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 #1

General comment: This paper presented a newly developed IBBCEAS system for measuring ambient HONO and NO2. The subject is within the scope of the journal, but the IBBCEAS techniques is not a new technique for measuring HONO and NO2. There are several papers with the same topic had been published in AMT recently (like Min et al., 2016; Duan et al., 2018). The authors should make it clear that what is new in this work.

Response: We appreciate the reviewer's point of view. The novelty of this paper is threefold:
(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.
Since novelty is a given (i.e., can be taken for granted) in any scientific manuscript and in our opinion the novelty of this work is transparent, we have chosen not to amend the manuscript in response to this comment.

The authors also should be more conservative, especially when using the statement like "state-of-the-art". Compared with the previous instruments mentioned before, this instrument (HODOR) does not have the best detection capacity in fact. The following comments should be addressed.

Response: Our apologies - we used the phrase "state-of-the-art" with a common dictionary definition of "the most recent stage in the development of a product, incorporating the newest ideas and the most up-to-date features" but realize that it could also be interpreted as "the most recent and therefore considered the best; up-to-the-minute". In response to the reviewer's comment, we have removed the adjective "state-of-the-art" from the abstract and added the underlined words to the sentence on line 355 as follows:

"The 60 s HODOR LOD was 240 pptv and of similar magnitude as the LODs of 180 pptv reported by Duan et al. (2018) and of 200 pppv Nakashima and Sadanaga (2017) and hence <u>on par with</u> state-of-the-art <u>instruments</u>."

Specific comments:

1. The fitting results showed in Fig. 4 have some problem, especially the fitting range from 365 to 370 nm. It seems that the measured HONO absorption coefficient (orange line) is quite similar to the unconvoluted cross section from Stutz et al., 2000 (may be the convolution is not so good). The author should give some explanations of the large residual.

Response: We agree with the reviewer that the residual ought to be void of structure. As stated in the text (lines 158-163), we convolved the reference spectra using the observed width of a Ne line

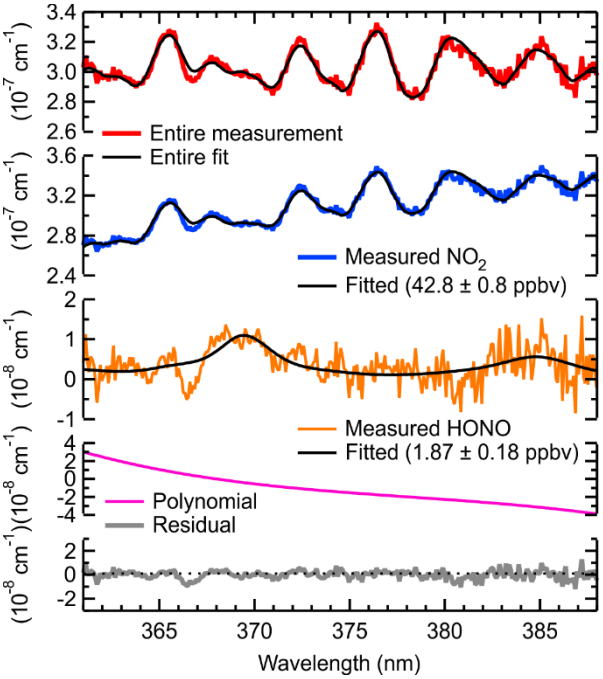
(1.04 nm). It certainly appears that the convolution between 365 and 370 nm may have been too aggressive, perhaps. The largest feature in the residual spectrum lines up with a trough in the NO_2 spectrum, whereas the second largest feature in the residual spectrum is located at the HONO peak (but is narrower). Outside this wavelength region, the agreement between observed and fitted spectra seems better. We cannot offer an explanation as to why that occurs but agree with the reviewer that a likely culprit is the instrument function used.

We acknowledged this issue in the conclusion section (see statement on lines 370-375 reproduced below). During the review phase, we were also made aware of a paper by Kleffman et al. (2006) who noted that some literature NO_2 absorption cross-sections contain HONO as an impurity.

"One of the challenges we encountered in the accurate retrieval of NO_2 and HONO was the convolution procedure and choice of cross-section. Literature values for NO_2 vary by up to ±6.2% (Harder et al., 1997; Burrows et al., 1998; Vandaele et al., 1998), such that the choice may introduce a systematic bias. In addition, some NO_2 reference spectra have been reported to contain HONO as an impurity (Kleffmann et al., 2006). Though not performed in this work, it may be advisable to use one's own reference spectra in future IBBCEAS NO_2 and HONO retrievals."

I also suggest the authors show the spectral fitting results from ambient measurement or the lab result of relatively low NO2 and HONO, which would be more representative.

Response: We have prepared such a figure as requested (see below). This particular spectrum was acquired on April 27, 2018, at 1:00 MST. Mixing ratios of 42.8 ± 0.8 ppbv of NO_2 and 1.87 ± 0.18 ppbv of HONO were retrieved.



We have updated the text in section 3.4:

Original: "Figure 4 shows an example fit containing NO_2 and HONO from a sample generated using the HONO generation system described in Sec. 2.6. The top panel shows the entire absorption (and the fit shown in black) along with the scattering coefficient of air. In this example, NO_2 (shown in blue) and HONO (shown in orange) mixing ratios of 109±0.2 ppbv and 23.9±0.4 ppbv were obtained, respectively."

Revised: "Figure 4 shows an example fit containing NO_2 and HONO in ambient air, collected on April 27, 2018, at 01:00 MST. The top panel shows the entire absorption (and the fit shown in black) along with the scattering coefficient of air. In this example, NO_2 (shown in blue) and HONO (shown in orange) mixing ratios of 42.8±0.2 ppbv and 1.9±0.2 ppbv were obtained, respectively."

We also updated the caption of Figure 4.

Original: "**Figure 4** Sample fit for laboratory generated NO_2 and HONO samples at 879.9 hPa and 296 K. The top panel shows the entire absorption spectrum. Shown below are the absorption spectra of NO_2 and HONO with their respective fit errors and the polynomial. The bottom panel shows the fit residual."

Revised: "**Figure 4** Sample fit of ambient air containing NO_2 and HONO sampled on April 27, 2018, at 01:00 MST. The top panel shows the entire absorption spectrum. Shown below are the absorption spectra of NO_2 and HONO with their respective fit errors and the polynomial. The bottom panel shows the fit residual."

2. The insert plot of Fig. 5b showed that the zero only had one point in each cycle. The author should check the transient time of mode changing from sampling mode to zero mode to make sure the zero mode without HONO.

Response: Data were acquired at 10 s, and fit results of the 10 s data were averaged to 60 s (which is the time resolution shown in the Figure). The transient time was checked. Figure S7 in the supplemental material section shows this at 1 s time resolution.

No changes were made to the manuscript.

3. Figure 5. The blue region represents lab sample and grey region represents zero, so what is the white region mean (Line 284, text indicated the indoor ambient air)?

The regions that are not color-coded (and hence appear white) are a combination of synthetic air or lab air to which various amounts of synthetic air containing HONO, NO_2 and zero air were added. We have modified the figure caption 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)** ...."

Why TD-CRDS only has such short measurement time period?

Response: The TD-CRDS HONO measurement is only valid when HONO is fully dissociated to NO (which is then oxidized in excess O_3 to NO_2). For the time period 23:30 - 00:10 of Figure 5, the TD-CRDS inlet converter temperature was ramped up and down several times to acquire thermograms (i.e., plots of signal vs. inlet temperature; an example is shown in Figure S4). The handful of data points shown in Figure 5b were collected when inlet temperatures were > 520 °C. After 00:10 of Figure 5, the TD-CRDS inlet temperature was constant (average $\pm$ 1 standard deviation of 523 $\pm$ 3 °C).

When sampling laboratory air the TD-CRDS does not have the necessary sensitivity to detect let alone quantify HONO since there is a large NO_x background that needs to be subtracted, as stated on line 287:

"In contrast to the IBBCEAS instrument, the TD-CRDS instrument was unable to quantify HONO in indoor air since the high NO_2 background introduces a large subtraction error in the heated channel."

In response to the reviewer's comment, we added the following to the caption of Figure 5:
"**b)** HONO mixing ratios reported by TD-CRDS (black) and IBBCEAS (orange). From 23:30 to 00:10, the TD-CRDS inlet converter temperature was ramped up and down several times to collect thermogram; only data collected at an inlet temperature >520 °C are shown here."

As the authors mentioned that GNOM suffered with interference of high NO2. I do not think the inter-comparison of HODOR with GNOM is appropriate to prove the measurement capacity of HODOR in measuring HONO.

Response: We, respectfully, disagree with the reviewer as TD-CRDS is quite accurate indeed when sampling a dilute mixture of HONO in zero air, i.e., in the absence of a large NO_x background. This is exemplified by the high degree of correlation as stated on lines 290-:

"The scatter plot of IBBCEAS vs. TD-CRDS HONO data (Fig. S6b; only data points when the synthetic source was sampled were included in the fit) has a slope of 1.01±0.01, an intercept of 0.01±0.24 ppbv and r^2 of 0.995. "

No changes were made to the manuscript in response to this comment.

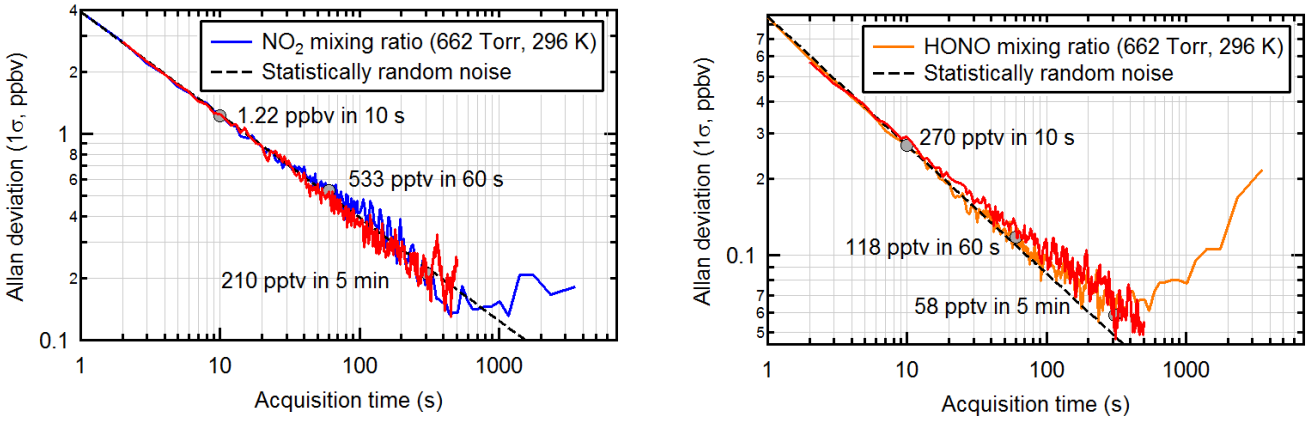
4. Allen deviation only used to study the system stability, the instrumental limit of detection should be characterized by the standard deviation. Figure 6 showed that the zero measurement results (here time resolution is 1 s?), and the LOD can be derived from the data. According to the result from Figure 6, the LOD for measuring HONO may be several hundred ppt (1 sigma).

Response: The reviewer is correct that Allan variance may (or may not) equal the usual variance, from which detection limits are calculated. Werle et al. (1993) state that "in the case of a white-noise dominated system, the Allan variance is equivalent to the variance of the mean and, as the variance of the mean is a measure for the detection limit, the Allan variance can be used to predict the detection limit." We added the following on line 298:

"Allan deviation analyses (Werle et al., 1993) ..... This analysis also allows an estimate of the LOD for each molecule for white-noise dominated data (Werle et al., 1993)."

In light of the reviewer's comment, we calculated standard deviations of the data shown in Figure 6 when averaged over 2, 3, 4, etc. seconds. We added those standard deviations to the plots in Figure 6 (in red color) and included those plots in our response here.

For NO_2, the Allan and standard deviations give quasi-equivalent results. For HONO, the results were dependent on the length of the data sample averaged: When all data were included, the plot resembled Fig. 6 of (Duan et al., 2018) because the average (used in the calculation of standard deviation) is "moving" i.e., changes due to drift. When a shorter segment (~2,000 seconds) of data are averaged, the Allan and standard deviations are also quasi-equivalent (see graph below). Hence, the use of Allan deviation to estimate the LOD is justified.



5. Line 110. How about the temperature control of CCD in the operation?

Response: The CCD was Peltier-cooled to -80 °C when in operation. We added the following on line 132:

"The instrument was turned on 30 min prior to measurements to allow for the LED temperature to stabilize and the CCD camera to cool to its operating temperature of -80 °C. "

Minor comments:

6. Line 160. Temperature sensor and pressure sensor mentioned are missed in Fig. 1.

Response: We modified the text section as follows:

" .. 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."

7. Line 103. Please add the manufacturer information of the spectrometer.

Response: We inserted the requested information: "The grating spectrometer (spectrograph and camera; Princeton Instruments Acton SP2156) has been described by Jordan et al. (2019)."

8. Line 125-130. The purity of N2 and Ar should be given.

Response: The gas purities are stated in Table 2. No changes were made in response to this comment.

9. Is the "celllength" the same as "cavity length" in the text, please unify.

Response: Corrected as requested.

10. Unify the font size of the title of Section 3.4

Response: Corrected as requested.

11. Line 356. pppv correct to pptv

Response: Fixed. Thank you.

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