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Discussions

Interactive comment on "Tropospheric HONO Distribution and Chemistry in the Southeast U.S." *by* Chunxiang Ye et al.

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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.

1) 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 background determined outside of the zero periods? Was a single value used for a flight, or was the background determined by interpolating between zeroes? 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. 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.

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 ug/m3 and sometimes comparable to NOx, so it would be useful to have other measurements to support this. Have similarly large nitrate values been measured outside of urban plumes over the SE US in other studies?

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?

2) 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.

3) 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.

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.

4) 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.

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.

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

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.

Figure 2 shows pNO3 in ppbv, which is in error.

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