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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Emission of nitrous acid from soil and biological soil crusts represents a dominant source of HONO in the remote atmosphere in Cyprus" *by* Hannah Meusel et al.

Anonymous Referee #3

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Summary: Soil samples used in this work are from soils collected from the field, manipulated in a controlled lab environment, and then measured fluxes extrapolated to compare with the missing HONO source calculated for the CYPHEX field campaign in the same location. Soils were collected and categorized from a gridded sampling scheme. HONO and NO fluxes were measured from the soils in replicates in order to quantify which surface soil community members, if any, were responsible for the majority of the HONO fluxes observed. The authors performed nice controlled experiments in the lab and found some interesting conclusions, counter to previous findings in similar soils by this group. The manuscript may be acceptable for publication in Atmospheric Chemistry and Physics, subject to a number of concerns being addressed.

Major Comments: 1. There is no experimental control of soils devoid of microbial activity for each soil type. One would think it necessary to fumigate (or otherwise kill) soil samples of each type to control for biotic versus abiotic HONO and NO emissions, yet this is not presented. If possible, the authors should consider acquiring this data and adding it to the manuscript for comparison and correction of the dataset.

2. The consideration of the effects of the measured soil pH on HONO release using the method of the Su et al. (2011) work is not considered in the interpretation of the data. What proportion of the emissions measured in each case can be ascribed to simple partitioning? What effect does this have on comparisons between soil types considered in this work when abiotic exchange is estimated versus measured (see comment above) in the experimentally measured fluxes? A major concern is that if abiotic partitioning from dead soils is not favored by calculation from bulk pH measurements and nutrient loadings (i.e. soil pH » pKa HONO) then another emission mechanism is active and should be considered/discussed.

3. Scaling to atmospheric relevance for the field campaign is very interesting, but inappropriate to include in the title of the manuscript. The linkage between microbial activity and HONO and NO emissions is of growing importance to constrain and these lab-based measurements help to do so. However, the uncertainty in the extrapolation of the data to the field campaign observations of the missing daytime HONO source covers an order of magnitude range, which means at the low end of the estimate these processes account for < 10 % of the daytime HONO source. The authors do not discuss this limitation and should do so. The title of the manuscript should also remove the HONO budget closure implications due to this significant uncertainty. An important consideration here is one that has been reported and discussed much in the atmospheric community over the past 5 years and that is the vertical structure of HONO, and by proxy, the daytime HONO source strength near the ground surface. Using soil HONO emissions to scale to measurements made nearly 6 m above the ground may add additional error in the budget closure calculation as the perceived missing source

changes with height. This topic and the relevant references would be a worthwhile addition to this component of the discussion as there are several reports on vertical structure of HONO in arid agricultural and rural regions in the US.

4. The quality of the nutrient data cannot be determined as no accuracy, precision, or detection limit values are presented. The data table in the supplement reports measurements of zero, which should be represented by '< LOD' and the detection limits determined by the experimental runs calculated (do not use the instrument manufacturer's stated values). My concern is that some of the measurements made on the samples are near the detection limits and therefore highly uncertain, which may confound the comparisons made throughout the manuscript. The authors should also be more cautious in their reported values from these measurements. Are these analytically certain to so many significant digits?

5. The experiments performed on nutrient content before and after experiments (Figure 4) is good to have performed, but this data does not need to be presented in the figure and would help make this a cleaner plot. The data can be replaced with one sentence in the manuscript stating that the nutrient levels were not different in the soils by performing the flux experiments. To that end, this suggests that the full set of measurements of these nutrients could be pooled and improve the statistical analyses performed, as it would reduce the standard error in the measurements. Further to this point, where replicate measurements have been made, the authors should be presenting the standard error of the mean and not the standard deviation as it aids in connecting the reader to the statistical results. The results and discussion surrounding the purpose, method selection, and outcomes of these statistical tests needs to be improved either through clarity in existing sections or expansion of the text.

6. Details on linear regression in Figure 7 needs to be presented. There is presumably large uncertainty in both measurements being compared and the appropriate considerations must be included prior to assessing the relationship between them, along with the associated uncertainty in the result. Building on this, the direction of the trend

and also its associated uncertainty from the regression fit may be more telling towards whether the strength of the coefficient is truly robust or being limited by the sample size available.

7. Figure 2 needs to be streamlined to present the relevant information for the contents of the manuscript. The level of detail here is not necessary.

Minor Comments:

Fix tense and plurality issues throughout the manuscript.

Many sentences have issues with comma splicing, making them long and the purpose of the sentence difficult to follow.

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