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CO2 hydrate equilibria at atmospheric partial pressures, and implications for atmospheric acidity

Brian Durham[1] and Christian Pfrang[2]
[1]University of Birmingham, United Kingdom of Great Britain and Northern Ireland (brian@oxpot.demon.co.uk)
[2]University of Birmingham, United Kingdom of Great Britain and Northern Ireland (c.pfrang@bham.ac.uk

We take moist air (and artificial air) at +0.35 ºC, variously doped with CO_2, and pass it through a chamber chilled to -3ºC. We record any depletion of CO_2 in the gas stream using differential NDIR spectrometers, and we then de-gas the melt-water to measure the captured CO_2, again with NDIR. Preliminary results are consistent with published curves for the CO_2/hydrate equilibrium at partial pressures relevant to the petrochemical industry and to industrial carbon capture as determined by Raman microscopy (Chazallon and Pirim 2018).

Extension of the CO_2/ice curve to atmospheric partial pressures allows a review of a range of related issues in atmospheric water, one of the more accessible being its acidity. The dissociation of CO_2 in water at equilibrium with 400ppm CO_2 at Earth surface is quoted as pH5.6, but fresh rainwater (and snow-melt) can be significantly more acidic, decaying exponentially to equilibrium with a half life of around four hours. This observation is consistent with published analyses of [CO_2] in rainwater that are significantly higher than the Henry equilibrium (Warneck 2000). Both would be explained if convection was resulting in CO_2/ice-formation in clouds within the boundary layer, in turn leading to deposition of supersaturated levels of CO_2 together with enhanced acidity.

This paper speculates on the local [H_3O^+] delivered by the melting of an ice particle that had grown in an atmosphere of 400ppm CO_2 at an altitude of 1km, and its dissipation through dilution by neighbouring unfrozen water drops and by slow release from the residue of hydrate caging in liquid water. For Earth surface it has potential implications for acid rain, for the solution and redeposition of carbonate rocks, and for ocean acidification.

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