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 <u>underlined</u>.

Anonymous referee #3

General comments: This is a really well written manuscript on the development of an IBBCEAS instrument at the University of Calgary, operated in the near UV between361 and 388 nm for the detection of HONO and NO2. Even though the authors pay excellent attention to experimental detail and characterize the performance comprehensively, including first measurements in ambient air, the technology in this manuscript, the measurement principles, the calibration approaches and verification methods are all known and not really new. Despite the very competent experimental description of the instrument, in my opinion, the manuscript is lacking novelty as illustrated in Table 1, where 7 other publications are listed using the same or very similar broadband cavity enhanced approaches. The authors themselves recently published a very similar instrument merely in a different spectral region. The most novel aspect in this manuscript concerning the area of 'CEAS for field detection of trace gas species' lies in the comparison of IBBCEAS with TD-CRDS, which is very brief in terms of a discussion and largely kept in the supplementary material. If the work is deemed publishable by the editorial board, then material from the supplementary material (S6,S7) should be moved into the main body of the text and further discussed.

Response: We thank the reviewer for this frank assessment. Briefly, the novelty of this paper is
(a) We describe a newly constructed instrument that has not been previously described,
(b) we validate the HONO response of the IBBCEAS via a comparison to blue diode laser TD-CRDS, which as far as we know has not been previously made, and
(c) we show sample ambient air data that have not been previously published.
Regarding the suggestion by the reviewer to reorganize the manuscript. The focus of this manuscript is on the new IBBCEAS, not on TD-CRDS, and we prefer to keep the manuscript focused on the CEAS. The supplemental material is posted on the publisher's web site in any case and is discerned to the community, so there is no need to reorganize the manuscript.

No changes were made in response to this comment.

The work would gain substantial merit from an investigation of the performance of the instrument in field applications especially under various experimental conditions and atmospheric environments. The long-term performance of IBBCEAS instruments in the field under more or less harsh conditions has not been scrutinized to a high standard in the literature yet, but this was unfortunately not within the scope of this work.

Response: We agree with the reviewer that a thorough assessment of the in-field performance of this instrument (and the inlet in particular) will be the next step. The following was added to the conclusion section:

"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)."

A few observations and comments in detail:

- The light source has an emitting area of 1.4x1.4 mm^2 ...(l79)

Response: The text was modified as suggested.

- 1200 grooves mm^{-1}... (l104)

Response: We thank the reviewer for catching this typo. It has been fixed.

- (Kraus, 2003) is missing in the reference list (l156).

Response: Our apologies. For some reason, the reference management software keeps removing this citation from the bibliography. It is Kraus, S.: DOAS Intelligent System Version 3.2.3505, Institute of Environmental Physics, University of Heidelberg, available at: https://doasis.iup.uni-heidelberg.de/bugtracker/projects/doasis/ (last access: 31 August 2018), 2003. and has been added to the reference list.

More information on the DOASIS retrieval could be given here.

Response: More information (such as range fitted, allowed spectral shifting, order of polynomial used in the fit, etc.) was given further down in the paragraph. We moved the sentence "Gas concentrations were extracted from a linear least square fit applied to the calculated absorption coefficient, followed by conversion to mixing ratios using the number density of air calculated from the ideal gas law and the temperature and pressure of the sampled gas, monitored using a K-type thermocouple (Omega) attached to the sample cell holder and a pressure transducer (MKS Baratron 722B) located next to where gases exit the sample cell and upstream of the mass flow controller. " to the end of the paragraph to make this clearer.

- third-order ... (l162)

Response: The text was modified as suggested.

- The inlet was guided through a partially open window. How far from the outside surface of the wall or window was the inlet line? How long was it? Was the instrument facing N,W,S, or E?

Response: The instrument sampled from a 1.8 m long FEP Teflon inlet at a flow rate of 2 slpm, with approximately 1/3 of the inlet line (~2 feet) protruding from the window. The window and inlet faced SE. We modified a sentence on line 247 from "The instrument's inlet was guided through a partially open window." to "The instrument sampled from a 1.8 m long FEP Teflon™ inlet at a flow rate of 2 slpm, of which ~1/3 was guided through a partially open window."

What can the authors say about losses in the inlet line. (p8 bottom and also p12)

Response Reviewer #2 also inquired about this issue. In short, 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, plus we observed fast rise times when adding HONO to the inlet and equally fast fall times when zeroing (e.g., Figure S7). We 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 to the introduction 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_2 with soot particles (Longfellow et al., 1999; Kalberer et al., 1999; Indarto, 2012) that may have deposited on the inlet lines and particle filter."

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)."

- In addition to the opening paragraph, there is also merit in the Rayleigh scattering cross-section measurements, as they confirm measurements in the literature from some time ago.

Response: We agree.

No changes were made.

- Cleanliness: : : (l268)

Response: Typo has been corrected. Thanks for catching this.

- The fact that the background in Fig. 4 is quite substantial and negative is not discussed in the manuscript. (p10)

Response: Fig. 4 has been replaced. Please also see our response to reviewer #1.

- The authors explain that the common approach in the literature to determine the LOD is not following the more strict recommendation of IUPAC, however, then they do not follow the recommendation either, as far as I can see. (p11)

Response: This is correct. The argument by Loock and Wentzell is based on the notion that the detection limit should be determined by determining the standard deviation of repeated measurements of at least one concentration near the LOD, rather than via repeated measurements of a blank as it is commonly done. The experimental challenge, of course, is to make sure that a stable, low concentration is indeed delivered.

No changes were made.

- The authors list a set of errors limiting the accuracy of their measurements and classify them as random. The literature cross-sections for the retrieval is a systematic error. The mirror reflectivity and RL are also systematic for a given set of measurements, until they are measured again. (p11)

Response: This is correct. What we had meant to say that these errors are independent from each other. We have removed the phrase "and random" and clarified that we added these uncertainties in quadrature (following how errors were propagated by (Min et al., 2016)).

- higher flow rate... (l330)

Response: The text was modified as suggested.

- pptv (typo l356)

Response: Fixed.

- The LED does not seem to emit between 330 and 400 nm as stated in the caption (l558)

Response: Indeed, the cavity output below ~350 nm is negligible. The superficial phrase "broadband (330 - 400 nm)" was removed.

- The effective pathlength... (l560)

Response: The text was modified as suggested.

- In Figure 5 the "blue time" and "grey time" are explained, the "white times" are unclear.

Response: Reviewer #1 also pointed this out - the caption has been changed as follows:

Original: "Figure 5 Time series of NO_2 and HONO mixing ratios for synthetic and laboratory air, averaged to 1 min. **a)** ...."

Revised: "Figure 5 Time series of NO_2 and HONO mixing ratios observed by HODOR, CRDS and TD-CRDS, averaged to 1 min. The instruments sampled for zero air (grey underlay), laboratory air (blue underlay) and laboratory air to which varying amounts of synthetic air containing NO_2, HONO and zero air were added (white underlay). **a)** ...."

(p23) - Specify the term "+/-1σ measurement uncertainty" in the caption further or include a cross-reference. (l575)

Response: We have removed the "±1σ" from the caption of Figure 5.

- ...sample ambient air data... improve phrase, caption Figure 7 (l586)

Response: The first phrase of the caption of Figure 7 was changed from "Time series of sample ambient air data averaged to 5 min." to "Sample ambient air data."

References

Duan, J., Qin, M., Ouyang, B., Fang, W., Li, X., Lu, K., Tang, K., Liang, S., Meng, F., Hu, Z., Xie, P., Liu, W., and Häsler, R.: Development of an incoherent broadband cavity-enhanced absorption spectrometer for in situ measurements of HONO and NO_2, Atmos. Meas. Tech., 11, 4531-4543, 10.5194/amt-11-4531-2018, 2018.

Min, K. E., Washenfelder, R. A., Dubé, W. P., Langford, A. O., Edwards, P. M., Zarzana, K. J., Stutz, J., Lu, K., Rohrer, F., Zhang, Y., and Brown, S. S.: A broadband cavity enhanced absorption spectrometer for aircraft measurements of glyoxal, methylglyoxal, nitrous acid, nitrogen dioxide, and water vapor, Atmos. Meas. Tech., 9, 423-440, 10.5194/amt-9-423-2016, 2016.