Review for Roberts and Liu, 2018

This manuscript by Roberts and Liu presents a fundamental laboratory experiment to determine thermodynamic data required to predict atmospheric fates of HNCO, CH3NCO, and three XCN species. HNCO and CH3NCO are toxic volatile organic compounds for which an accurate understanding of their atmospheric fate is critical. In particular, ambient measurement of HNCO was first made possible a few years ago by Roberts and coworkers themselves. Although a number of studies have focused on the chemical behavior of HNCO since then, fundamental thermodynamic data, such as those presented here, are still lacking. XCN species are novel species whose atmospheric importance has been implied but has not been fully established.

The experiments were conducted with well established methods, measurements and analyses were performed with cautions, and the choice of experimental conditions is thoughtful. Publication in ACP should be considered, but not before substantial revisions are made.

Major comments:

1) The use of Henry's law constant.

Throughout the entire manuscript, I am concerned about the current use of effective Henry's law constant (H_{eff}) vs the intrinsic Henry's law constant (H_{HNCO} or H). It seems that the authors fully understand the difference between the two, though the usage of H and H_{eff} is inconsistent and misleading. I would recommend the authors first use a few sentences in the introduction or method to clarify the difference between H_{eff} and H, and then revisit each H and H_{eff} throughout the manuscript to revise them accordingly.

Here are some particular examples:

- Line 309 and Eq. (1). My understanding is that the H determined using the experimental method and Eq. (1) is in fact H_{eff}. The authors should clarify that.
- The only intrinsic Henry's law constant appears is in Eq. (4) and related discussions. The authors decide to temporarily use H_{HNCO} here.

2) Atmospheric implication (Section IV) is one of the most important sections in the manuscript and requires some revisions. In particular:

- While the focus of this study is heterogeneous processes, the authors mention about the gas-phase fates of HNCO and CH3NCO in a rather sporadic manner. I was under an impression that the gas-phase loss of HNCO and CH3NCO is less important than the heterogeneous process, until I saw the OH rate coefficient of CH_3NCO (3.6e-12 cm3 molec-1 sec-1) and realized that it is actually very important for CH_3NCO. I would suggest the authors extend the discussion of atmospheric fate to include gas-phase loss processes for a more complete picture.
- The introduction to the loss processes, e.g., deposition velocity, uptake coefficient, etc., is very insightful and resourceful. However, the authors perform the actual analysis at a rather abstract level after a full-bodied introduction. In particular, the fate of HNCO is summarized into a couple of numbers in Table 2, which in a sense self-negates all the detailed analyses performed by authors themselves. Given all the HNCO data at different pH and temperatures etc., HNCO

deserves a more detailed discussion in a separate paragraph, and perhaps with additional diagrams.

- It is surprising that the in-cloud rxn value of HNCO in Table 2 is directly taken from another study. Why don't the authors derive this value from their own data from this study using Eq. 16? I thought that was the whole purpose of doing all the analyses for HNCO.
- HNCO and other compounds' water solubility varies significantly across temperatures and pH. What condition is used to derive logKow in Table 2? No explanation is provided. When the Henry's law constant of HNCO varies to such an extent, is Kow of HNCO helpful at all?

3) Miscellaneous typos, mistakes, etc. Each of them is minor by itself, but the overall quality of this manuscript should be improved to achieve a professional level.

Minor and technical comments:

- Line 204 "at several at several"
- Line 284: as the authors point out, the selectivity of the detection method is indeed important. Did the authors try using CIMS and PTRMS which should be able to verify the selectivity of the Nr method?
- Line 314 "phi/V"
- Line 324 "cc/min" should be made consistent with "ml/min" used previously (Line 297)
- Line 345 redundant
- Line 348 "volumetric flow rate to solution volume" is already defined as phi/V previously.
- Line 351 "lass rate"
- Line 365 should define effective Henry's law coefficient as H_{eff} here.
- Line 386 394. Please consider citing this paper for salting in/out and Setschenow constants: Wang et al. EST 2014 10.1021/es5035602
- Line 411 R10: out of curiosity, H2O can be technically treated as a type of ROH. Any suggestion on why the reaction mechanisms of HNCO towards H2O and ROH are different?
- Line 624-625: By using an extremely polluted condition, I guess the authors are trying to derive the lower limit of Aerosol Dep. lifetime. This should be clarified somewhere, perhaps as a notes to Table 2.
- Line 682 check the unit of k
- Figure 4: The figure contains data for Heff HCN, but nothing is mentioned in the caption.