

We thank the reviewer for their time and comments, which are reproduced in *italic font* below. Our responses are shown in regular font. Text added to the manuscript is underlined.

*Anonymous referee #2*

*The manuscript describes a new instrument for the detection of HONO using cavity enhanced absorption spectroscopy. The paper is well written and gives a detailed description of the detection method. It is well suited for the journal. I recommend publication after addressing the following points:*

*I am missing a more detailed discussion of potential interferences from for example HONO formation / conversion of nitrogen oxide species in the inlet system. Did the authors do any kind of tests to exclude that HONO is artificially formed in the inlet system?*

Response: We agree with the reviewer that inlet artefacts are an important consideration for any instrument when sampling ambient air. In our instrument, all wetted surfaces in the inlet system were constructed from inert FEP Teflon tubing and PFA fittings. We observed fast rise times when adding HONO to the inlet and equally fast fall times when zeroing (e.g., Figure S7) and hence do not believe that partitioning of HONO to or from the inner walls of the inlet was significant in the experiments presented here. We also did not notice any evidence for formation of HONO within the inlet system. Having said this, we agree that if the instrument is deployed for longer-term ambient air measurement, the inlet system needs to be scrutinized more. The following was added on line 336 (in the section on accuracy):

"Not included in this estimate are potential systematic errors resulting from the spectral convolution and fitting procedure (Sect 2.4), and photolysis of the fitted species within the optical cavity, and potential inlet artefacts (which were not characterized under atmospheric conditions)."

The following was added to the conclusion on line 382:

"Though not examined in this work, interferences may arise when sampling for long periods in heavily polluted environments from soot deposition on inlet filters and walls. In such situations, it would be advisable to monitor inlets for HONO production (or loss) upon aging, for example using a setup as recently described by Duan et al. (2018)."

*The potential of such interferences are also not mentioned in the introduction (page 2 line 36).*

Response: We agree that this should have been mentioned and have added the following on line 43:

"A considerable advantage of open-path instruments is the lack of any inlet and associated chemistry, such as loss of HONO due to partitioning onto inlet walls (Duan et al., 2018) or HONO formation, for example from reaction of NO<sub>2</sub> with soot particles (Longfellow et al., 1999; Kalberer et al., 1999; Indarto, 2012) that may have deposited on the inlet lines and particle filter."

*It should be mentioned that interferences are detected and corrected for in LOPAP instruments (page 2 line 33-35).*

Response: We have added the following statement: "In practice, interference from NO<sub>2</sub> and O<sub>3</sub> can be accounted for with a two-channel system and interference from PAN can be avoided by sampling at low pH (Kleffmann et al., 2006). Interference from HO<sub>2</sub>NO<sub>2</sub> is likely only significant in cold environments such as the poles since this compound is prone to thermal decomposition (Legrand et al., 2014)."

*What was the purity of the NO used for the production of NO<sub>2</sub> by ozone titration? Was there any artificial signal observed from impurities in the NO cylinder (page 7, line 180)?*

Response: Apologies for omitting this. The cylinder was supplied by Scott-Marrin and was filled with (101±1) ppmv nitric oxide in oxygen-free and moisture-free nitrogen. Scott-Marrin certified an NO<sub>2</sub> content of <0.5 ppmv. In practice, the amount of NO<sub>2</sub> emitted was larger (mainly because the oxygen-free nitrogen was combined with pure oxygen prior to delivery (Figure 1C) - if nitrogen was used as a ballast, the NO<sub>2</sub> content was negligible). On occasion, we monitored the NO<sub>2</sub> and NO<sub>x</sub> output of the gas delivery system in two parallel CRDS channels and typically found a ratio of 1.85% NO<sub>2</sub> in NO<sub>x</sub>.

Another important detail that we should have mentioned that once the setup used to deliver NO<sub>2</sub> was assembled, it always remained under flow of oxygen to keep moisture and impurities that might be present in room air (e.g., HNO<sub>3</sub> or HONO) out of the lines, i.e., to keep the tubing dry and clean.

We added the following on line 184:

"Briefly, NO<sub>2</sub> was generated by mixing the output of a standard NO cylinder (Scott-Marrin, 101±1 ppmv in oxygen- and moisture-free nitrogen)" with O<sub>3</sub> produced by illuminating a flow of O<sub>2</sub> (99.99%, Praxair) by a 254 nm Hg lamp followed by dilution with zero air to vary the product concentration. When not in use, the setup remained under O<sub>2</sub> flow to prevent moisture and other impurities from contaminating the tubing."

*What is the precision and accuracy of the conversion efficiency (the number of digits given here suggests a very high precision) (page 8 line 217)?*

Response: We agree with the reviewer that the number of digits used is optimistic. The value 83.8% was derived from a box model simulation which is precise as far as the computation is concerned, but of course relies on several assumptions, such as rapid mixing of the added ozone with the sample gas and accuracy of rate coefficients. The most important uncertainty is the rate coefficient for NO + O<sub>3</sub>, which NASA/JPL puts at ±10%.

We added the following on line 227: "The accuracy of this correction factor is limited by knowledge of the rate coefficient for the oxidation of NO by O<sub>3</sub>, ±10% (Burkholder et al., 2015)."

*Are the measurements in Fig. S4 examples for a typical measurement or can the measurements repeated with high accuracy that always the same concentrations are observed (page 8 line 221-225)?*

Response: This is an example of a typical measurement. With the HONO source used in this work, it was challenging to always produce the same concentration; however, this didn't matter in practice since scatter plots of the two measurements were ultimately generated (e.g., Figure S6b). We have modified the section of text as follows:

"Figure S4 shows a sample TD-CRDS inlet temperature scan when the output of the source described in Sec. 2.6 was sampled. In this particular example, the heated channel (to which excess O<sub>3</sub> was continuously added) measured ~137.5 ppbv of NO<sub>y</sub> (NO<sub>x</sub> + HONO) ..."

*Did the authors have a closer look, if the intercept in the regression for lab measurements holds for low NO<sub>2</sub> values (page 10 line 290)? For ambient air measurements, this would be a significant source of errors, but might be here due to the large range of values here.*

Response: We agree with the reviewer that intercept is larger than desired and is driven by the large range of values as the reviewer states. No changes were made.

*Was the GNOM instrument running in parallel also during ambient air measurements (section 3.8)? If so could the authors show the correlation of NO<sub>2</sub> with the HODOR instrument? I would prefer to see the regression analysis in the main paper.*

Response: Unfortunately, the GNOM instrument was not operated during the ambient air measurements. No changes were made.

## References

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