Molecular Dynamics simulations indicate solvation and stability of single-strand RNA at the air/ice interface, supporting a primordial RNA world on Ice

Steven Neshyba[1], Ivan Gladich[2], **Penny Rowe**[3], Maggie Berrens[1], and Rodolfo Pereyra[4]

[1]University of Puget Sound, Chemistry, Tacoma, Washington, United States of America (nesh@pugetsound.edu)
[2]Hamad Bin Khalifa University, Doha, Qatar (igladich@hbku.edu.qa)
[3]NorthWest Research Associates, Redmond, Washington, United States of America (penny@nwra.com)
[4]National University of Córdoba, Córdoba, Argentina (pereyra@famaf.unc.edu.ar)

Timeline of the hypothesized RNA World

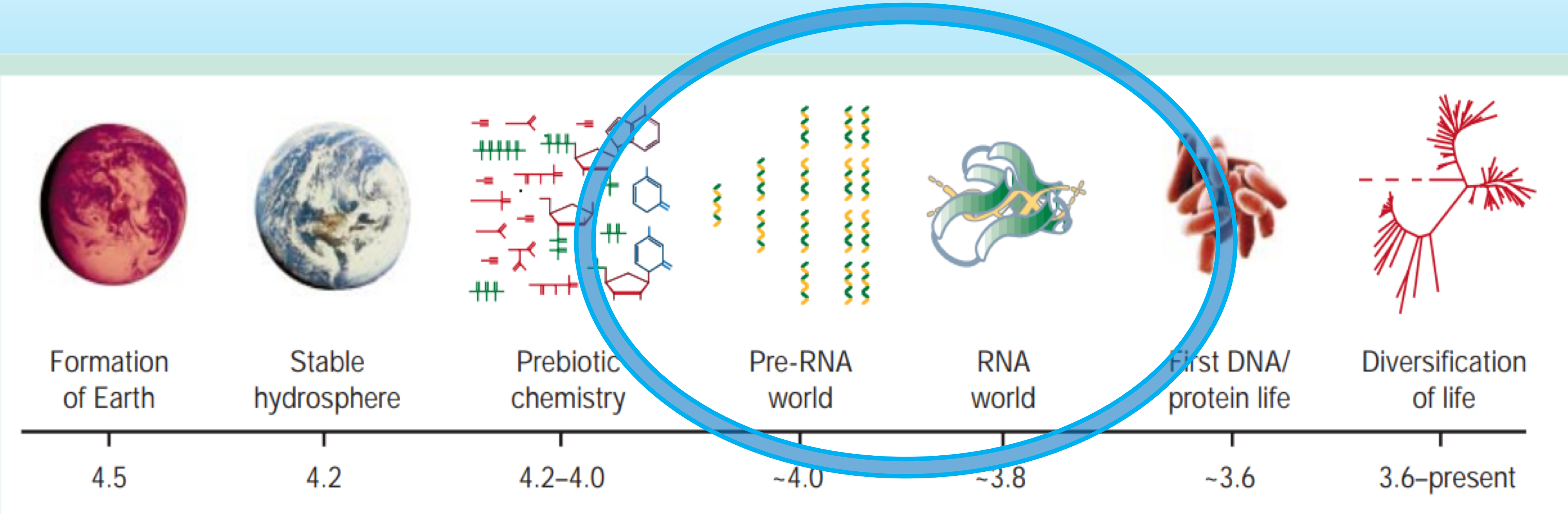
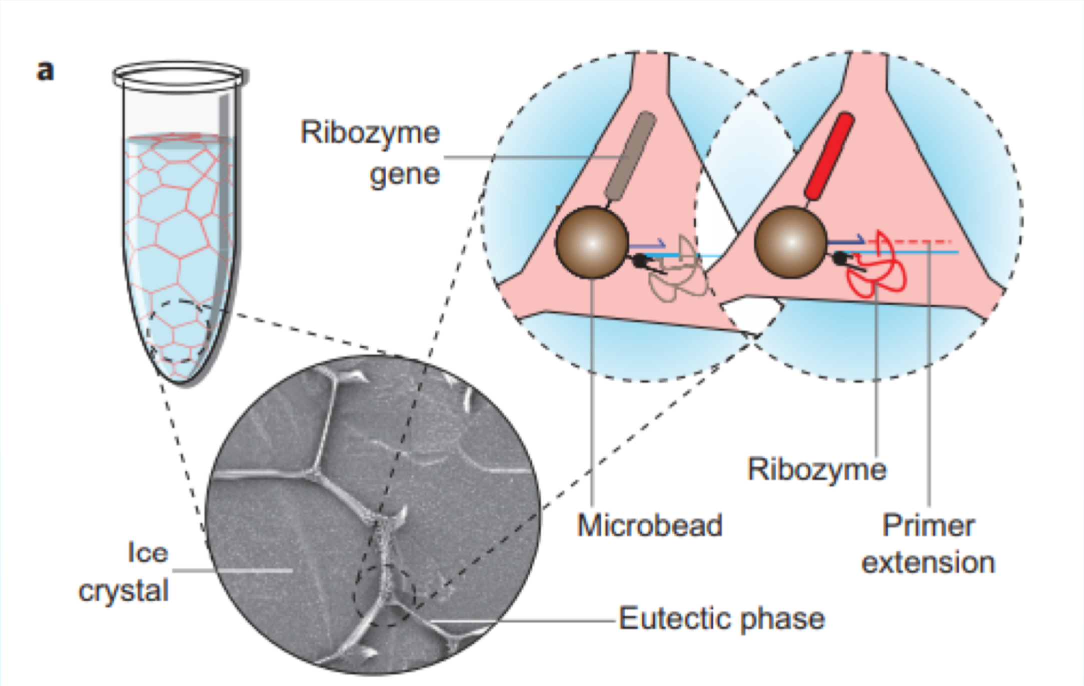


Figure 1 Timeline of events pertaining to the early history of life on Earth, with approximate dates in billions of years before the present.

Joyce, 2002

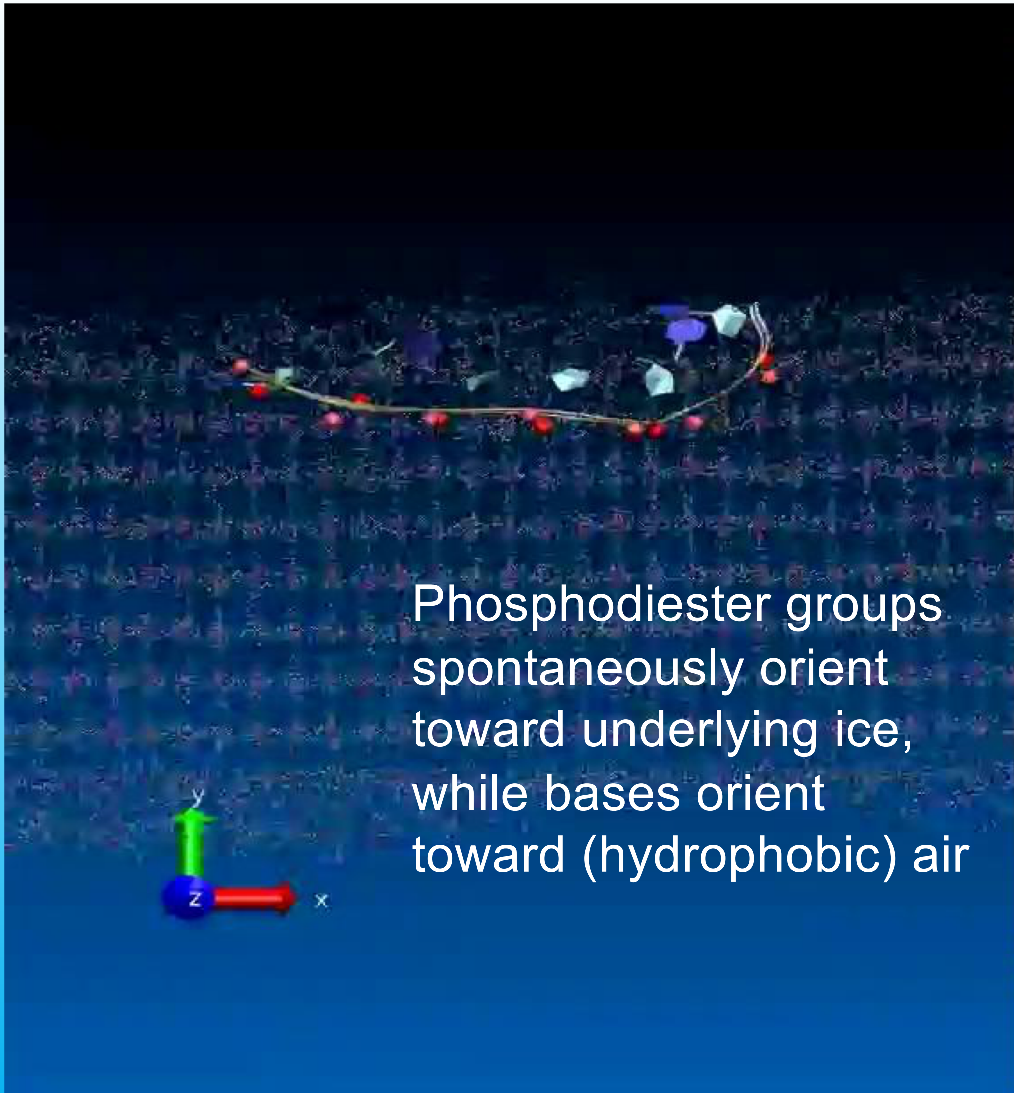
Low temperatures as a remedy for the hydrolysis problem

Attwater et al, 2010:
"Our results support a wider role for ice as a predisposed environment, promoting all the steps from prebiotic synthesis to the emergence of RNA self-replication and precellular Darwinian evolution"



Attwater et al, 2013

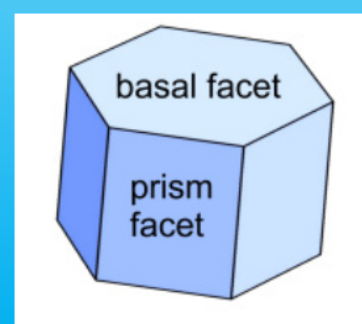
Molecular Dynamics (MD) of single stranded RNA on the basal facet of ice



Sequence: CCUUCGGG

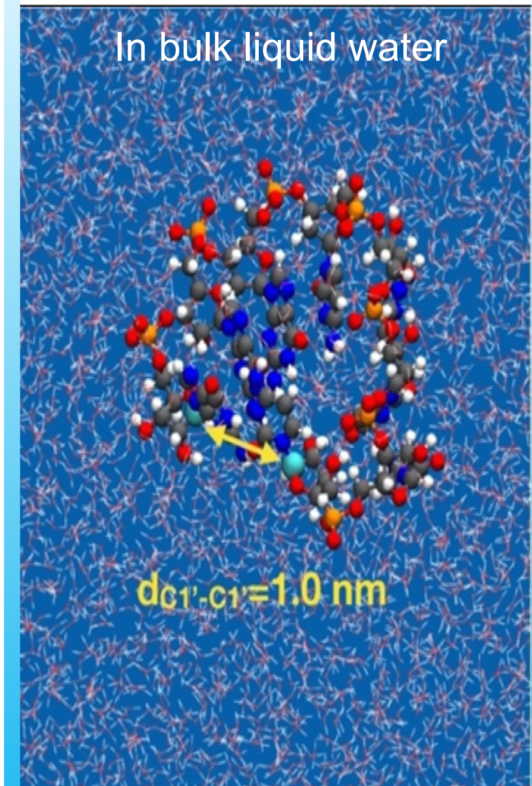
“Ice-solvated”, i.e., solvated by a quasi-liquid layer at the air/ice surface)

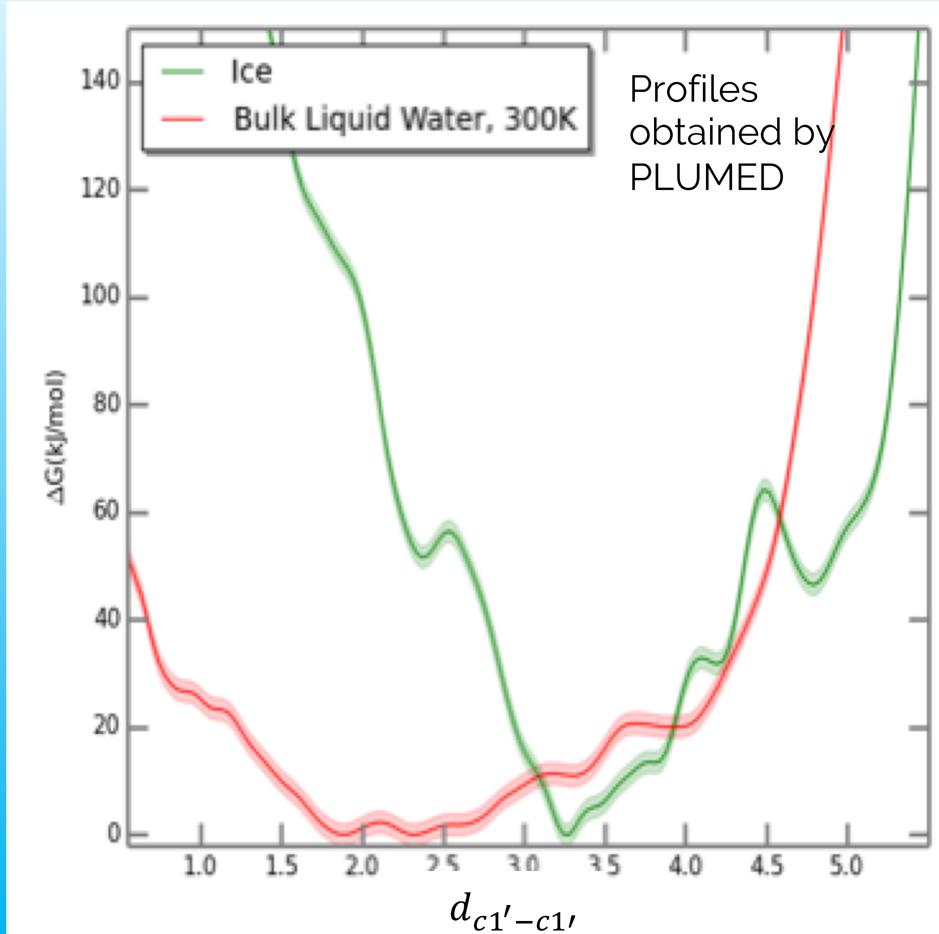
Comparisons possible with prior MD work on aqueous RNA by Bottaro et al, 2016.

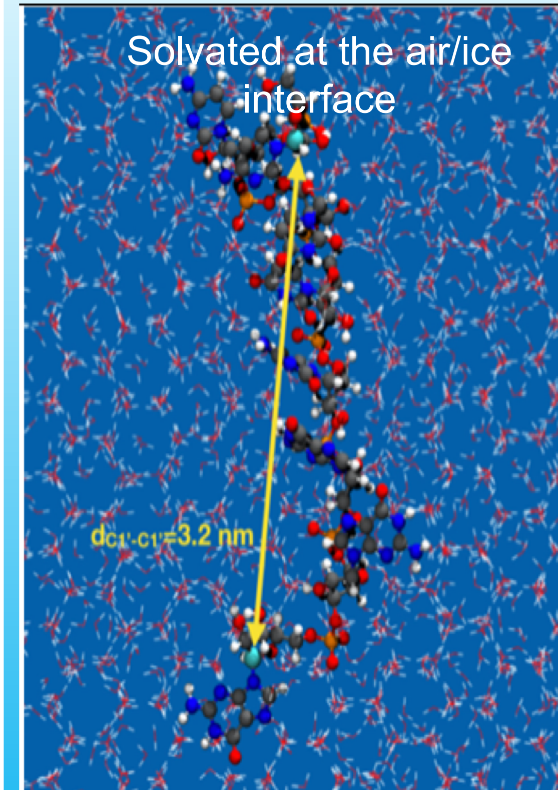


www.snowcrystals.com/faceting/faceting.html

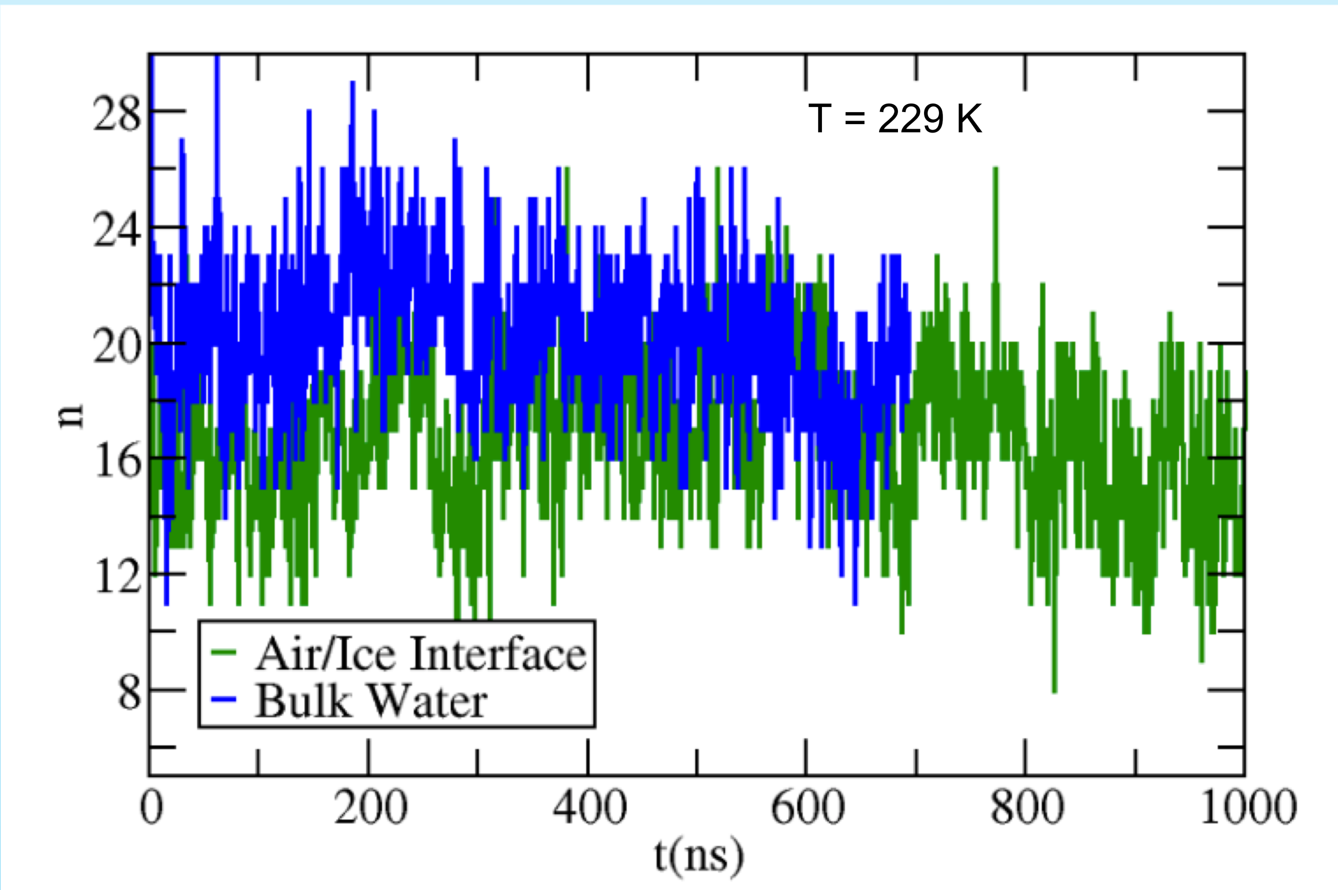
Thermodynamics: free energy profiles on the basal surface of ice exhibit a robust minimum at a much more extended

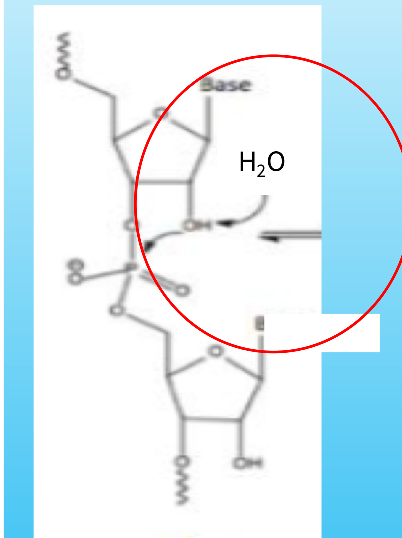




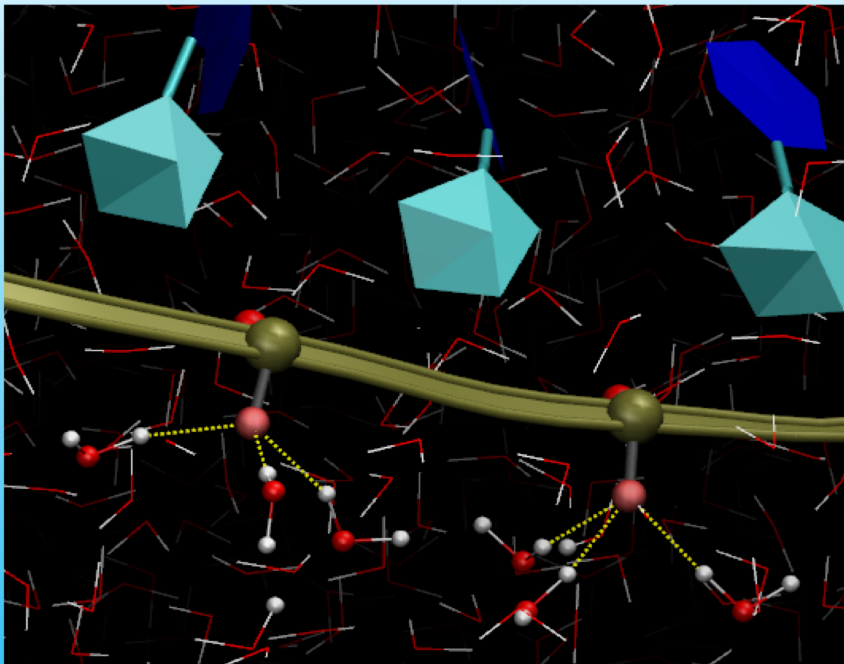


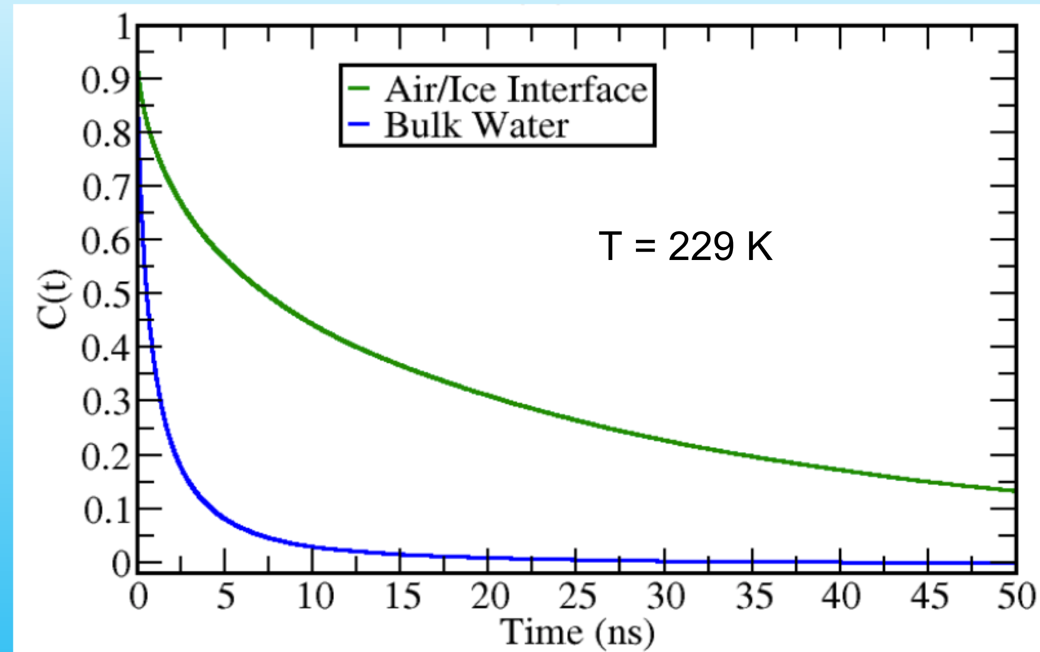
Precursor to hydrolysis: the rate of contact between $OH(2')$ & solvent indicates slightly lower susceptibility





Long-lived H-bonds between phosphodiester oxygens and solvent indicate much greater resilience to hydrolysis





Geometry of phosphodiester H-bonding to solvent is similar to that of aqueous phosphate [Chen et al, 2015; Moelbert et al, 2004; Sharma and Chandra, 2017; Dill et al, 2005], a potent kosmotrope.

Conclusions

- The air/ice interfacial environment has a distinctive impact on the orientation of surface-solvated single-strand RNA: bases turn toward the (hydrophobic) air/ice interface, while anionic phosphate oxygens align with the underlying ice lattice.

- The rate of contact between $OH(2')$ and solvent suggests that ice-solvated RNA is somewhat less susceptible to initiation of hydrolysis, compared to aqueous RNA at the same temperature.

- H-bond lifetimes of anionic phosphodiester oxygens suggest that hydrolysis ice-solvated RNA will be much less likely to complete (compared to aqueous RNA).

- These findings thus offer the possibility of a role for an ancient RNA world on ice distinct from that considered in extant elaborations of the RNA world hypothesis.

References

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