Response to interactive comment on manuscript on “Tropospheric HONO Distribution and Chemistry in the Southeast U.S.” by A. Neuman

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General comments: *The manuscript "Tropospheric HONO Distribution and Chemistry in the Southeast U.S" by Ye et al. presents airborne measurements of reactive nitrogen compounds in the troposphere. They measure HONO to be larger than can be explained by known formation processes and find that known mechanisms explain only 20% of the daytime HONO source in background air masses. Understanding HONO formation and loss is important to understanding the photochemistry of the atmosphere, but the results here require further support to be useful in constraining reactions that produce HONO. Some specific concerns are detailed below.*

Response: We would like to thank Andy Neuman for his time and efforts in preparing this detailed and comprehensive comment. We have revised the manuscript accordingly to address his questions and concerns. Specific concerns and questions are addressed below in this Response .

Major concerns:
Q1: *The discussion of the measurements and their uncertainties are insufficient, and many of the experimental descriptions are qualitative. Substantially greater quantitation is required to support the stated 1 ppt detection limit. For example, zeros were performed "periodically" (line 125), and the baselines were subtracted from the total signal. How frequently were these backgrounds performed, and how was the back- ground determined outside of the zero periods? Was a single value used for a flight, or was the background determined by interpolating between zeroes?*

Response: HONO measurement technique has been described in detail in the previous method paper (Zhang et al., 2012), therefore only brief description of the instrument was given in the manuscript to provide the key pieces of information (lines 117 – 144 in the original manuscript). We have added significant amount of information to the revised manuscript as suggested. To answer the reviewer’s questions, more details are provided below; please refer to the method paper (Zhang et al., 2012) for instrumental details, such as HONO sampling, baseline substation and interference correction, nitrite derivatization, and absorbance measurement of azo dye derivative by LPAP technique.

HONO was measured by two separate LPAP (long-path absorbance photometer) systems. Each system ran a 30-min measurement and zero cycle, with 20 min sampling ambient air and 10 min sampling “zero-HONO” air for baseline correction, and with a 15-min time offset between the two sampling cycles (Figure 1a). The “zero-HONO” air was generated by directing the ambient air stream through a Na_2CO_3-coated denuder to remove HONO while allowing most of interfering species (NO_x, PAN, and particulate nitrite) to pass through. The combination of overlapping ambient signals from the two systems provide a continuous HONO concentration measurement (Figure 1b, solid black circles). The absorbance signals were sampled at a rate of 1 Hz (Figure 1a, blue and red circles), and were averaged into 1-min or 3-min data (Figure 1b, blue and red circles, 1-min averaging). The averaged signals were converted into concentrations based on calibration slope and air sampling and liquid flow rate information. The baseline correction was made by subtracting the ambient signals by the extrapolated line between the two adjacent stable “zero-HONO” air signals (Figure Sb, blue and red lines). The “zero-HONO” air signals were stable most of the time, and the slow drift in the baseline can be easily corrected for. The baseline was sometimes found to change rapidly in two circumstances: when the aircraft was transacting through a high NO_x plume, and when the ambient pressure changed rapidly and significantly during the rapid ascending to or descending from high altitudes. For the first case, the interference from other reactive nitrogen species in high NO_x plumes can be corrected by subtracting from the ambient air signals the increases in the “zero-HONO” air signals measured by the other HONO system. However, this correction was rarely needed, since the increases in the “zero-HONO” air signals were usually quite small even in the urban and power plant plumes (Figure 2). In the second circumstance, large baseline drifts were observed when the flow state of the scrubbing solution was disturbed by rapid pressure fluctuations. The up-shifting or down shifting of the baseline may result in over-correction or under-correction; the over-correction could then result in negative concentration numbers, as Andy Neuman pointed out. Fortunately, altitude changes in the PBL during the race-track profiling did not disrupt the liquid flow pattern enough to cause rapid baseline shift (Figure 1a). If the baseline shift was found to be caused by rapid pressure fluctuations and if reasonable baseline correction could not be made, the data points were excluded from analysis, regardless of the sign or magnitude of the data.

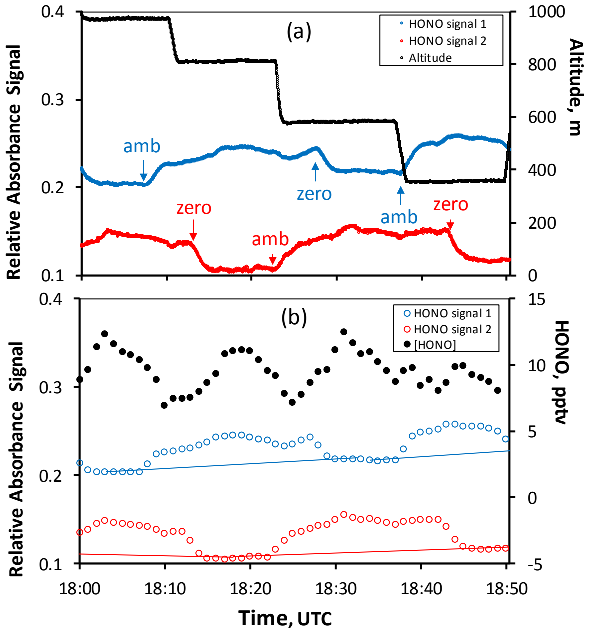

Figure 1. Time series of 1-Hz raw absorbance signals (blue and red circles) and flight altitude (black circles) (a) and of 1-min averaged absorbance signals ((blue and red circles) and calculated HONO concentrations (black solid circles) (b), during NOMADSS RF#4 on June 12, 2013. The blue and red lines in panel (b) are the baselines extrapolated from the two adjacent “zero-HONO” air measurements.

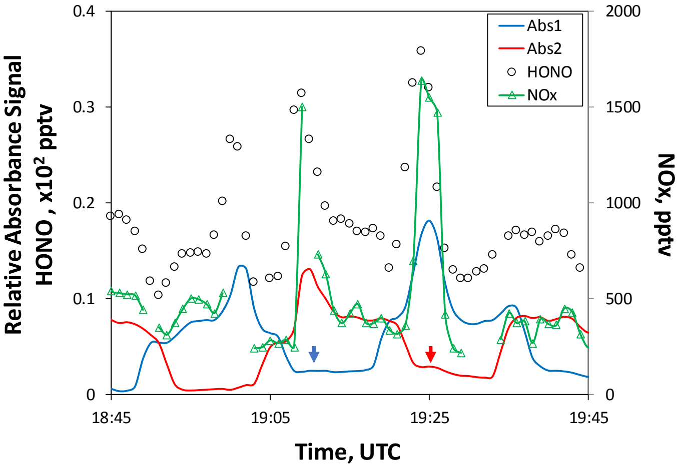

Figure 2. One-hour time series of 1-Hz absorbance signals from two HONO systems (blue and red lines), 1-min averaged HONO (black circles) and NO_x (green triangles with line) during NOMADSS RF11 on June 29, 2013. The blue and red arrows indicate the slight increases in the "zero-HONO" air signals due to potential interferences from NO_x, particulate nitrite and PAN in the power plant plumes.

The time resolution is defined as the 90% response time based on the signal transition from "zero-HONO" air to ambient air. The lowering of the flow rates of scrubbing and reagent solutions and increase in the length of liquid plumbing tubing resulted in a longer response time (200 s) compared to that reported for the ground-based system (110 s) (Zhang et al., 2012). The lower detection limit of the method was estimated to be ≤1 pptv, based on 3 times the standard deviation of the zero air signal (N >10). An overall uncertainty of ±(1 + 0.2 [HONO]) pptv was estimated, combining the uncertainties in signal acquisition and processing, air and liquid flow rates, standard preparation, and baseline correction. Again, the estimated overall uncertainty of ±(1 + 0.2 [HONO]) pptv is significantly higher for the aircraft HONO measurements than that of ±(1 + 0.05 [HONO]) pptv for the ground HONO measurements (Zhang et al., 2012), in part due to pressure fluctuation and baseline drifting on the aircraft.

***Q2:** The inlet residence time of 0.8 s is very large. What happens in a NOx plume? Wouldn't there be a contribution from NO2 conversion to HONO on the inlet? A description of the inlet length and flow would be helpful.*

Response: Andy Neuman has made some fair comments regarding the long inlet deployed on the C-130 for HONO measurement. It was only during the instrument integration when we learned that there were exhaust vents next the inlet ports near our instrument location. The vented aircraft cabin air might significantly contaminate our HONO measurement. To avoid the potential contamination artifacts, the inlet port on the other side of the aircraft was used. A heated 7-m long 3/8"-ID PFA inlet line was thus needed and used, and a high flow rate (210 L min^{-1}) ambient air was drawn by an auxiliary blower to reduce the air sample residence time in the inlet line. The resulting residence time in the inlet line is 0.14 s, not 0.8 s stated in the original manuscript. We regret the error and have made the correction in the revised manuscript.

Our group has examined the potential interference from heterogeneous NO_2 reactions on the inlet wall surface on HONO measurements many times and in different environments, and have found it not to be significant. Figure 3 shows the result of such an experiment conducted recently in downtown Albany. HONO in the ambient air was measured by two HONO systems, one with a regular inlet, and the other with or without adding long piece of heated PFA tubing (10 m long, 1/4-OD and 1/8”-DI). At a sampling flow rate of 2 L min^{-1}, the residence time of air sample in the PFA tubing was ~2.4 s, about 17 times longer than 0.14 s for the aircraft systems. The ambient NO_2 concentration varied from ~1 to ~6 ppbv during the measurement period. The comparison of the two time series by measured the two HONO systems shows no discernible difference within the estimated uncertainty, regardless if the extra long tubing was added or removed (Figure 3).

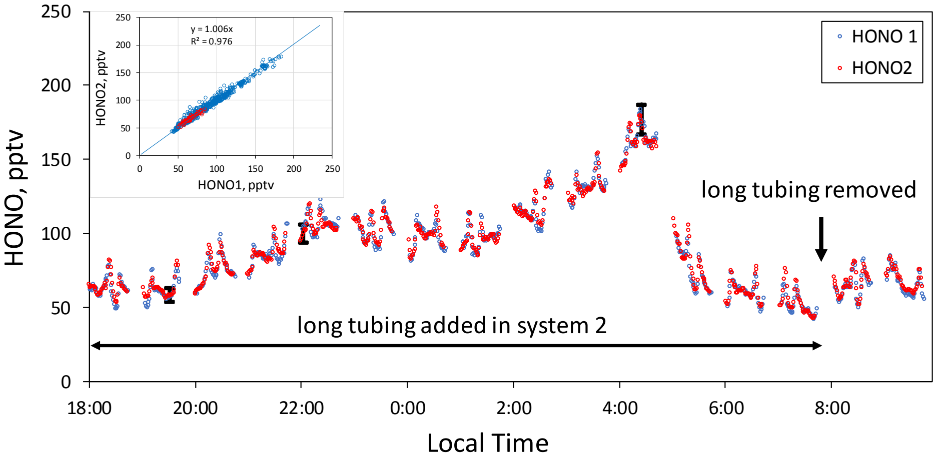

Figure 3. Ambient HONO concentrations measured by two HONO systems in Downtown Albany during April 19-20, 2016. A 10-m PFA tubing (1/8”-DI) was added to the inlet of system 2 (red circles) from at 17:00 on April 19, and was removed at 7:53 on April 20, as indicated by the black arrow. The three black bars at 19:31 and 22:02 on April 19 and at 4:25 on April 20 indicate estimated measurement uncertainties at the measured concentrations. The insert is the scatter plot of the measured HONO concentrations by the two systems. The red symbols represent the measurements by the two systems with the same short inlets (10-cm long, 1/16”-ID), and the blue symbols represent the measurements when an additional 10-m tubing (1/8”-ID) was added to system 2. The line is the linear best fit for the data.

The accuracy of HONO measurements was also confirmed by comparison with a limb-scanning differential optical absorption spectroscopy (DOAS) (Ye et al., 2016a). When measuring in wide power plant plumes where HONO mixing ratios exceeded the lower detection limits of both instruments, the agreement between these two instruments was very good, within the assessed uncertainties (Extended Data Fig. 3 in Ye et al., 2016a).

***Q3:** If the HONO measurement is a difference between total signal and background, I am surprised that there are no values below zero in Figures 2 and 3. Are there really never any instances when HONO falls to zero? Perhaps the interferences are underestimated.*

Response: The signals for the “zero-HONO” air were quite stable, and ambient signals were well above the baselines, even at the data sampling rate of 1 Hz (Figure 1a). As explained in the response to Q1, ambient signals are always higher than the baseline signals extrapolated from the adjacent “zero-HONO” signals, except during the rapid ascending to and descending from high altitudes; large baseline drifts were observed when the flow state of the scrubbing solution was disturbed by rapid pressure fluctuations. Overcorrection of the upward-shifting baseline may sometimes result in negative values in HONO concentration. However, the data were excluded from analysis if the baseline shifts caused by rapid pressure fluctuations could not be reasonably corrected, regardless of the sign or magnitude of the data.

***Q4:** Please mention briefly how surface area density was determined from SMPS data. Wouldn’t SMPS also provide a constraint on aerosol mass that could be useful for verifying the pNO3 measurements? Some of values of pNO3 in remote regions are very large - up to 0.5 and Have similarly large nitrate values been measured outside of urban plumes over the SE US in other studies?*

Response: The surface area density was calculated by the following equation:

$$\mathrm{S/V} = \sum(4\pi r_i^2) \times n_i$$

where r_i and n_i represent the radius and number density of aerosol particles. A perfect sphere was assumed for aerosol particle in the calculation.

The mean (±1std) and median of pNO_3 in the Southwest US were 76 (±45) pptv and 66 pptv in the PBL, and 35 (±39) pptv and 15 pptv in the free troposphere, within the range of reported particulate nitrate in rural atmosphere (Heald et al., 2012 and paper therein). The high pNO_3 concentrations were observed in the PBL during the first racetracks of the RFs #4 and #5 west of Centreville, AL, and during the RF 11 around Auburn, AL (Figures 1 and 2). Agricultural activities in this region may release enough NH_3 to convert some of the gaseous HNO_3 into pNO_3, as observed by Neuman et al. (2003). We have calculated the aerosol mass using the SMPS data as suggested. However, no robust relationship was found between aerosol mass and the concentration of pNO_3. We were not able to do the same analysis as that in Neuman et al. (2003) due to poor resolution and missing data points of HNO_3 and the lack of NH_3 data.

***Q5:** I cannot find the mentioned UHSAS data or DOAS data in the project archive. What does a "very good" agreement mean (line 142)? Again, quantifying the agreement and showing data would strengthen the paper.*

Why is OH estimated using a prior study (line 246), when the OH measurements listed in Table 1 could be used?

Response: No HONO data from DOAS is available in the project archive, because the ambient HONO concentrations (11.2 ± 4.3 pptv in the PBL and 5.6 ± 3.4 in the FT as measured by LPAP) were mostly below the lower detection limits of the DOAS instrument (30 pptv) during the NOMADSS study. Good HONO measurements were made by both the DOAS ant the LPAP in wide power plant plumes during RF 7 over Ohio River Valley, and results have been intercompared (Extended Data Fig. 3 in Ye et al., 2016a). We found the HONO concentrations from the two instruments closely tracks each other, and the agreements were within the assessed uncertainties. The readers are encouraged to read the paper for more information.

Both this manuscript and the "prior" paper by Kaser et al. (2015) were based on results from the NOMADSS study, and the same OH measurement dataset was shared and used by the two papers. Since the information on OH levels during the flight had been published, it is appropriate to reference the paper.

***Q6:** A very large photolysis rate for pNO3 is used to explain HONO formation, but this rate isn't consistent with the data shown. It is difficult for me to understand the difference between "determined photolysis rate" and "ambient photolysis rate" (section 3.3), but both are extremely large and comparable to the loss rate for isoprene. The nitrate photolysis rates give a nitrate lifetime of approx. one hour, which is less than the lifetime of NOx. How can nitrate ever accumulate in the atmosphere if its lifetime is so short? Are there any other studies that find a very short lifetime for nitrate? The large nitrate photolysis rate is inconsistent with the nitrate abundance and distribution reported here and cannot explain the HONO abundance.*

Response: Indeed, a very large photolysis rate constant was used for pNO_3 in our calculation. The pNO_3 photolysis rate constant was determined in the laboratory using the aerosol samples collected on board the C130 during the NOMADSS field study (Ye et al., 2017a). Several recent laboratory studies have demonstrated that surface nitric acid and particulate nitrate can be photolyzed at much higher rates than gaseous nitric acid, by 2-3 orders of magnitudes (Baergen and Donaldson, 2013; Du and Zhu, 2011; Ye et al., 2016b, 2017a; Zhou et al., 2003; Zhu et al., 2010, 2015). While NO_2 has been found to be the dominant product from HNO_3 photolysis on clean and dry laboratory surfaces (Ye et al., 2016b; Zhou et al., 2003; Zhu et al., 2010, 2015), HONO is the major product on natural surfaces and in ambient aerosols (Ye et al., 2016b, 2017a).

The "determined photolysis rate constant" ($J_{pNO_3}^N$) is the laboratory determined photolysis rate constant using the ambient aerosol samples. It has been normalized to tropical noontime conditions at ground level (solar zenith angle = 0 °), so that it can be compared with results in other studies. The $J_{pNO_3}^N$ value varies over a wide range, from 8.3×10^{-5} s^{-1} to 3.1×10^{-4} s^{-1} among the samples, with a median of 2.0×10^{-4} s^{-1} and a mean (± 1 standard deviation) of 1.9 (± 1.2) × 10^{-4} s^{-1}. A median $J_{pNO_3}^N$ value of 2.0×10^{-4} s^{-1} was used in the calculation.

The "ambient photolysis rate constant" is the pNO_3 photolysis rate constant (J_{pNO3}) under the ambient conditions. It varies with the time of the day, the location, and the cloud coverage. J_{pNO3} was calculated by scaling $J_{pNO_3}^N$ (~2.0×10^{-4} s^{-1}) to ambient light conditions using the measurement-derived J_{HNO3} (Eq. 3).

Yes, some recent studies also showed the short lifetime of particulate nitrate in low-NO_x environments (Reed et al., 2017; Ye et al., 2016a, 2017b). Many laboratory studies have also shown fast photolysis rate constant for surface HNO_3 and pNO_3 (Baergen and Donaldson, 2013, Ye et al., 2016b, 2017a; Zhou et al., 2003; Zhu et al, 2010; Zhu et al., 2015), lending support to our argument that pNO_3 photolysis can be an effective renoxification pathway. However, particulate nitrate is in a dynamic equilibrium with gas-phase HNO_3, the later accounts for a larger (or even dominant) fraction of total nitrate (pNO_3+HNO_3) and is photochemically inert. The overall photolysis of pNO_3+HNO_3 would be much slower than indicated by J_{pNO3}. In addition, oxidation of NO_x via several reactions will replenish the pNO_3+HNO_3 reservoir. Our results reported in this manuscript and in an earlier paper (Ye et al., 2016a) suggest that there is a rapid cycling in reactive nitrogen species in the low-NO_x atmosphere, sustaining the observed levels of HONO and pNO_3.

Q7: *The different air mass types are not explained, and it isn't clear if or how they were separated. Benzene is used to identify urban plumes, but how are power plant plumes and biomass burning plumes identified? Could there be a large biomass burning plume contribution to the observations? Were some plumes a combination of sources? CO or acetonitrile measurements could be used to identify air mass influences. Similarly, SO2 was measured and could be used to identify power plant plumes. I could find no mention of any meteorological conditions. Without a more thorough description of the ambient conditions and ancillary measurements, it is very difficult to compare these results with other studies.*

Response: As suggested, we have added CO, acetonitrile and SO_2 as tracers to identify plumes in the revised manuscript (Figure S1). Based on the low levels of acetonitrile during the reported flights in this manuscript, we did not observe any significant contribution from biomass burning (Figure S1). The original assignments of plumes are further confirmed by these tracers: The CO peaks in plumes U1,U2 and U3 (A, B, C in the original Figures 2 and 7) suggest that they were under influenced by urban activities, and the lack of CO peaks in plumes P1-P4 (D, E, F, G in the original Figures 2 and 7) suggest that they were power plant plumes. A high SO_2 peak also accompanied a high NO_x peak in the power plant plume P4 (G in the original Figures 2 and 7).

We did employ the meteorological information in our discussions, for examples, using the wind speed and wind direction to calculate the transport time of plumes from a power plant (original lines 358-362) and back trajectories in explaining horizontal HONO variations (original lines 401-409). Ancillary measurements, including OH, NO, NO_2, aerosol number and size distribution, isoprene, J values, …, were used in calculations and discussion throughout the manuscript.

Q8: *The large reduction in PBL mixing time (line 262) between noon and afternoon is very surprising and differs from previous studies. By noon in the summer, the mixing time should be much less than 1.5 h.*

Response: The mixing time can be influenced by several factors, such as the surface albedo and cloud coverage. The longer than expected mixing time was calculated using the vertical isoprene profile and may be due to the combined effect of these factors. We have also found significant variations in mixing time in RF #4, #5 and #17. Nevertheless, HONO photolytic lifetime was still much shorter than the mixing time even if the later was reduced by half; the conclusion of this section remains unchanged, i.e., the contribution of ground HONO source was not important to the overall HONO budget in the PBL, due to low ground source strength and/or slower transport than its photolysis loss.

Q9: Relevant literature is not referenced, and the differences with previous measurements are not discussed. We published a very similar paper, using aircraft HONO measurements at the same time and location and under the same SAS umbrella (Neuman at al., HONO emission and production determined from airborne measurements over the Southeast U.S., JGR, 2016), but oddly, that paper is not referenced. We found that known HONO production mechanisms explained the HONO abundance, and we did not need to invoke unknown sources. In contrast, the studies referenced in the introduction (lines 29-30, line 103) report much larger values ranging from 100s of pptv to ppb levels. Why do the HONO values reported here differ from previous measurements, which range from indistinguishable from zero to ppbv levels? Meaningful comparisons to previous studies (some conducted at the same time and location) are essential for understanding the findings reported here.

Response: We finished our first draft of this manuscript over two years ago, before the publication of the mentioned paper (Neuman et al., 2016). Although we have made significant changes to the draft during the subsequent revisions, we failed to update the references. We regret the omission. The paper by Neuman et al. (2016) has been referenced and discussed in the revised manuscript (lines 64, 95, 211, 215, 239, 282, 403).

We agree that meaningful comparisons to previous studies are essential for understanding the reported data. We compared our results with those from other two airborne studies (Zhang et al., 2009; Li et al., 2014) in the original manuscript, and have added more discussions and comparisons with Neuman et al. (2016) in the revised manuscript.

Although the two aircraft studies, SENEX on NOAA's WP-3D and NOMADSS on NSF/NCAR's C-130, were conducted at the same time and location and under the same SAS umbrella, they had been focused on somewhat different objectives. The main objective of TROPHONO project (one of the three projects in NOMADSS) was to investigate daytime HONO formation mechanisms and the role of nitrate photolysis in aerosol particles in the cycling of reactive nitrogen species in the troposphere. All the C-130 flights were conducted in the daytime during NOMADSS except the RF#18 (from late-afternoon to midnight). And the results reported in this manuscript were mostly from rural background air masses, with only a few small urban and power plant plumes in RE#11. On the other hand, the WP-3D spent far more time in various plumes and at nights during the SENEX (Neuman et al., 2016). We would like to point out that there are actually no major disagreements between the two aircraft-based studies when the overlapped measurements are compared. Similar to what reported by Neuman et al. (2016), we found that the NO_x-related reactions (mainly the NO+OH reaction) accounted for nearly all the required HONO source in the large fresh power plant plume (NO_x ~ 20 ppbv) encountered during the RF #7 to Ohio River Valley (lines 375-378 in the original manuscript). In the low-NO_x background air masses, the mean HONO concentration was 11.2 ± 4.3 pptv in the PBL and 5.6 ± 3.4 in the free troposphere (Table 2), which is within the range from -15 pptv to 10 pptv (±15 pptv uncertainty) (Neuman et al., 2016).

As pointed out by Andy Neuman, the studies referenced in Section "1. Introduction" reported significantly higher HONO concentrations, up to hundreds of pptv in the rural environments and several ppbv in the urban environments. The reported values include lower daytime and higher nighttime HONO concentrations. Most of these measurements were made on ground stations, and thus under direct influence of the ground HONO sources, including direct emissions (combustion sources and soil emission sources), heterogeneous and photochemical reactions of precursors (e.g., NO_2, PAN and HNO_3) on ground surfaces, and gaseous reactions with elevated reactant concentrations. The measurements on aircrafts, on the other hand, minimized the influence of the ground HONO sources, as we discussed on section 3.2. Therefore, the airborne measurement data would provide a better insight into the HONO chemistry within an air parcel.

***Q10:** Line 85 states that nearly all HONO measurements have been made at ground sites, but that dismisses the many studies of HONO vertical gradients using DOAS and from towers (e.g. Young et al, Vertically Resolved Measurements of Nighttime Radical Reservoirs in Los Angeles and Their Contribution to the Urban Radical Budget, ES&T, 2012; Stutz et al., Simultaneous DOAS and mist-chamber IC measurements of HONO in Houston, TX, Atmospheric Environment, 2010; Vandenboer 2013 in the references). And the authors themselves have many papers that detail airborne measurements.*

Response: We would like to point out that the HONO gradient measurements using DOAS and from towers are still ground-based, and that we did reference many of the related literatures (Kleffmann et al., 2003; Li et al., 2014; Stutz et al., 2002; Villena et al., 2011; Wong et al., 2011, 2012, 2013; Ye et al., 2015; Zhang et al., 2009) when discussing HONO vertical measurements and airborne measurements in the introduction (the paragraph starting line 85 in the original manuscript).

***Q11:** smaller points I don't know what an N(V) level is (line 216)*

Response: We have much few data points of HNO_3, due to poor time resolution and more technical difficulties with the system (bubble formation/baseline shift, especially at high altitudes). The HNO_3 levels in the PBL were 305 ± 87 pptv in RF4, 291 ± 81 pptv in RF 5, 342 ± 108 pptv in RF11, 105 ± 38 pptv in RF 17, and 206 ± 73 pptv in RF18, accounting for 70% - 85% of (N(V).

***Q12:** Data averaging is not explained. The time resolution of HONO and pNO3 are listed as 3 min and 6 min, yet 1 min data are shown. How are the data averaged in figure 3? The values do not match those shown in Figure 2, but the binning and averaging are never described.*

Response: The absorbance signals were sampled at 1 Hz, much higher rate than the time resolutions of HONO and pNO_3 (see Figure 1 in this Response). The 1-min or 3-min averages were used to convert absorbance signals to concentrations, based on flow rate and calibration information (please see our responses to Q1 and Q2 for more details on method and data processing). We have added the above information to the revised manuscript and have used 3-min HONO data and 6-min pNO_3 data for the revised figures, as suggested.

***Q14:** Figure 2 shows pNO3 in ppbv, which is in error.*

Response: Thank you for pointing out the error; the error has been corrected.

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