has been changed as suggested by the referee.

(1) Comments from Referee: Page 3, line 95. Suggest "…used for ground-based measurements…"

(2)Response to RC1: This has been corrected as suggested by the referee.

(1) Comments from Referee: Page 3, line 96-7. Suggest "…the particular constraints related to airborne measurement…".

(2)Response to RC1: This has been changed as suggested by the referee.

(1) Comments from Referee: Page 3, line 99. Suggest "In this study, the specifications…are described based on thorough laboratory…"

(2)Response to RC1: The text has been changed as suggested by the referee.

(1) Comments from Referee: Page 4, line 107. Suggest (if correct) "…and located outside the HALO fuselage…"

(2)Response to RC1: The pylon is a part of the fuselage. The sentence has been reworded. Line 123:

(3) Author's changes in manuscript: "….installed inside a pylon located on the outside of the HALO fuselage"

(1) Comments from Referee: Figures 1, 2, and 3. While photographs can be nice, schematic diagrams are more useful to see the path of sample, reagents, and signals throughout the system. Suggest limiting to only one or two photographs and add diagrams to show the details.

(2)Response to RC1: The photos have been replaced by schematic diagrams as suggested by the referee.

(1) Comments from Referee: Page 4, line 117. Mixing and pressure regulation are mentioned here, but no detail is given. This is relevant to the general comment given earlier. Suggest adding more detail in the text and perhaps in Figures 1 and 2. Suggest discussing how DUALER I and II are different and describe why the changes do indeed result in improved performance.

(2)Response to RC1: The text has been extended with the description of the DUALER operation and Figure 2 has been included to highlight differences between DUALER I and II. Lines 127 to 151:

(3) Author's changes in manuscript: "Briefly, sampled air enters PeRCEAS through the DUALER pre-chamber, which is at a lower pressure than that outside of the HALO, through an orifice in a truncated cone, i.e. a nozzle. From this pre-chamber the air is pumped simultaneously through the two flow reactors and a bypass line. At the upper addition point a mixture of CO or N_2 and NO enters each reactor. At the lower addition point, a flow of N_2 or CO enters each reactor. This enables the CO and N_2 flows in the two reactors within the DUALER to be switched simultaneously but out of phase with one another from the upper to the lower addition point. At the addition points, the reagent gases enter the reactor through eight circular distributed 1 mm holes to facilitate the rapid mixing with the sampled air. During measurements, the pressure in the pre-chamber and both reactors is held constant. However, there is a small pressure fluctuation during the switching of flows between the upper and lower mixing point. The flow passing through each reactor enters a CRDS NO_2 detector. Afterwards, the sample flows together with the air from the bypass line are scrubbed for CO and NO and exhausted by the pump.

The DUALER inlet comprises two PeRCA chemical reactors having alternating measurement modes, which are out of phase with one another. During the first part of the measurement cycle, the first reactor and detector are in amplification mode, while simultaneously the second reactor and detector are in background mode. In the second part of the cycle, the CO addition point in both reactors is switched. Consequently, the first reactor and detector are then in background mode while the second reactor and detector are in amplification mode. In the analysis of the measurements, the amplification and background signals from both detectors are combined appropriately. This improves accuracy and temporal resolution of the resultant RO_2^* data set (see 3.1).

In the DUALER, a stable pressure in the pre-chamber is achieved by a pressure regulator, which controls the flow through the bypass line. As noted the flow rate through the reactors is held constant during measurements. Consequently, when the outside air pressure changes, the bypass flow rate from the pre-chamber is changed. The outer dimensions, shape, form and weight of the DUALER are constrained by the inlet pylon in use with the research aircraft HALO. After the first version of the DUALER (from now on called DUALER I) was flown, the inner dimensions of the pre-chamber were further optimised to reduce the wall losses and turbulence in the pre-chamber. For this, in the DUALER II the volume of the pre-chamber was increased by extending its vertical extent, the length of the truncated cone on top of the reactors was reduced in 3 mm, and the volume of the reactors was increased to 130.5 ml from the 112 ml in DUALER I. These changes resulted in a higher eCL and improved pressure stability in DUALER II as compared to DUALER I. Figure 2 shows the upper part of both DUALER I and DUALER II."

(1) Comments from Referee: Page 4, line 124. The term "piezo electric stack" is not a common term and needs more description.

(2)Response to RC1: A piezo electric stack features a longitudinal deformation when voltage is applied. A mirror mounted on piezo electric stack was used to achieve mode matching between the single mode laser and resonator in Hortsjann et al., 2014. This is not used in the current configuration of PeRCEAS.

The text has been accordingly modified (now lines 158-159).

(3) Author's changes in manuscript: "With this, the fine adjustment of the laser is simplified and improved, and the piezo electric stack used to achieve mode matching between the single mode laser and the optical cavity in Hortsjann et al., (2014) becomes unnecessary and is removed."

(1) Comments from Referee: Page 4, line 128. Suggest adding the manufacturer of the beam camera (MKS Ophir). This is very cool, by the way!

(2)Response to RC1: The text has been extended. Line 162:

(3) Author's changes in manuscript: "During alignment procedures and for test purposes, a beam camera (BM-USB-SP907-OSI, Ophir Spiricon Europe GmbH) monitors the beam profile and simplifies the identification of misalignments or loss of performance of the optical system."

(1) Comments from Referee: Page 4, line 132-3. Suggest "…other sensor data such as pressure, flow, temperature, and humidity." You don't need "etc." if using "like" or "such as".

(2)Response to RC1: This has been corrected as suggested by the referee.

(1) Comments from Referee: Page 5, line 149. Would "occasionally" be better than "exceptionally"?

(2)Response to RC1: This has been changed as suggested by the referee.

(1) Comments from Referee: Page 5, line 150-152. The term that should be used is "Allan Variance" rather than "Allan Deviation". Perhaps the equation for its calculation should be given, since there is also modified Allan Variance that can be used. Also, give one or two references (e.g. Allan, 1966; Allan et al., 1991; Allan and Levine, 2016). Suggest "…was investigated using calculated Allan Variance in the measurement…". Also "…the optimum integration time for the three PeRCEAS detectors is between 20 s…". Do you use 20-30 second averaging in the data analysis? The timing of the instrument cycling should be shown, perhaps in Figure 1 or in a separate figure.

(2)Response to RC1: The analysis of the Allan deviation was used to infer the detection limit of the measurement resulting from random noise. The plot of the Allan deviation has been replaced by another plot of the Allan variance as suggested by the referee. The text has also been extended for clarification. Lines 195-206:

(3) Author's changes in manuscript: " To optimize the mode time and thus also the modulation cycle, the Allan variance (Allan, 1966; Werle et. al., 1993) was analysed for PeRCEAS. Given a time series of N elements and a total measurement time t_{acq}, $t_{acq} = f_{acq} \cdot N$, where f_{acq} is the frequency of acquisition, then the Allan variance is defined as:

$$\sigma_x^2(\tau) = \frac{1}{2}\langle (x_{i+1} - x_i)^2 \rangle_\tau \qquad \text{(Eq.2)}$$

where x_i is the mean over a time interval of a length τ, being $\tau = f_{acq} \cdot m$; and m the number of elements in a selected interval. The use of $\langle \ldots \rangle$ denotes the arithmetic mean. The square root of the Allan variance is the Allan deviation. For random noise, the Allan deviation at any given integration time determines the detection limit of the measurement.

The Allan variance plot for measurements of 5.6 ppbv NO_2 at 200 mbar and 23 °C is shown in figure 5. As can be seen, the optimal averaging time for the three PeRCEAS detectors is in the range between 20 s and 50 s. The corresponding minimum (2σ) detectable mixing ratio is < 60 pptv (3.15×10^8 molecules cm^{-3} for these P and T conditions). Slow temperature drifts over longer averaging times impact on both the laser and the resonator characteristics. This behaviour is observed for averaging times longer than 60 s."

(1) Comments from Referee: Page 5, line 155. The phrase "…over the modulation time…" need explanation. This might be apparent with addition of a figure showing the instrument cycle timing.

(2)Response to RC1: The text has been extended for clarification at the beginning of 3.1. (Lines 181-194), and Figure 4 has been added:

(3) Author's changes in manuscript: "The mode time is defined as the time selected for the measurement in either amplification or background mode. The modulation time is the time taken for a complete measurement cycle, which comprises the sum of one amplification and one background mode. The PeRCEAS measurement cycle is illustrated in Figure 4. The ΔNO_2 for each detector is calculated from the ring down time of two consecutive modes using Eq.(1). If the mode time is adequately selected, the RO_2^* retrieved per measurement cycle is identical in both measurement lines, as the two reactors are operated out of phase with one another. The final RO_2^* data is calculated as the mean of the RO_2^* determined from the ΔNO_2 and eCL of both detectors for a given measurement cycle. The time resolution of the RO_2^* measurement is then equal to the mode time. After switching modes, a small pressure pulse leads to an oscillation of the NO_2 signal. Consequently, the first 20 s of each mode are not used in data analysis. The time lag arising from the time taken for the sample flow between the CRDS detector and the point of switching is typically less than 8 s (see Table 3).

Typically, 650 to 800 ring down times of the NO_2 absorption are averaged per second and the measurement of NO_2 is made at 1 Hz. Individual ring down times are occasionally saved for sensitivity studies. Modulation and mode times are selected empirically. The optimised values are a compromise between the time taken for the detector signal to stabilise after the CO/N_2 flow is switched between the addition points, and the temporal variability of the chemical composition of the air probed"

(1) Comments from Referee: Page 5, line 161. Suggest "…signals generated that minutely varied the sampled NO_2…"

(2)Response to RC1: The paragraph has been rewritten for clarification. Lines 211-217:

(3) Author's changes in manuscript: "Temperature changes of the CRDS affect: i) the diode laser emission, both its amplitude and wavelength; ii) the mode matching between laser and detector, and consequently the τ_0. The effect of the variations in τ, resulting from changes in room or HALO cabin air temperatures, on the accuracy and precision of the ΔNO_2 determination was investigated by a series of laboratory experiments. For this, modulated concentrations of NO_2 in the flow were generated. This was achieved by alternating between two selected NO_2 concentrations once per minute. The temperature of the CRDS detector, T, and τ were then measured. Detector temperature gradients over a time t, i.e., $\Delta T/\Delta t$, determined by the temperature within the CRDS housing close to the photodiode detector, were induced by controlled changes in the room temperature."

(1) Comments from Referee: Page 5, line 164. "…time resolution using each background…". This will be more obvious with a graphical representation.

(2)Response to RC1: Now the figure 4 depicts the PeRCEAS measurement cycle.

(1) Comments from Referee: Page 5, line 167. "… molecules cm^{-3} for typical measurement conditions".

(2)Response to RC1: This value refers to the value in molecules cm^{-3} corresponding to 150 pptv at the particular T and P of the measurement. The text has been changed for clarification. Line 220:

(3) Author's changes in manuscript: "….the experimental precision of the ΔNO_2 determination remains within (2σ) 150 pptv (= 7.3×10^8 molecules cm^{-3} at 200 mbar and 23°C). "

(1) Comments from Referee: Page 5, line 170. "It is inferred from laboratory calibrations that a 60 s modulation cycle is an optimum comprise between…". The term "modulation cycle" is not apparent here, but perhaps would be with a graphical representation of the instrument cycle. Also, add more discussion why fluctuations last so long (20 s).

(2)Response to RC1: Figure 4 now depicts the measurement cycle. The text has also been extended as suggested by RC1 to clarify the modulation cycle (see lines 181-190:

(3) Author's changes in manuscript: "The mode time is defined as the time selected for the measurement in either amplification or background mode. The modulation time is the time taken for a complete measurement cycle, which comprises the sum of one amplification and one background mode. The PeRCEAS measurement cycle is illustrated in Figure 4. The ΔNO_2 for each detector is calculated from the ring down time of two consecutive modes using Eq.(1). If the mode time is adequately selected, the RO_2^* retrieved per measurement cycle is identical in both measurement lines, as the two reactors are operated out of phase with one another. The final RO_2^* data is calculated as the mean of the RO_2^* determined from the ΔNO_2 and eCL of both detectors for a given measurement cycle. The time resolution of the RO_2^* measurement is then equal to the mode time. After switching modes, a small pressure pulse leads to an oscillation of the NO_2 signal. Consequently, the first 20 s of each mode are not used in data analysis. The time lag arising from the time taken for the sample flow between the CRDS detector and the point of switching is typically less than 8 s (see Table 3)."

(1) Comments from Referee: Page 5, line 172. The detection for NO_2 should be performed with a background level of ozone in the sample, since this is how ambient measurements are performed. Was this is the case?

(2)Response to RC1: As described in the text, the detection limit for NO_2 was determined by measuring a modulated signal generated by dilution of NO_2 from commercial standard cylinders in synthetic air to get 11.5 and 12.1 ppbv NO_2 as background and amplification signals respectively. This is equivalent to adding a background level of O_3, since O_3 is converted totally in NO_2 before reaching the detector. In further complementary measurements it has not been observed any significant variation in the NO_2 detection limit for variations up to 100 ppb in the O_3 background produced by an ozone generator.

(1) Comments from Referee: Page 5, line 173-4. It is not obvious that larger modulation times lower the representativeness of the averages. Do you mean variability in peroxy radicals or in the background? For the latter, the
instrument is continually measuring the background, and it should be well accounted for. If you mean the peroxy radicals, while one-minute (or quicker) data are nice, longer averages can still be useful in adding understanding of tropospheric free radical behavior. Suggest rewording the last sentence of this paragraph.

(2)Response to RC1: In instruments based on PERCA modulated signals, the modulation time determines the resolution. In contrast to ground based measurements, as the airborne platform additionally moves horizontally and vertically, modulation times longer than 120 s can be critical to mirror the peroxy radical and background variabilities in the encountered air masses.

This part of the text has been rewritten to address the criticism of the referee and moved to 3.2.2 for clarification (Lines 310-316):

(3) Author's changes in manuscript: "As explained in section 3.1. the simultaneous use of two detectors measuring out of phase results in the temporal resolution of the RO_2^* data being 60s. In this way, the horizontal resolution of the PeRCEAS airborne measurements, which depends on the speed and altitude of HALO, is typically between 7 and 15 km. Longer modulation cycles than 120 s result in noisy and unrepresentative averages for ambient measurements in air masses having significant short term variability of O_3 and NO_2. To keep the temporal resolution of the RO_2^* data to be equal to the mode time, the rapid and complete conversion of ambient O_3 into NO_2 within the PeRCEAS is required. For this, the NO concentration added at the inlet has to be sufficient for a complete titration of the sampled O_3 before reaching the detector."

(1) Comments from Referee: Page 6, line 176. "Sample and reagent gas flows...".

(2)Response to RC1: It has been changed as suggested by the referee.

(1) Comments from Referee: Page 6, line 176-184. This discussion of reaction time should also include discussion of how the size of the reactor was selected, since it also affects the reaction time (reactor volume / total flow). Perhaps this would be a good place to discuss the approach to ensuring mixing of reagent gases with the ambient air sample. How was this done? Were fluid dynamical calculations performed? Were flow visualization approaches used? Related to this: how do you ensure that no components of the inlet system are leaking?

(2)Response to RC1: The length of the reactor is limited by the design of the pylon. Therefore the reaction time is constrained by the mechanical borders and the flow conditions selected. The description in section 2 has been extended for clarification.

The reagent gases are added to the reactor through eight 1mm holes distributed circularly around the reactor tube in order to maximise the mixing with the ambient air sample.

The tightness of the inlet is controlled regularly by applying an overpressure of 0.8 mbar after closing all openings of the inlet and using leak soap at the connections. CO leakages in the aircraft are a critical safety issue in the aircraft. Therefore the inlet tightness is checked before installation and the potential critical points of the instrument are tested by adding He overpressure and using a He detector.

(1) Comments from Referee: Page 6, line 182. "…lower explosion limit (LEL) in air of 12.5% v/v at room temperature…".

(2)Response to RC1: The text has been changed as suggested by the referee.

(1) Comments from Referee: Page 6, line 189. Remove extra space between "air" and "and", and between "CO" and "in".

(2)Response to RC1: This has been corrected as suggested by the referee.

(1) Comments from Referee: Page 6, line 190. Suggest "…between safety requirements, limiting…".

(2)Response to RC1: This has been corrected as suggested by the referee.

(1) Comments from Referee: Page 6-7. Effective chain length. This might be good place to discuss experiments to determine the optimum NO concentration for the amplifier chemistry. Also, perhaps near the end of this section, discuss how the effective chain length values are used in the data analysis. In other words, have estimates of the HO_2/RO_2 ratio been made and used to apply the two eCL (for HO_2 and $HO_2+CH_3O_2$) values? If so, how is it done?

(2)Response to RC1: The text of section 3.2.1: Effective chain length has been rewritten to address this criticism. Additional experiments were undertaken to determine the eCL as a function of [NO]. These are reported in figure 7 and also in table 2. The concentration or mixing ratios of HO_2 and RO_2 are not known in ambient air. Thus the ratio is also not known. The RO_2^* values are reported to be the sum of $HO_2 + \alpha{\cdot}RO_2$. Using CH_3O_2 as a surrogate for all RO_2 , the values of α, which depends on [NO], have been determined by modeling and measurement in Table 2. The relevant paragraph in section 3.1.1 now reads as follows (Lines 302-306):

(3) Author's changes in manuscript: "Table 2 summarises the simulated PeRCEAS sensitivity for the HO_2 and CH_3O_2 detection for different NO mixing ratios in the reactor at 300 mbar. Up to 10 ppm NO ([NO] 7,29 x 10^{13} molecules cm^{-3}) the difference in sensitivity remains within the PeRCEAS uncertainty. The ratio of the eCL_{CH3O2} /eCL_{HO2} is defined as α. The estimated values of α from modelling and measurements are given in table 2. For the assessment of air masses the measurements of $HO_2 + \alpha{\cdot}RO_2$, where $\alpha RO_2 \approx \alpha{\cdot}CH_3O_2$, are compared with atmospheric model values of $HO_2 + \alpha{\cdot}RO_2$."

(1) Comments from Referee: Page 6, line 197. "…to the conversion into RO_2.". Include that the approach is based on O_2 actinometry, as opposed to other approaches reported in the literature (such as N_2O actinometry, calibrated NIST photodiodes).

(2)Response to RC1: The text has been extended for clarification. Lines 247-258:

(3) Author's changes in manuscript: "The eCL of the DUALER reactors is determined in the laboratory by using a calibrated source of peroxy radicals. The latter uses the photolysis of water vapour at 184.9 nm (see Schultz et al., 1995). Briefly, a known water vapour - air mixture is photolysed by a low pressure mercury (Hg) lamp. A nitrous oxide (N_2O) absorption filter attenuates the intensity of 184.9 nm radiation. This is achieved by varying the N_2O/N_2 ratio in the filter absorption zone. The photolysis of H_2O makes an OH and H. In air, the H reacts with O_2 in a termolecular reaction to make HO_2. The photolysis of oxygen molecules yield oxygen atoms, O which react with O_2 in a termolecular reaction to make O_3 (see Reichert et al., 2003). CO is added to the gas mixture in the source to convert the OH into HO_2 radicals. As a result, each absorbed photon by a water vapour molecule generates two HO_2 molecules. Alternatively, the addition of a hydrocarbon, RH, leads to the conversion of OH to a RO_2, and consequently to a 1:1 mixture of HO_2 and RO_2 for calibration. The concentration of HO_2 or RO_2, and O_3 is thus proportional to the intensity of 184.9 nm electromagnetic radiation. As the absorption coefficient of N_2O (Cantrell et al., 1997) does not change significantly around 185 nm (σ_{N2O}=14.05×10^{-20} cm^2 molecule^{-1} at 25 °C with a 0.02×10^{-20} cm^2 molecule^{-1} K^{-1}, temperature dependency), different HO_2 and RO_2 radical amounts can be produced for a constant H_2O concentration.."

(1) Comments from Referee: Page 6, line 205. "…are changed stepwise every ten minutes from 8 pptv…". Also, note that too much reagent added to the calibrator has the potential to affect the inlet chemistry. This can be seen by a change in the background with change in radical concentration, which is not expected if the background is mostly due to ozone.

(2)Response to RC1: The radical calibration procedure does not require any change in reagents. Different radical mixing ratios are generated by attenuation of the light of the Hg/Ne UV lamp used for photolysis of H_2O and O_2 by using different concentrations of N_2O as absorption filter, as described in previous publications (e.g, Reichert et al., 2003). In each step of the calibration both the amplification and the background signal change due to the effect of the light attenuation in the photolysis of H_2O and O_2 leading to radicals and O_3 respectively.

The text has been extended for clarification:

(3) Author's changes in manuscript: In line 255 "….The concentration of HO_2 or RO_2, and O_3 is thus proportional to the intensity of 184.9 nm electromagnetic radiation, and the absorption coefficient of N_2O (Cantrell et al., 1997) does not change significantly around 185 nm (σ_{N2O}=14.05×10^{-20} cm^2 molecule^{-1} at 25 °C with a 0.02×10^{-20} cm^2 molecule^{-1} K^{-1}, temperature dependency), different HO_2 and RO_2 radical amounts can be produced for a constant H_2O concentration."

(3) Author's changes in manuscript: In line 271: "The O_3 generated by the radical source is converted in the DUALER to NO_2 by its reaction with NO, which is in excess. Therefore the O_3 entering the reactor during the radical calibration is detected as NO_2 in the background and amplified signals."

(1) Comments from Referee: Page 6, line 206-7. "…is determined from the slope of the measured ΔNO_2 levels versus the calculated radical amounts. Example data is shown in…". Suggest rewording the end of the last sentence on this page, since the concentration of NO ***within the inlet*** is 30 ppmv. Perhaps "…and added reagent NO to achieve 30 ppmv within the inlet."

(2)Response to RC1: We agree with the comment of the referee but the original figure has been removed.

(1) Comments from Referee: Page 7, line 208. Suggest "In Figure 7, the PeRCEAS eCL versus the inlet NO concentration…". In Figure 7, why aren't data shown for lower NO concentrations, such as used by your group in the past and by other researchers?

(2)Response to RC1: This figure has been extended for eCL values at lower NO concentrations as suggested.

(1) Comments from Referee: Page 7, line 212. "…concentration, eCL values increase with…".

(2)Response to RC1: The text has been modified as suggested.

(1) Comments from Referee: Page 7, line 16-18. Suggest describing how wall losses were determined. Are they constant or are they affected by the cleanliness of the inlet? Suggest putting all the rate coefficients used for both reactor pressures into Table 1. It is interesting that a level of 3 ppbv O_3 was used, presumably because this is what comes out of the calibrator. Suggest also running the model with ambient-like levels of O_3.

(2)Response to RC1: The wall losses are not determined experimentally. The result of eCL calibrations before and after the measurement campaigns indicate that the effect of cleanliness of the inlet in the eCL is within the experimental error.

The text has been extended (Lines 289-297) and new simulations have been performed for clarification. Table 1 includes now all rate coefficients taken from JPL- Publication 15-10 (Burkholder et al, 2015). The model is initialised with 3ppb O_3 because this is the mixing ratio produced during the calibration. Sensitivity studies have shown no significant change in the simulated eCL up to 100ppb O_3. This is also confirmed by the experimental values shown in figure 17.

(1) Comments from Referee: Page 7, line 221. Inlet pre-chamber is not defined anywhere. This should be shown in the schematic diagram discussed earlier. If the radical losses in the model do not agree with what you think they arein the DUALER II inlet, suggest you perform experiments to determine what they are. The model of this simple chemistry should be much closer to the observations that a factor of two!

(2)Response to RC1: The inlet pre-chamber is now described in detail in section 2. As mentioned in the previous response, the text has been extended for clarification (Lines 280-306) and new simulations have been performed to better reproduce the pre-chamber + reactor configuration in PeRCEAS.

(1) Comments from Referee: Page 7, line 222. Suggest "…shows measured eCL versus modeled CL for the…".

(2)Response to RC1: This figure has been replaced.

(1) Comments from Referee: Page 7, line 223-4. "The $CL_{modeled}/eCL_{measured}$ ratio averages about 2 for HO_2…". Actually, the ratio is more than 2 for the 200 mB measurements and is about 2 for the 300 mB HO_2 measurements. Only the 300 mB HO_2+CH_3O_2 measurements are about 1.5. Does this mean that the inlet wall loss changes with reactor pressure? Were the rate coefficients in the model changed to reflect the reactor pressure? Perhaps the model wall loss values should be adjusted based on new laboratory measurements. The chemical amplifier chemistry is simple enough that a box model should be able to accurately reproduce laboratory data such as this. Add error bars to the points to represent total uncertainty in the measured and modeled values. Perhaps perform regressions of data.

(2)Response to RC1: As already mentioned, the text has been modified (lines 280-301) and new simulations of the pre-chamber losses have been included. The inlet wall losses are calculated using Eq.4.

$$k_w = 1.85\left(\frac{v^{1/3}D^{2/3}}{d^{1/3}L^{1/3}}\right)\left(\frac{S}{V}\right) \quad \text{(Eq.4)}$$

S is the surface area in cm^2, V the volume in cm^3, L the length and d the diameter of the flow tube in cm, v the velocity of the gas in cm s^{-1}, and D is the diffusion coefficient, which is calculated to be D_{HO2}=0.21 and D_{CH3O2}=0.14 in cm^2 s^{-1}. At different pressures the velocity of the gas in the reactor changes and therefore the k_w.

The modeling data obtained for 300mbar is now shown in Figure 8. These agree reasonably with the corresponding experimental results.

(1) Comments from Referee: Page 7, line 226. This reviewer does not like the term "titration" in this context, even though it is widely used in the community. Suggest using "conversion" instead.

(2)Response to RC1: In the case of PeRCEAS the term titration is used correctly when the conditions are selected to achieve the complete conversion of O_3 into NO_2 before reaching the detector.

The title of 3.2.2 has been replaced by "conversion of ambient O_3 into NO_2" and the text in lines 310-316 has been extended for clarification:

(3) Author's changes in manuscript: "As explained in section 3.1. the simultaneous use of two detectors measuring out of phase results in the temporal resolution of the RO_2^* data being 60s. In this way, the horizontal resolution of the PeRCEAS airborne measurements, which depends on the speed and altitude of HALO, is typically between 7 and 15 km. Longer modulation cycles than 120 s result in noisy and unrepresentative averages for ambient measurements in air masses having significant short term variability of O_3 and NO_2. To keep the temporal resolution of the RO_2^* data to be equal to the mode time, the rapid and complete conversion of ambient O_3 into NO_2 within the PeRCEAS is required. For this, the NO concentration added at the inlet has to be sufficient for a complete titration of the sampled O_3 before reaching the detector. "

(1) Comments from Referee: Page 7, line 227. This reviewer disagrees that ozone in the sample has to be completely converted toNO_2. Why is this? It seems that partial conversion, as long as it is stable, would be fine.

(2)Response to RC1: We agree with RC1 that the partial conversion of O_3 in the sample, as long as stable and identical for both reactor-detector lines of the PeRCEAS is sufficient to determine RO_2^*. However the total conversion of O_3 in the system enables the quantification of radical data at a 60s temporal resolution as explained in

figure 4. Malfunctioning of one of the reactor-detector line yields the RO_2^* to be determined at 120 s. In this case, if O_3 is completely converted and the simultaneous O_3 and NO_2 measurements on board are of sufficient accuracy, a RO_2^* time resolution of 60s may still be feasible.

(1) Comments from Referee: Page 7, line 228. Figure 9 has a lot of information that could be presented in a more straightforward way. Suggest plotting the ozone lifetime (or three lifetimes) versus the reactor NO at the two pressures. This could be shown in one plot.

(2)Reply to RC1: The figure 9 has been changed as proposed by the referee.

(1) Comments from Referee: Page 7, line 229. There is no reason to require conversion of 100-200 ppbv of ozone to 1-2 pptv in the inlet. Conversions of 99% are more than sufficient. Suggest changing this paragraph as Figure 9 is changed.

(2)Reply to RC1: The text has been changed for clarification (Lines 310-322).

(1) Comments from Referee: Page 7, line 230. The wavelength 409 nm is mentioned, but everywhere else it indicates that the lasers operate at 408 nm.

(2)Reply to RC1: This is a typo and has been corrected.

(1) Comments from Referee: Page 7, line 236. Suggest "…which are captured by…". This reviewer disagrees that the radicals and NO_2 from PAN-like compounds cancels and does not lead to interference. Yes, the NO_2 from the decomposition should be like ambient NO_2 and be corrected for by the background measurement. But the radicals formed from the decomposition will amplify and appear like ambient radicals. This is an interference! Suggest rewording this paragraph. Some direct laboratory measurements of the interference would also be helpful. Figure 11 shows that the PAN interference is greater at lower reactor pressures. Why would this be the case. At reduced pressure, the decomposition is slower and the time is the reactor is shorter. Suggest checking the modeling.

(2)Response to the RC1: The time of reaction of CH_3CO_2 produced by the decomposition of PAN with NO is the same in both amplification and background modes. The decomposition of PAN is therefore a potential interference, if PAN decomposes between the upper and lower gas addition points in the reactor working in amplification mode. Taking into account the residence times given in Table 3 and the reactor temperatures showed now in figures 18 and 19, for most operating conditions the potential interference will remain below 2 pptv and can be considered negligible (see also answer to RC2).

The chemistry involved in the formation of CH_3O_2 from the PAN decomposition has now been revised, and all the rates have been taken from the recommendations in JPL 15-10, with the equilibrium rate constant from Zhang et al., (2011).

There is no significant difference in the production of CH_3O_2 radicals with the pressure. The differences observed in the graph are the result of the conversion of molecules·cm^{-3} in mixing ratios.

The figure has been updated and the text has been modified for clarification. Lines 324-339:

(3) Author's changes in manuscript: "Peroxyacyl nitrates ($RC(O)OONO_2$) such as peroxyacetylnitrate, PAN and peroxypropionyl nitrate can decompose thermally inside PeRCEAS. The extent of the decomposition to peroxy radicals and NO_2 depends on the time and the temperature. If the thermal decomposition occurs at shorter time scales than the modulation time, they can be a significant interfering source of radicals which are chemically amplified and lead to additional NO_2. In a rapidly changing background the RO_2^* determination might be affected according to the temperatures and sample residence times between the gas addition points in the DUALER (Table 3).

To evaluate this effect the production of peroxy radicals from the thermal decomposition of 1 ppb PAN at different temperatures and pressures has been simulated. The results obtained with a box model (Ianini, 2003) including the reactions:

$CH_3COO_2NO_2 \rightarrow CH_3COO_2 + NO_2$ (R1)

$CH_3COO_2 + NO \rightarrow CH_3 + CO_2 + NO_2$ (R2)

$CH_3 + O_2 + M \rightarrow CH_3O_2$ (R3)

are depicted in figure 11. The rate coefficients used are taken from Burkholder et al., (2015).

The $[CH_3O_2]$ produced does not vary significantly at the pressures investigated. As the temperature of the PeRCEAS reactors during flight generally remain under 290 K, this source of radicals is considered to be negligible for most operating conditions. The thermal stability of the PAN analogues is similar to that of PAN but they are usually at much lower concentrations than PAN in the atmosphere and also assumed to be a negligible source of error."

(1) Comments from Referee: Page 8, line 259-260. It is not clear what is meant by "based on the similarity of the eCL values". Suggest rewording this sentence, and perhaps this entire paragraph to make the message clearer.

(2)Response to RC1: It was concluded that the eCL dependency observed was related to the relative humidity and not to the absolute $[H_2O]$ because the eCL measured by Reichert et al., (2003) at 20°C and 30°C did not differ within the experimental errors although corresponding to significantly different absolute water concentrations. The text has been modified for clarification. Lines 354-358:

(3) Author's changes in manuscript: "The effect of changes in the sampled air humidity on the eCL has been reported and studied by Mihele and Hastie, (1998) and Mihele et al., (1999). Reichert et al. (2003) investigated the dependency of the eCL for ground based measurements at 20 °C and 30 °C and standard pressure, i.e., keeping the relative humidity but almost doubling the absolute water concentration. The obtained eCL values did not differ within the experimental error and confirmed the dependency of eCL on the relative humidity. All these measurements were performed at a pressure of one atmosphere and for 3.3 ppmv NO ([NO] 8.12 x 10^{13} molecules cm^{-3})."

(1) Comments from Referee: Page 8, line 263. Suggest replacing "as shown exemplary" with "with an example shown".

(2)Response to RC1: The text has been modified.

(1) Comments from Referee: Page 8, line 265. "…sample humidity do not lead to…".

(2)Response to RC1: The text has been modified.

(1) Comments from Referee: Page 8, line 267. "…is subject to two types of errors which either are: a) intrinsically…"

(2)Response to RC1: The text has been modified as suggested.

(1) Comments from Referee: Page 8, line 268. "…in the laboratory, or b) result…"

(2)Response to RC1: The text has been modified as suggested.

(1) Comments from Referee: Page 8, line 273. Equation 3 is very similar to Equation 1. Suggest eliminating Equation 3 and referring back to Equation 1 in this discussion. Perhaps change Equation 1 slightly, if needed.

(2)Response to RC1: The text has been modified as suggested and Equation 3 has been eliminated.

(1) Comments from Referee: Page 9, line 279. "…Vandaele et al. [2002] with the normalized laser spectrum from the corresponding detector.". Also, "The values obtained have been…"

(2)Response to RC1: The text has been modified as suggested.

(1) Comments from Referee: Page 9, line 284. "…depicts a sample comparison of spectra…".

(2)Response to RC1: The figure has been moved to the supplementary information as suggested by RC2 and the text has been modified. Lines 387-388 :

(3) Author's changes in manuscript: "A sample comparison of spectra obtained for the three PeRCEAS detectors is included in the supplementary information (Figure SI-1)."

(1) Comments from Referee: Page 9, line 294. "The effective σ_{NO2} obtained agrees within…".

(2)Response to RC1: The sentence has been corrected.

(1) Comments from Referee: Page 9, line 297. The tau symbol disappeared. You have a lot of ambient data. Does τ_0 vary significantly as the CRDS cell mirrors are exposed to ambient air?

(2)Response to RC1: The τ_0 symbol disappeared during the processing of the document. This is a typo and has been corrected. The variation in the value of τ_0 during the measurement depends on the composition of the air probed. As shown in figure 1, the sample air goes through a 5µm filter before reaching the PeRCEAS detectors. This filter removes most of the ambient particles. As a consequence there is only a gradual change in τ_0 over the 10 hours flight which is not critical for the measurement. The filter is replaced after each flight.

(1) Comments from Referee: Page 9, line 300. "…measurement requires accurate…"

(2)Response to RC1: The text has been modified as suggested.

(1) Comments from Referee: Page 9, line 302-3. "…are the radical calibration…". "…which is estimated to be…".

(2)Response to RC1: The source of uncertainty is actually the generation of radicals during the radical calibration.

(1) Comments from Referee: Page 10, line 307. "The errors associated…".

(2)Response to RC1: The text has been modified as suggested.

(1) Comments from Referee: Page 10, line 312. "…reactor 2, respectively, and…".

(2)Response to RC1: The text has been modified as suggested.

(1) Comments from Referee: Page 10, line 315-6. Delete "during the airborne measurement of RO_2*".

(2)Response to RC1: The text has been modified as suggested.

(1) Comments from Referee: Page 10, line 317. "The noise in the NO_2 signal is enhanced by…".

(2)Response to RC1: The text has been modified as suggested.

(1) Comments from Referee: Page 10, line 319. "…cabin temperature could increase…".

(2)Response to RC1: The cabin temperature varies depending on the characteristics of the flight. The sentence has been reworded.

(1) Comments from Referee: Page 10, line 320. "…stability of the CRDS signal and the accuracy of the supporting measurements."

(2)Response to RC1: The text refers to measurements taken before flying to check the overall performance of the instrument. Therefore they are rather "reference" than "supporting" measurements.

(1) Comments from Referee: Page 10, line 323-4. Again, this reviewer does not agree with this statement. Since you are continually measuring the background and the amplified signal plus background, variations should be accounted for. Only changes happening faster than one second should have influence, unless there is something about the data analysis that is not obvious from the presentation in the paper. Suggest looking into why step changes in ozone should affect the signal for more than a few seconds. Also, step changes are extreme for ambient

measurements. Even changes as the aircraft changes altitude are likely to be gradual unless a pollution layer is encountered.

(2)Response to RC1: The PeRCEAS operating conditions has been clarified in text and in answers to previous questions of RC1. These clarifications have already addressed this issue.

(1) Comments from Referee: Page 10, line 335. "…a standard deviation of the order of…".

(2)Response to RC1: The sentence has been corrected.

(1) Comments from Referee: Page 11, line 353. "…detector signals can be significantly affected…".

(2)Response to RC1: The text has been changed as suggested.

(1) Comments from Referee: Page 11, line 360. "…airborne measurements and is difficult to implement in…".

(2)Response to RC1: The text has been changed as suggested.

(1) Comments from Referee: Page 11, lines 364-370. Do you mean running the inlet at reduced pressure results in lower eCL values? Doesn't PeRCEAS continually measure the signal and background as mentioned in the second line? What do differences in detector accuracy (do you mean sensitivity) do to affect their uncertainties? Suggest adding a reference to the last sentence of this paragraph (about RO_2 interferences in LIF).

(2)Response to RC1: As now better described in section 2 the pressure in the inlet is controlled in the pre-chamber during the flight. This pre-chamber is however associated with radical wall losses which reduce the eCL as discussed in 3.2.1. The losses in the pre-chamber depend on the residence time in the pre-chamber. The latter depends on the volume of the pre-chamber and the total flow rates.

The sentence has been extended for clarification. Lines 469-470:

(3) Author's changes in manuscript: "The pressure regulation in PeRCA based airborne instruments results in lower eCL than ground based ones. This is attributed to radical losses in the pre-chamber prior to the addition of reagent gases for the radical chemical amplification."

(2)Response to RC1: The reference Fuchs et al. (2011) is already cited in the introduction. However, it has additionally been included here as suggested by the referee.

(1) Comments from Referee: Page 12, line 374. "Figure 20 shows sample data of RO_2* measured…".

(2)Response to RC1: The text has been changed as suggested. The figure 20 is now Figure 18.

(1) Comments from Referee: Page 12, line 382. Mention that the flagged values are shown in Figures 20 and 23. Also mention this in the figure captions.

(2)Response to RC1: The flags are now mentioned in the text and in the corresponding figure captions (now figures 18 and 19) as suggested by the referee.

(1) Comments from Referee: Page 12, line 386. "…in more detail in Figure 23…".

(2)Response to RC1: The text has been modified (Lines 483-487). The corresponding figure is now Figure 19.

(1) Comments from Referee: Page 12, line 388. Not sure what is meant by "the signal is not affected by altitude changes", since there are jumps in ΔNO_2 when the altitude changes. Suggest rewording to make the point clearer.

(2)Response to RC1: Although ΔNO_2 changes by altitude changes the value of RO_2* does not change significantly. The text has been accordingly rewritten. Lines 489-492:

(3) Author's changes in manuscript: "As can be seen in the figure, pressure fluctuations due to dynamic pressure changes have been reduced by up to 80 % in the improved PeRCEAS. Although the measured ΔNO_2 is affected by altitude changes, the value of the retrieved RO_2^* does not change significantly except for the maximum climbing rate directly after take-off.".

(1) Comments from Referee: Page 12, line 402. "…over a 60 s integration…".

(2)Response to RC1: The sentence has been corrected.

(1) Comments from Referee: Page 12, line 404. While PeRCEAS may be suitable for measurements up to 12 km, no data were shown at this altitude. Suggest rewording this sentence.

(2)Response to RC1: The sentence has been reworded. Line 506:

(3) Author's changes in manuscript: "The performance of the PeRCEAS instrument has been proven to be suitable for airborne measurements during different campaigns onboard HALO."

(1) Comments from Referee: Page 12, line 405. "…campaigns onboard HALO.".

(2) Response to RC1: The sentence has been changed as suggested.

(1) Comments from Referee:

References

- Line 449. "…peroxy radicals by chemical amplification…".
- Line 452. Two references are together. Need carriage return after "1993".

(2)Response to RC1: The references have been corrected.

Tables

(1) Comments from Referee: Suggest heading for "second addition point" to be changed to "reaction times", and "to detector" changed to "transfer times".

(2)Response to RC1: The headings have been changed to "reactor residence time" and "total residence time".

(1) Comments from Referee: Page 3, Edwards et al., Inlet pressure should be 200 mB.

(2)Response to RC1: This has been corrected.

Figures

(1) Comments from Referee: Most of the figures need larger symbols and bolder lines (4, 5, 6, 10, 12, 14, 17, 18, 19, 20, 23). In many of the plots, the legend is covered by data. Suggest enclosing legend in a box with a white background.

(2)Response to RC1: This has been corrected.

(1) Comments from Referee: See comments earlier about Figures 1-3.

(2)Response to RC1: These figures have been removed and replaced as suggested.

(1) Comments from Referee: Figure 2 caption. "…Top view of the…". "..the laser beam is highlighted (purple) for…". "…exiting the cavity is depicted."

(2)Response to RC1: This figure has been removed and replaced as suggested.

(1) Comments from Referee: Figure 4. Change y-axis to Allan Variance. Describe what the lines depict (linear fits to data less than 10 seconds?).

(2)Response to RC1: This figure has been removed and replaced as suggested. A subplot has been added to show the detector stability for 40 minutes measurement by detector FH as an example. These data are used for the Allan variance study of the corresponding detector.

(1) Comments from Referee: Figure 5. It is not obvious what this figure is trying to show. It appears to this reviewer that the point is temperature changes affect the τ_0 of the detector, but the retrieved ΔNO_2 is affected very little. Why not do this experiment with two detectors as done for radical measurements? This would be a more realistic representation of the actual measurement situation.

(2)Response to RC1: The objective of this experiment is to illustrate the effect of temperature drifts in the ΔNO_2 at the detector. Since both NO_2 detectors are identical in the operating conditions, it is sufficient to show the results obtained with one detector. The text has been extended for clarification

(1) Comments from Referee: Figure 6. Are the equations determined from linear fits? Are they standard or bivariate fits?

(2)Response to RC1: The equations are determined from linear and bivariate fits, considering the errors in x and y axis. This plot is now placed in the supplementary information.

(1) Comments from Referee: Figure 7. Why are not data shown for lower values of NO? Suggest more work going from 0 to 3.5E14 NO with at least 10 points for each instrumental condition (pressure and radical type). Perhaps also show the same y-axis for both plots.

(2)Response to RC1: The data series have been extended as suggested.

(1) Comments from Referee: Figure 8. Since it is mentioned in the text, suggest adding 1:1.5 line. As discussed earlier, perhaps more modeling with more realistic wall loss rates needs to be done.

(2)Response to RC1: The text has been extended and the results of additional modeling have been depicted in a new figure.

(1) Comments from Referee: Figure 9. Changes to this figure suggested earlier (plot O_3 lifetime versus NO). If it is kept the same, suggest labeling each sub-figure and referring to those labels in the caption.

(2)Response to RC1: The figure has been changed as suggested.

(1) Comments from Referee: Figure 10. This shows that 60% of the O_3 is converted with NO of 3 ppmv, and 90% at 6 ppmv. This means that the instrument could be run with much lower NO levels.

(2)Response to RC1: This issue has been extensively discussed in previous answers.

(1) Comments from Referee: Figure 11. It is stated that PAN interference is not a problem with PeRCEAS, but this plot shows that with reaction times of 3 seconds (compared to 2.6 to 3.1 seconds for the two DUALER inlets), up to 6 pptv of peroxy radicals can be produced from 1 ppbv of PAN. Is this representative of the conditions for which the instrument has been used? This figure could be changed to plot the fraction conversion of PAN (CH_3O_2 produced / PAN) versus temperature for the two DUALER reaction times.

(2)Response to RC1: As mentioned before, the simulations have been revised and the plots updated. The ratio CH_3O_2 produced/PAN is $<10^{-3}$ for the range of temperatures expected in the reactors, which are in the outer part of the fuselage. The maximum interference expected remains below 1-2 pptv for the transition times given in Table 2.

(1) Comments from Referee: Figure 13. There are places where the ambient water is below the inlet water. How can this be? This figure could be changed to plot inlet H_2O versus ambient H_2O with the points colored by altitude.

(2)Response to RC1: The figure has been changed as suggested by the referee. The relative humidity sensor at the inlet has been re-calibrated. After applying the calibration correction all the above mentioned values of inlet humidity lower than the ambient humidity have disappeared. The plot has been updated.

(1) Comments from Referee: Figure 14. How can the lack of dependence on water vapor be explained, given that it is a purported to be related to one of the amplification chemistry reactions (HO_2+NO)? Perhaps modelling of these data would be instructive. Also, suggest showing data down to the lowest water values possible.

(2)Response to RC1: The dependency on water vapour only becomes significant at higher water number concentrations for NO 45 ppm at 300 mbar as shown in figure 14. New measurements at 10 and 30 ppmv NO at 300

mbar show a clear dependency with [H_2O] which has to be taken into account in the analysis of ambient air measurements at these operating conditions. The water vapour dependence of eCL decreases significantly from 10 ppm to 45 ppm. For water vapour concentrations in the range of 4 -14 x 10^{16} molecule cm^{-3} of relevance for HALO flights, this results in the water vapour impact on the eCL being negligible. This is shown more explicitly in the new figure 14 in the manuscript.

As described in Hastie et al., (1991) and Reichert et al., (2003), the chain length (CL) of a PERCA reactor can be expressed using a resistance model as follows:

$$\frac{1}{CL} = \left(\frac{1}{CL_{HO_2}} + \frac{1}{CL_{OH}}\right)$$

$$\frac{1}{CL} \approx \frac{1}{P_{NO_2}} \cdot \left(\frac{\sum HO_2 \text{ removal rates}}{\sum HO_2 \text{ propagating rates}} + \frac{\sum OH \text{ removal rates}}{\sum OH \text{ propagating rates}}\right)$$

And if only the predominant processes are considered, as:

$$\frac{1}{CL} \approx \frac{1}{P_{NO_2}} \cdot \left(\frac{k_{w_{HO_2}}}{k_{NO+HO_2}} + \frac{k_{w_{OH}} + k_{HONO} \cdot [NO]}{k_{CO+OH} \cdot [CO]}\right)$$

P_{NO_2} is the probability of radical conversion into NO_2, $k_{w_{HO_2}}$ and $k_{w_{OH}}$ are the wall losses of HO_2 and OH respectively, k_{NO+HO_2} is the rate constant for NO_2 production from HO_2 + NO reaction, k_{HONO} is the rate constant for HONO formation from OH + NO reaction, k_{CO+OH} is the rate constant for HO_2 formation from OH + CO reaction. In the presence of H_2O, HO_2 water clusters are formed as postulated by Reichert et al., (2003).

Our interpretation of these novel results is that as the NO is increased from mixing ratios of 10 to 45 ppm at 300 mbar, the CL_{OH} is reduced, because the rate of the chain termination termolecular reaction of OH with NO making HONO increases, and the CL_{HO_2} is increased, because the rate of the propagation reaction between NO and HO_2 increases. As a result the CL begins to be dominated by the CL_{OH}, which is independent of water vapour.

The new series of measurements are now included in Figure 14 and the text has been extended for clarification (Lines 359-371):

(3) Author's changes in manuscript: "In this work radical mixtures were sampled at 25 °C for relative humidity between 2 % and 25 %. This leads to a ca. 20 times increase in the absolute [H_2O]. These conditions cover the [H_2O] expected for a larger T range (-20 – 30 °C) during airborne measurements in the free troposphere at 200 and 300 mbar inlet pressures. Figure 13 shows the [H_2O] in the air probed versus the [H_2O] in the inlet for real measurements on board of the HALO aircraft. The results in figure 14 for 45 ppm NO ([NO] 3.28 x 10^{14} molecules cm^{-3} at 300 mbar) indicate that variations in the sample humidity do not lead to additional uncertainty in the RO_2^* retrieval as the PeRCEAS eCL remains invariable within the experimental error up to [H_2O] ~ 1.4 x 10^{17} molecules cm^{-3}. In contrast, for 10 ppm and 30 ppm NO in the reactor ([NO] 7.29 x 10^{13} and 2.19 x 10^{14} molecules cm^{-3} at 300 mbar) the eCL shows a clear dependency with the ambient [HO_2]. The comparison with the eCL values obtained by Reichert et al (2003) at 1 atmosphere indicate a eCL dependency on [H_2O], temperature and pressure having a different pattern for 45 ppm NO in the reactor. This is explained by invoking the competition in the amplification chain length, CL, between HO_2

and OH removal rates, as explained in Hastie et al., (1991) and Reichert et al., (2003). At [NO] ~ 3.28 x 10^{14} molecules cm^{-3} the CL begins to be dominated by the rate of the termination termolecular reaction of OH with NO, which is independent of water vapour. This eCL dependency has to be taken considered in the analysis of ambient air RO_2^* measurements."

(2)Response to RC1: The [H_2O] values showed are the lowest water values experimentally possible with the used set up.

(1) Comments from Referees: Figure 15. Can the laser emission be adjusted so all three detectors peak at the same wavelength? This would definitely help them to behave more similarly. Suggest changing the right-hand y-axis to go from 0 to10, and to average the cross-section data to a lower resolution, say 0.1 nm, to make the plot message clearer.

(2)Response to RC1: The laser emissions cannot be changed. They are multimode diode lasers with fixed emission at a particular measurement condition. The plot has been modified as suggested and moved to the SI as suggested by RC2.

(1) Comments from Referee: Figure 16. Not sure this figure is necessary, since the data are plotted in Figure 17.

(2)Response to RC1: This figure has been moved to the SI.

(1) Comments from Referee: Figure 19. change caption "..while changing O_3…", "…in the source with estimated 15%...". It is not clear why the perturbations to ΔNO_2 last so long (up to 40 seconds) when the background should be measured on the 1 second time scale. Is this a data processing issue? Suggest checking why this is the case.

(2)Response to RC1: The ΔNO_2 from each detector is calculated from individual ring down times. Consequently, variations in the background affect the calculated ΔNO_2. This effect is cancelled out when averaging the ΔNO_2 of both detectors to retrieve RO_2^* which consequently remains unaffected. The variation of O_3 from the calibrator is not instantaneous but takes around 1 minute to become stable. This is the reason why ca. three RO_2^* values are affected by each change in O_3.

(1) Comments from Referee: Figure 20. There are big swings in the inlet pressure even when the altitude is not being changed. Why is this?

(2)Response to RC1: The figure has been modified for clarification. The pressure spikes are related to dynamical pressure changes in the inlet, e.g. caused by changes in velocity of the aircraft and air turbulences.

(1) Comments from Referee: Figures 21 and 22. These figures could be eliminated

(2)Response to RC1: These figures have been removed as suggested by both referees.

Response to RC2

General comments:

(1) Comments from Referee: 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.

(2)Response to RC2: The first three figures have been replaced by instrument schematics as suggested by the referee.

(1) Comments from Referee: 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.

(2)Response to RC2: A new figure (Figure 2) showing the changes to the pre-chamber and reactors in the DUALER has been included as suggested by the referee. In addition the text has been rewritten (Lines 143-151):

(3) Author's changes in manuscript: "In the DUALER, a stable pressure in the pre-chamber is achieved by a pressure regulator, which controls the flow through the bypass line. As noted the flow rate through the reactors is held constant during measurements. Consequently, when the outside air pressure changes, the bypass flow rate from the pre-chamber is changed. The outer dimensions, shape, form and weight of the DUALER are constrained by the inlet pylon in use with the research aircraft HALO. After the first version of the DUALER (from now on called DUALER I) was flown, the inner dimensions of the pre-chamber were further optimised to reduce the wall losses and turbulence in the pre-chamber. For this, in the DUALER II the volume of the pre-chamber was increased by extending its vertical extent, the length of the truncated cone on top of the reactors was reduced in 3 mm, and the volume of the reactors was increased to 130.5 ml from the 112 ml in DUALER I. These changes resulted in a higher eCL and improved pressure stability in DUALER II as compared to DUALER I. Figure 2 shows the upper part of both DUALER I and DUALER II."

(1) Comments from Referee: 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).

(2)Response to RC2: A new figure (Figure 4) has been added showing the ring down times of the two detector, the retrieved ΔNO_2 and HO_2 for a laboratory calibration of HO_2. The text has been rewritten (Lines 127-142):

(3) Author's changes in manuscript: "Briefly, sampled air enters PeRCEAS through the DUALER pre-chamber, which is at a lower pressure than that outside of the HALO, through an orifice in a truncated cone, i.e. a nozzle. From this pre-chamber the air is pumped simultaneously through the two flow reactors and a bypass line. At the upper addition point a mixture of CO or N_2 and NO enters each reactor. At the lower addition point, a flow of N_2 or CO enters each reactor. This enables the CO and N_2 flows in

the two reactors within the DUALER to be switched simultaneously but out of phase with one another from the upper to the lower addition point. At the addition points, the reagent gases enter the reactor through eight circular distributed 1 mm holes to facilitate the rapid mixing with the sampled air. During measurements, the pressure in the pre-chamber and both reactors is held constant. However, there is a small pressure fluctuation during the switching of flows between the upper and lower mixing point. The flow passing through each reactor enters a CRDS NO_2 detector. Afterwards, the sample flows together with the air from the bypass line are scrubbed for CO and NO_x and, exhausted by the pump.

The DUALER inlet comprises two PeRCA chemical reactors having alternating measurement modes, which are out of phase with one another. During the first part of the measurement cycle, the first reactor and detector are in amplification mode, while simultaneously the second reactor and detector are in background mode. In the second part of the cycle, the CO addition point in both reactors is switched. Consequently, the first reactor and detector are then in background mode while the second reactor and detector are in amplification mode. In the analysis of the measurements, the amplification and background signals from both detectors are combined appropriately. This improves accuracy and temporal resolution of the resultant RO_2^* data set (see 3.1). "

(1) Comments from Referee: Furthermore, a detailed description of how the mixing ratio of 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).

(2)Response to RC2: The objective of the present publication is to explain the dominant factors affecting the overall performance and accuracy of PeRCEAS for the determination of the hydroperoxyl, HO_2, and organic peroxy radicals, RO_2, which react with NO to form NO_2, when deployed on the the HALO aircraft. The operating conditions of PeRCEAS are optimised for the specific sampling position used, cabin location of the instruments, safety requirements and the type of flight tracks and altitude profiles which were flown by HALO. Generally, these limitations are often different in different campaigns. For this reason, this manuscript does not aim at describing a unique universal set of PeRCEAS operating conditions.

(1) Comments from Referee: 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

(2)Response to RC2: The performance of DUALER I and DUALER II are now compared in section 5. In addition some key improvements of performance of DUALER II compared with DUALER I are highlighted. Lines 488-493:

(3) Author's changes in manuscript: "Two hours of measurements from the flight on the 19.03.2018 are shown in Figure 19 as an example of the third airborne deployment of PeRCEAS within the EMeRGe campaign in Asia. As can be seen in the figure, pressure fluctuations due to dynamic pressure changes have been reduced by up to 80 % in the improved PeRCEAS. Although the measured ΔNO_2 is affected by altitude changes, the value of the retrieved RO_2^* does not change significantly except for the maximum climbing rate directly after take-off. Furthermore, the beam camera and the motorised mirror mounts enable the identification and immediate correction of small misalignments. This improves significantly the instrumental performance while simplifying maintenance."

Specific comments:

(1) Comments from Referee: 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.

(2)Response to RC2: The piezo optical alignment system was used in the previous PeRCEAS configuration using a single mode laser as reported by Horstjann et al., (2014). The actual PeRCEAS detector described here, has a multimode diode laser. The alignment is done manually using motorized mirror mounts while the beam camera confirms the TEM^{00} mode. The beam profile is checked manually in a regular basis but is not a feedback parameter for the alignment of the mirrors.

The text has been extended. Lines 153-155:

(3) Author's changes in manuscript: "The optical cavity remains similar to that described in Horstjann et al., (2014), i.e., a V-resonator of ca. 100 cm^3 volume formed between glued highly reflective mirrors (reflectivity, R = 99.995 %, diameter, d = 0.5", radius of curvature, roc = 100 cm, AT Films, USA) on the side of a Teflon coated aluminium cuboid."

(1) Comments from Referee: 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

(2)Response to RC2: The text has been extended for clarification at the beginning of 3.1. (Lines 181-190) and Figure 4 has been added:

(3) Author's changes in manuscript: "The mode time is defined as the time selected for the measurement in either amplification or background mode. The modulation time is the time taken for a complete measurement cycle, which comprises the sum of one amplification and one background mode. The PeRCEAS measurement cycle is illustrated in Figure 4. The ΔNO_2 for each detector is calculated from the ring down time of two consecutive modes using Eq.(1). If the mode time is adequately selected, the RO_2^* retrieved per measurement cycle is identical in both measurement lines, as the two reactors are operated out of phase with one another. The final RO_2^* data is calculated as the mean of the RO_2^* determined from the ΔNO_2 and eCL of both detectors for a given measurement cycle. The time resolution of the RO_2^* measurement is then equal to the mode time. After switching modes, a small pressure pulse leads to an oscillation of the NO_2 signal. Consequently, the first 20 s of each mode are not used in data analysis. The time lag arising from the time taken for the sample flow between the CRDS detector and the point of switching is typically less than 8 s (see Table 3)."

(1) Comments from Referee: Page 5, line 160, "…detector temperature. For this different detector temperature gradients, ΔT, where applied to modulated signals generated by varying the sampled NO_2 concentration…" it's not clear why the investigators modulated 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.

(2)Response to RC2: A modulated signal was applied in order to study the effect on similar signals to those measured by PeRCEAS during operation i.e., a ΔNO_2 between amplification and background modes. The text has been extended for clarification. Lines 211-221:

(3) Author's changes in manuscript: "Temperature changes of the detector affect: i) the diode laser emission, both its amplitude and wavelength, and ii) the mode matching between laser and detector, and consequently the τ_0. The effect of the variations in τ, resulting from changes in room or HALO cabin air temperatures, on the accuracy and precision of the ΔNO_2 determination was investigated by a series of laboratory experiments. For this, modulated concentrations of NO_2 in the flow were generated. This was achieved by alternating between two selected NO_2 concentrations once per minute. The temperature of the CRDS detector, T, and τ were then measured. Detector temperature gradients over a time t, i.e., $\Delta T/\Delta t$, determined by the temperature within the detector housing close to the photodiode, were induced by controlled changes in the room temperature.

Figure 6 shows the effect of introducing temperature perturbations in a modulated NO_2 signal between 11.5 and 12.1 ppbv measured at 200 mbar and 23 °C. As can be seen in the figure, a temperature perturbation affects both precision and accuracy of the retrieved ΔNO_2. For temperature gradients up to $\Delta T/\Delta t \approx 7$ °C h^{-1} the experimental precision of the ΔNO_2 determination remains within (2σ) 150 pptv (= 7.3 x 10^8 molecules cm^{-3} at 200 mbar and 23°C). "

Concerning T stability as states in 4.2, Lines 406-407:

"Generally, in-flight variations in the HALO cabin temperature affect minimally the accuracy of the RO_2^* determination. "

(1) Comments from Referee: Page 6, line187, "…of the sampled O_3 by NO to form NO_2 also depends on the concentration of 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 NO was decided on for a reagent mixing ratio and discuss flow rate choices for both NO and CO.

(2)Response to RC2: The purpose of this section is to present the effect of changing gas concentration and flows. A balance between different and competing effects, leads to a selection of the optimal operating conditions for a specific measurement campaign. 30 ppm NO has just been selected for some of the testing series in this work as an example for suitable reagent mixing ratio. The paper has been revised and additional measurement series have been included to illustrate some of the effects investigated.

(1) Comments from Referee: 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.

(2)Response to RC2: This part of the text has been modified taking into account the comments of both RC1 and RC2.

(1) Comments from Referee: Page 7, line 216, "The model was initialized with 9% CO, 3 ppb O_3, 50 pptv HO_2…" why was 3 ppb 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 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.

(2)Response to RC2: The model is initialised with 3 ppbv O_3 because this is the mixing ratio produced during the calibration. Sensitivity studies have shown no significant change in the simulated eCL up to 100ppb O_3. This is also confirmed by the experimental values shown in figure 17.

Following the recommendation of RC2, the simulations have been extended varying the wall losses in the pre-chamber to match the experimental values, as now explained in the text (Lines 280-306). PeRCEAS is operated at low pressures and the pressure regulation during operation prevents the direct experimental determination of the wall losses in the pre-chamber. However these are estimated by comparison with the model used.

(1) Comments from Referee: Page 7, line 222, "figure 8 shows eCL vs CL", the authors should include error bars on these data.

(2)Response to RC2: The error bars on the experimental data are now included and the figure has been extended with more experimental and model data.

(1) Comments from Referee: Page 7, line 228, "Figure 9 depicts the 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 NO mixing ratio curve and plot all 4 conditions (ie pressure and O_3 mixing ratio) on 1 figure for varying 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.

(2)Response to RC2: The figures have been modified as suggested by the referee.

(1) Comments from Referee: 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.

(2)Response to RC2: The discrepancies between measured eCL and simulated CL for the reactor are attributed to errors in the estimate of the wall losses in the inlet pre-chamber and not to the overall performance of the box model. The chemistry involved in the formation of CH_3O_2 from the PAN decomposition has now been revised, and all the rates have been taken from the recommendations in JPL 15-10, with the equilibrium rate constant from Zhang et al (2011).

The results do not change for NO mixing ratios in reactor within 10 and 45 ppm.

During the flight the PeRCEAS reactors are located inside the pylon on the upper part of the HALO fuselage. As a consequence, the temperature in the reactors, which is measured, remains below 290K (Figure 20 and 21). Consequently the maximum interference expected for the transition times given in Table 3 will be below 2 pptv, which is within the measurement error. The figure (now Figure 11) has been updated and the text has been extended for clarification (Lines 324-339).

(1) Comments from Referee: Page 8, line 252, "Figure 12 shows the variation of the eCL for 45 ppm NO within a pressure range…", The reviewer does not understand why this experiment was done with 45 ppm NO when the decided upon mixing ratio of NO addition seems to be 30 ppm 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.

(2)Response to RC2: The figure has been extended with similar measurements for 10 ppm NO. The results at 10 ppm NO confirm the recommendation of ΔP= 100 mbar as the minimum operating pressure. The text (Lines 348-353) and the figure (now Figure 12) have been updated.

(1) Comments from Referee: Page 9, line 284, "Figure 15 depicts exemplary a comparison of spectra…", the reviewer does not believe including NO_2 absorption cross section and detector spectra is a useful figure for the main text of this paper. Remove or include in the SI.

(2)Response to RC2: The figure has now been included in the supplementary information. Line 387:

(3) Author's changes in manuscript: "A sample comparison of spectra obtained for the three PeRCEAS detectors is included in the supplementary information (Figure SI-1)."

(1) Comments from Referee: Page 9, line 289, "In addition, the effective σ_{NO2} 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.

(2)Response to RC2: The figure has now been included in the supplementary information. Lines 392-396:

(3) Author's changes in manuscript: "The result of applying Eq. (5) to the PeRCEAS detectors is depicted in Figure 15. The detectors sampled known mixtures of NO_2 from commercial gas cylinders in synthetic air at 200 mbar as shown in the supplementary information (Figure SI-2)."

(1) Comments from Referee: 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_2 number density [molecules/cm^3] vs. α, as the slope has the physical meaning of the absorption cross section of NO_2.

(2)Response to RC2: In the case of CRDS α is not measured but must be calculated by using Eq.1. In contrast, the variables plotted in the Figure 15 (originally Figure 17) are taken directly from the measurement and do not require a prior knowledge of τ_0. In addition the y intercept of the plot is the $1/\tau_0$ for each detector.

(1) Comments from Referee: 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?

(2)Response to RC2: Temperature drifts can be minimised during laboratory measurements. Concerning in-flight measurements, it is mentioned in section 4.3 that the temperature in the HALO cabin remains reasonably constant and therefore is not a significant source of uncertainty during the airborne measurement. An example can be seen in Figures 18 and 19. A sentence has been added to clarify this point. Line 406:

(3) Author's changes in manuscript: "Generally, in-flight variations in the HALO cabin temperature affect minimally the accuracy of the RO_2^* determination."

(1) Comments from Referee: 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?

(2)Response to RC2: In this publication there is no intention to define one unique set of operating conditions for the PeRCEAS on HALO. Several concentrations and mixing ratios for NO were investigated. At 300 mbar the longest series of measurements was selected in the figure 18 to show the best statistics in the reproducibility and stability of eCL over time. These measurements were carried out to investigate the dependence of eCL on NO. Figure 18 (now Figure 16 in the revised version) is been replaced to show the data obtained at 300 mbar and 30 ppmv as suggested by the referee. The estimates of the corresponding eCL values for 10 ppm, 30 ppm and 45 ppm NO are now listed in Table 4.

(1) Comments from Referee: Page 10, line 330, "As can be seen in Figure 19…", it would be useful to plot actual O_3 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_3 mixing ratio is changed the NO_2 signal displays a large amount of noise (50 ppb). It is also unclear what D1, D2, SG and BG stand for in this figure.

(2)Response to RC2: The original figure 19 is now Figure 17. The O_3 mixing ratio is set at the calibrated O_3 generator and cannot be measured during the PeRCEAS measurement, as it is converted in NO_2.

The variation observed in the ΔNO_2 (the NO_2 signal is not plotted) is not related to any change in the pressure but to the change in the O_3 concentration in the sample, which takes approximately 1 minute to stabilise after changing the set point in the ozone generator.

The meaning of D1, D2, AP and BG are now explained in the figure caption. SG has been replaced by AP for clarification.

(1) Comments from Referee: 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.

(2)Response to RC2: As now described in section 2 the pressure in the inlet in PeRCEAS is controlled in the pre-chamber during the flight. This pre-chamber prior to the reactors causes radical losses which do not have the ground based instruments without pressure chamber and pressure regulation. The sentence has been extended for clarification. Lines 469-470:

(3) Author's changes in manuscript: "The pressure regulation in PeRCA based airborne instruments results in lower eCL than ground based ones. This is attributed to radical losses in the pre-chamber prior to the addition of reagent gases for the radical chemical amplification."

(1) Comments from Referee: Page 11, line 367, "the detection limit and uncertainty of PeRCA based instruments are strongly depending on the variation of O_3 and NO_2 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_3 and NO_2 are measured (as often are in aircraft field campaigns) can a correction method not be proposed and evaluated?

(2)Response to RC2: The effect of O_3 changes in the RO_2^* determination has been shown and quantified in figure 17. Simultaneous measurements of O_3 and NO_2 onboard can be used as a reference for the correction of the background variations in individual cases, but a method that relies on other measurements to correct the background will have to deal with different instrumental resolutions and sources of errors of the instruments involved.

(1) Comments from Referee: 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 ΔNO_2 earlier in the manuscript. If NO_2 and O_3 mixing ratio data is available from the flight, this would be useful to include as well.

(2)Response to RC2: The figure 20 (now figure 18) has been modified to include the inlet and detector temperatures as suggested by the referee.

(1) Comments from Referee: 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.

(2)Response to RC2: This figure has been removed as suggested by the referee.

(1) Comments from Referee: 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.

(2)Response to RC2: The figures 18 and 19 have been modified to make clearer the improvement made as suggested by the referee.