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'Caged': Is capture of CO₂ hydrate a reality in the atmospheric water cycle?

Brian Durham and Christian Pfrang Oxford, United Kingdom (brian@oxpot.demon.co.uk)

This paper builds on our poster presentation to RMetS Annual Conference Exeter 2017 summarising available analyses of CO_2 in rainwater. Titration, volatile acidity and degassing have each yielded values that are supersaturated with respect to the Henry equilibrium in pure water (see chart), and this paper raises a potential source of 'supra-Henry' capture via hydrate cages in cloud ice.

The available literature on the thermo-chemistry and kinetics of CO_2 hydrate and semi-clathrate is currently repositioning itself away from avoidance of hydrate blockages in petrochemical processes and towards capturing gases in fossil fuel electricity generation, relevant to climate change (Chernov et al 2016). The enthalpy change reported for fusion of CO_2 hydrate looks promising for capture (Anderson, G. K. 2002). Industrial capture presently demands a promoter molecule and around 50 atmospheres pressure (Gholinezhad et al 2011), neither of which would be available in the atmosphere, but industry also demands a relatively high capture rate, while the mol fraction target implied by the above analyses of atmospheric deposition is lower by several orders of magnitude.

Given reported supra-Henry values for CO_2 in rainwater and snow-melt, could the quasi-liquid film enveloping a growing cloud ice particle under convective chilling provide a kinetic and thermo-chemical environment favourable to hydrate formation?