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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-

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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.

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