Referee 2:

The measurement of NOx in remote air is very challenging, in particular because of the difficulty of accurately determining the NO2 artefact of photolytic convertor-CLD (P-CL) measurements, the current gold standard technique for accurate NOx measurements.

This manuscript, while not entirely novel as the authors point out in terms of presenting an alternative quartz glass converter for P-CL measurement of NO2, is very useful especially because of the discussion of laboratory experiments to investigate the instrumental background produced by the photolytic converter in the NOc channel and the characterisation of an improved convertor.

I recommend publication after the following points have been addressed:

We would like to thank the referee for the positive feedback and the recommendation for publication.

Pg 5. A NO_2-> NO conversion efficiency of 14% (or even 20% for the original convertor) is very low (i.e. Andersen et al. 2020 report CEs of >50%). I suggest the authors mention that a higher CE is desirable for improved accuracy and perhaps suggest ways this could be implemented.

We agree with the referee that a higher CE would be desirable. This could easily be implemented when operating the converter at higher pressures. However, for aircraft measurements, an additional uncertainty would result from calculating altitude-dependent CEs. Additionally, operation at higher pressure also increases the fractional dissociation of thermally unstable NOx reservoir species which decay in the converter which is shown in Figure S1a of the Supplement. The referee made a good point here and this is a topic which we want to investigate in more detail in the future. We have added some text for clarification.

Lines 152 ff.: Please note that the low conversion efficiencies in both converters result from the operation at low pressures which we have implemented to pursue aircraft measurements where altitude changes are accompanied by pressure variations. Operating the converter at lower than minimum ambient pressure levels (max. ~15km flight altitude) has the benefit of a constant conversion efficiency. The fractional dissociation of thermally unstable NOx reservoir species increases with increasing pressure in the converter which can be seen in Figure S1a of the Supplement. On the other hand, a higher conversion efficiency would be desirable for improved accuracy of the measurement.

Pg 5. "Therefore, a pre-chamber measurement is operated for 20 seconds every 5 minutes where ozone is added to the sample gas flow". What is the efficiency of the pre-chamber volume (i.e. how much of the added NO from the calibration gas reacts with O3) ? It should be >98% or so.

For the NO channel, the efficiency of the pre-chamber is >96% and for the NOc channel it is close to 100%. We have added this information to the main text.

Lines 166 ff: The residence time in the pre-chamber allows for the reaction of O3 and NO and the relaxation of NO2* before entering the main reaction chamber (pre-chamber efficiency > 96% for the NO channel and ~ 100% for the NOc channel)

Pg 5 Ln 141. The "constant temperature of 25oC" in the convertors is not monitored, and so could presumably be a lot higher when the LED lights are on. The authors rightly point out that accurate determination of this temperature is critical for the calculations of the NO_2 artefact. It would also be highly beneficial to perform measurements of e.g. PAN degradation to confirm the artifact calculations (and, indirectly, indicate the temperature in the chamber).

The "constant temperature of 25°C" in this sentence refers to the reaction chambers where the reaction of NO and O3 takes place, not the converter where NO2 is converted to NO. The temperature in the reaction chambers is constantly monitored and has a maximum variation of ± 0.1°C.

The temperature in the photolytic converter was not monitored during CAFE Africa. However, the temperature of the gas outflow was measured to be 40°C which we assume to be identical to the inner temperature for our calculations. The temperature in the alternative quartz converter is likely lower as the sample gas does not get in direct contact with the LEDs.

Based on the temperature of the conventional converter and the short residence time we calculate that only 0.1 % of any PAN would thermally decompose to NO2. Therefore, a PAN mixing ratio of 200 pptv would result in release of only 0.2 pptv NO2. This is consistent with findings by Reed et al. (2016).

Lines 159 ff.: Additionally, the sample gas flow in the type 2 quartz converter does not have contact with the LEDs which likely minimizes the sample gas heating and consequently the thermal interferences when passing through the converter.

Lines 199 f.: Please note that it was not possible to measure the temperature inside the converter. Instead, the temperature of the gas outflow from the converter in the ring channel was measured which we equate to the inner temperature.

Ln 178. "Please note that the instrumental background for the NO data was determined by nighttime measurements of NO instead of zero air measurements ..." How often was night-time NO determined and what was the variability?

The instrumental background for the NO data was determined from one night-time flight on August 26, 2018 and was 5.0 ± 5.3 pptv (1s integration time). In comparison, zero-air measurements throughout the campaign were on average 4 ± 7 pptv which is in close agreement. We have added text for clarification.

Lines 207 ff.: Please note that the instrumental background for the NO data was determined at 5.0 ± 5.3 pptv during a nighttime measurement on August 26, 2018 of NO as presented by Tadic et al. (2021) and previously described by Lee et al. (2009). The instrumental background determined via zero-air measurement was similar with 4 ± 7pptv (Tadic, 2021) (measured four to six times per MF).

Page 7. An calculation of uncertainty for both NO and NO2 measurements is missing from the Experimental section.

Thank you for noting this. We have determined the precision from the reproducibility of the NO calibrations and the detection limit from the reproducibility of the zero air measurements. This gives a precision of 3% (1σ). The accuracy of the secondary NO standard is 4%. For the NO channel and the NOc channel using the alternative quartz

converter, the detection limit is around 5pptv. When using the old converter, the detection limit in the NOc channel is more difficult to determine due to the observed memory effects and is estimated at >10pptv. We have added the information to the manuscript.

Lines 180 ff.: The precision is determined from the reproducibility of the NO calibrations and is 3% (1σ). The NO concentration is 4.96±0.21ppmv which gives a 4% uncertainty of the used secondary standard. The resulting NO calibration mixing ratio is 15.8±0.7ppbv. The detection limit is given by the reproducibility of the zero air measurements which is around 5pptv for the NO channel and the NOc channel using the type 2 quartz converter. The detection limit is higher when using the type 1 converter, but difficult to determine due to the observed memory effects and estimated at >10pptv.

Ln 193 "Please note that these data (OH and HO2) are still preliminary" Are final data yet available? This would be highly desirable since HO_2 and OH are required for the calculation of [PNA], and CH_3O_2 is calculated from HO_2 and required to calculate MPN.

We regret that final data are still not available. The high uncertainty of the data is based on difficulties associated with the calibration method for the data. We do not expect any changes to be significant.

Figure 5. Please include all data in the figure legend (including BG) and explain the orange dotted lines in the caption. The word "exemplarily" is not needed in the caption.

For a better overview of the figure we would prefer to limit the legend to the entries which are necessary for distinguishing the data. This is only relevant for the NO2 data traces while the other traces are specified by the according y axis label. BG is the abbreviation for background and the orange dashed lines indicate the simultaneous occurrence of an aircraft descent, an increase in water vapor concentrations and a peak in the NO2 CLD data. We have added text in the caption for clarification and removed the word "exemplarily".

Figure 5: Temporal development of the instrumental background (BG), NO, water vapor, and calculated and measured NO2 for measurement flights 10 and 12. The orange dashed lines indicate the simultaneous occurrence of a rapid decrease in altitude, an increase in water vapor concentration and a peak in the NO2 CLD data.

Lns 352 onwards. The authors demonstrate convincingly that memory effects of the porous convertor coupled to water vapour changes are a strong driver of changes in the instrumental NOc background. However, the adsorbing/desorbing of NO molecules will likely also be affected by pressure as well. Could the authors comment on this?

The memory effect is related to processes which occur in the converter which is held at a constant pressure at all times and the observed processes can therefore be considered independently of pressure. Generally, variations in pressure will affect competitive adsorption of NO and other air molecules and the transport of trace gases to the surface.

Ln 400 onwards. I congratulate the authors on their much improved photolytic convertor and its apparent stability and insensitivity to varying humidity and lack of memory effects. I would recommend also that experiments are conducted with varying pressure to evaluate pressure-dependence of the background.

We thank the reviewer for this friendly comment and the suggestion. We are very interested in the role of pressure and are planning some studies on how pressure affects the conversion efficiency and whether it is more favorable to perform aircraft measurements with a constant, but low CE or a high, but variating CE.

Ln 470 onwards. In the Conclusions section, the authors could consider adding recommendations on airborne NO2 measurements by P-CL, i.e. avoiding constant altitude changes in flight, which will inevitably change the background, and ensuring sufficient background measurements at each altitude change. This would be useful for the community.

We have added this advice regarding the use of the conventional blue light converter to the conclusions section. Regarding the quartz converter, we are confident that the background is not subject to changes as a consequence of altitude changes based on our laboratory investigations. However, this still needs to be confirmed by field experiments.

Lines 518 ff.: If a conventional blue light converter is still in use, we would suggest to avoid constant altitude changes in aircraft applications. Instead, we highly recommend the application of an alternative photolytic converter made from quartz glass (…)