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Atmospheric Chemistry and Physics Discussions

Interactive comment on "How does deposition of gas phase species affect pH at frozen salty interfaces?" *by* S. N. Wren and D. J. Donaldson

Anonymous Referee #2

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The authors present a study of pH at the gas-ice interface using glancing-angle laser-induced fluorescence spectroscopy. They monitored changes in pH at the surfaces of samples of frozen salt water, artificial seawater or freshwater upon exposing the samples to HCl or NH3. The choice of ice samples is intended to reveal any differences in the pH behavior of "Brine" (salt water) vs. "quasi-liquid layer" (freshwater) type interfaces. pH was determined using a surface-active organic indicator (harmine).

This article may be suitable for publication in ACP after the following comments are addressed:

- The gas-phase HCl and NH3 concentrations used in this study are much higher than atmospherically relevant concentrations. A true liquid HCl-water solution (i.e., a brine) is known to form when pure water ice is exposed to HCl in sufficiently high concen-

trations (see phase diagram from Molina (1994), reproduced in McNeill et al., 2006). I am concerned that these experimental conditions may lie in that part of the HCl-ice phase diagram, possibly complicating the interpretation of the experimental results for the freshwater ice samples. Can the authors justify the choice of such high gas-phase concentrations? Was pH investigated at any other gas-phase concentration?

- Can the authors comment on how the presence of Harmine, a large organic molecule, might perturb the chemical or physical state of the ice surface, especially when brine is not expected to be present?

- More technical specifics regarding the experiments performed and the results should be provided in the abstract.

- A few more experimental details regarding the use of Harmine as a pH indicator at the ice-air interface should be given rather than referring the reader to Wren and Donaldson for all experimental information regarding this technique.

- Figure 3 caption should read: "100 ppm of HCl in N2 was introduced to the chamber"

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 19277, 2012.