The Supplementary Information for

Structure making and breaking effects of ions on the anomalous diffusion of water revealed by machine learning potentials

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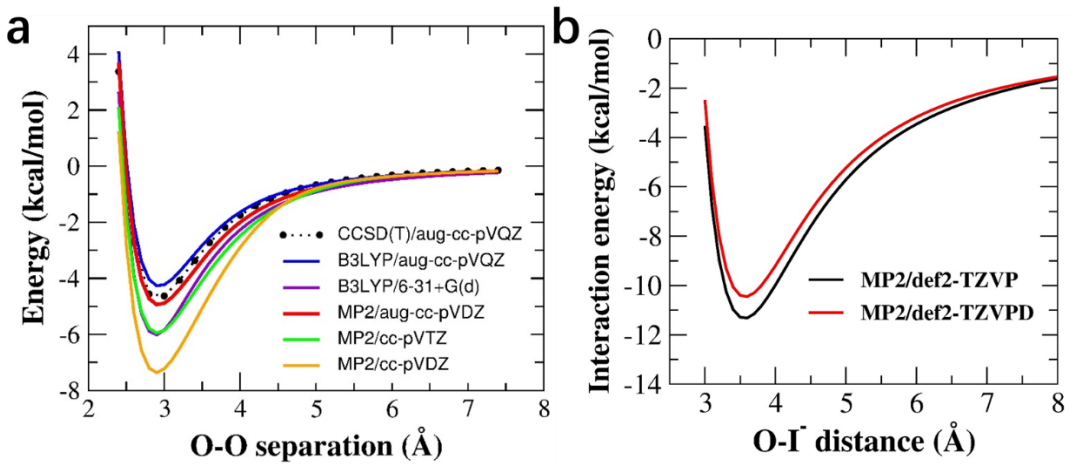
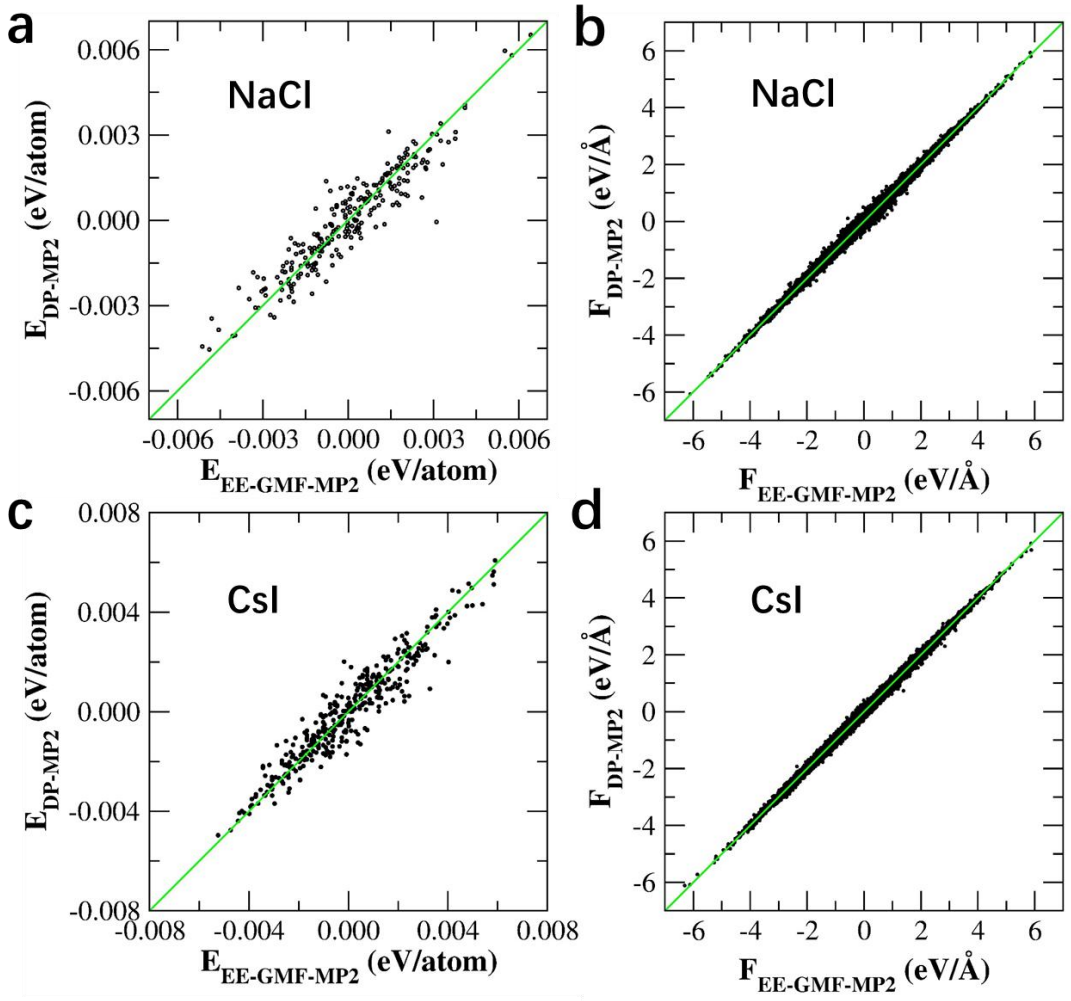
Supplementary Table 1. Classical and quantum predicted absolute self-diffusion coefficients (Å²/ps) of water in NaCl and CsI solutions across different salt concentrations using salt-specific DP-MP2 potentials.

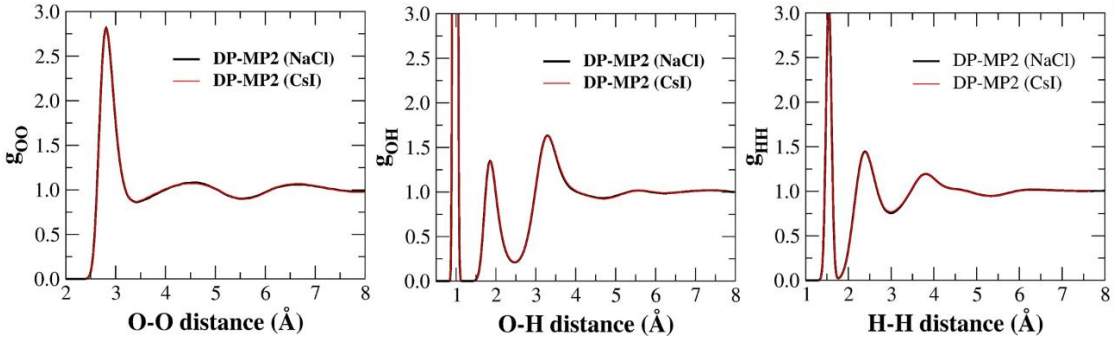
	NaCl solution		CsI solution	
	MD	CMD	MD	CMD
Pure water	0.224±0.005	0.241±0.016	0.214±0.004	0.245±0.006
1M	0.203±0.009	0.226±0.016	0.231±0.003	0.262±0.015
2M	0.178±0.006	0.204±0.012	0.244±0.006	0.284±0.018
3M	0.153±0.005	0.180±0.018	0.262±0.008	0.294±0.017



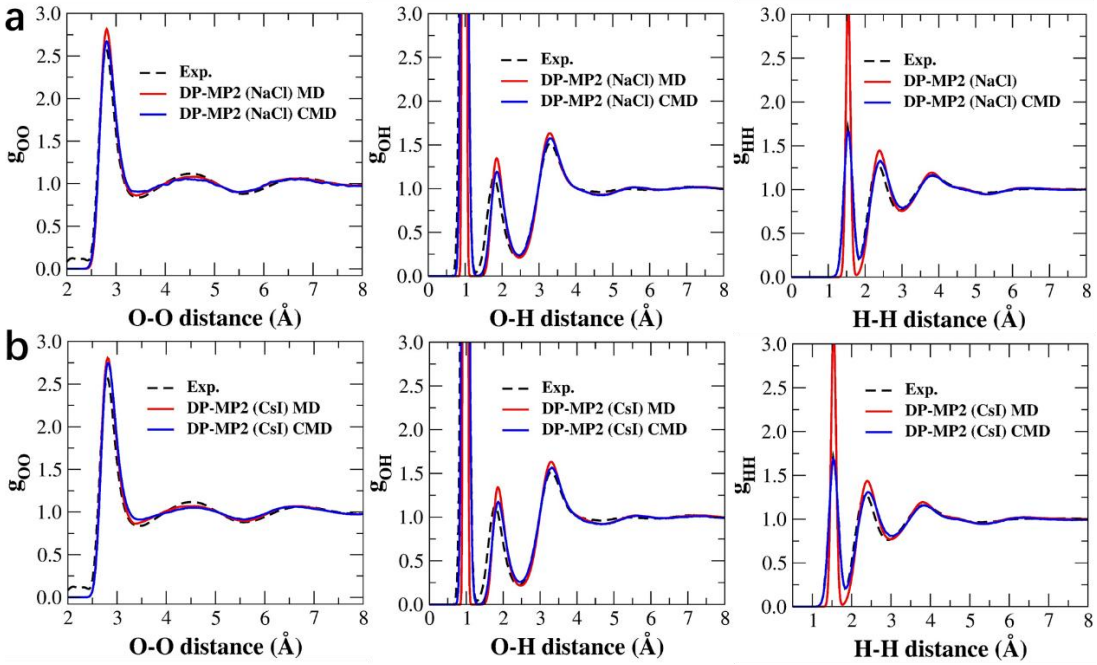
Supplementary Figure 1. a) Interaction potential energy curves between two water molecules calculated at different levels of theory. b) Interaction energy profiles between water molecule and iodide ion calculated by MP2 method using def2-TZVP and def2-TZVPD basis sets, respectively.



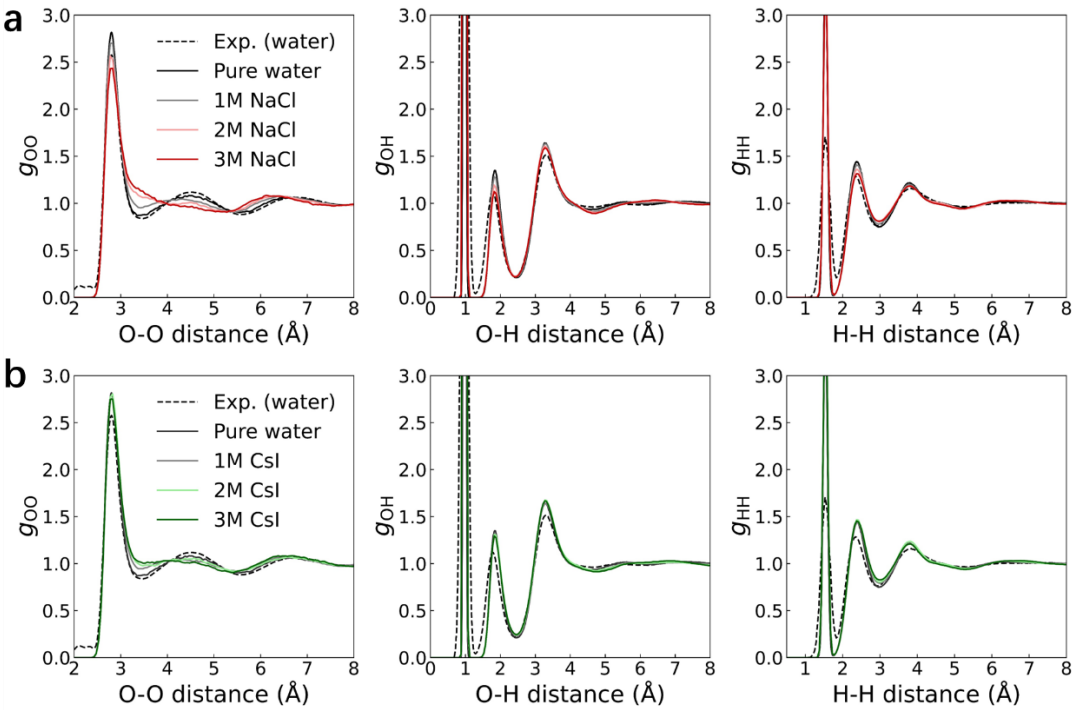
Supplementary Figure 2. The consistency between the predicted (a, c) energies and (b, d) atomic forces by the DP-MP2 potentials and the reference values calculated by EE-GMF fragmentation at the MP2 level of theory on the independent (a, b) NaCl and (c, d) CsI solution test sets, respectively.



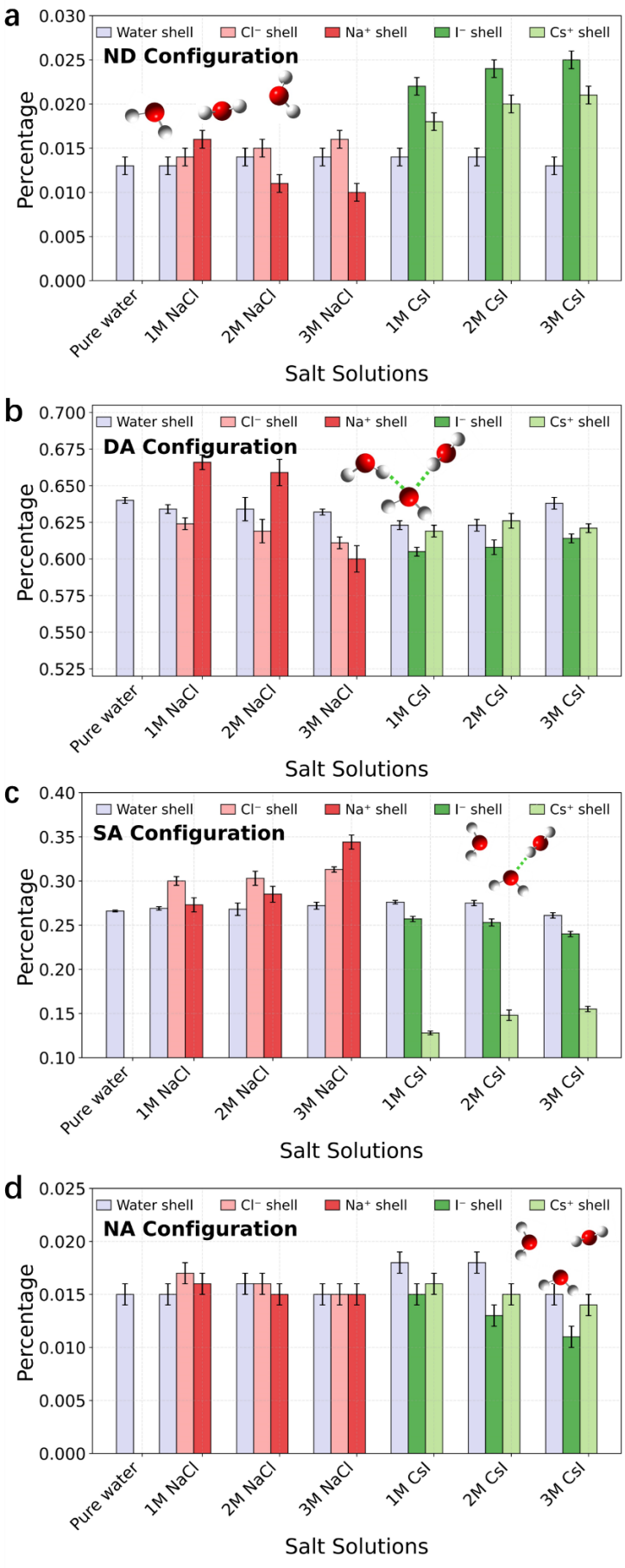
Supplementary Figure 3. The consistency of DP-MP2 (NaCl) and DP-MP2 (CsI) potentials in simulating the O-O, O-H, and H-H RDFs of liquid water from classical MD simulations.



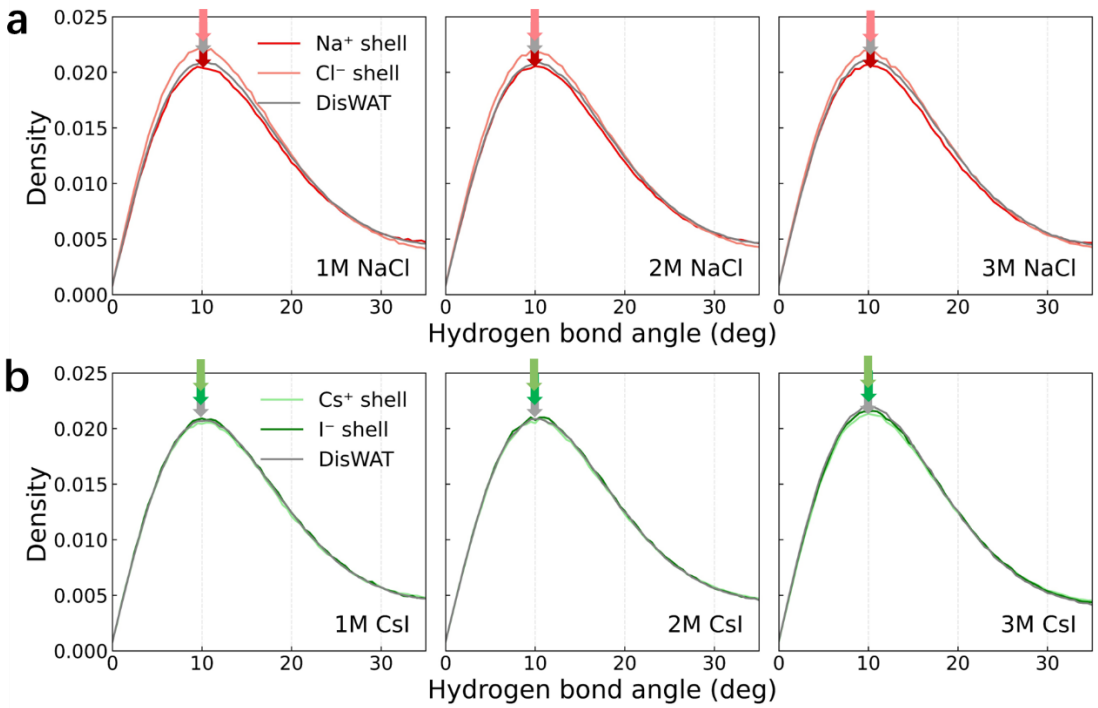
Supplementary Figure 4. The classical and quantum simulated O-O, O-H, and H-H RDFs of liquid water using (a) DP-MP2 (NaCl) and (b) DP-MP2 (CsI) potentials, respectively, with reference to the experimental data[1, 2].



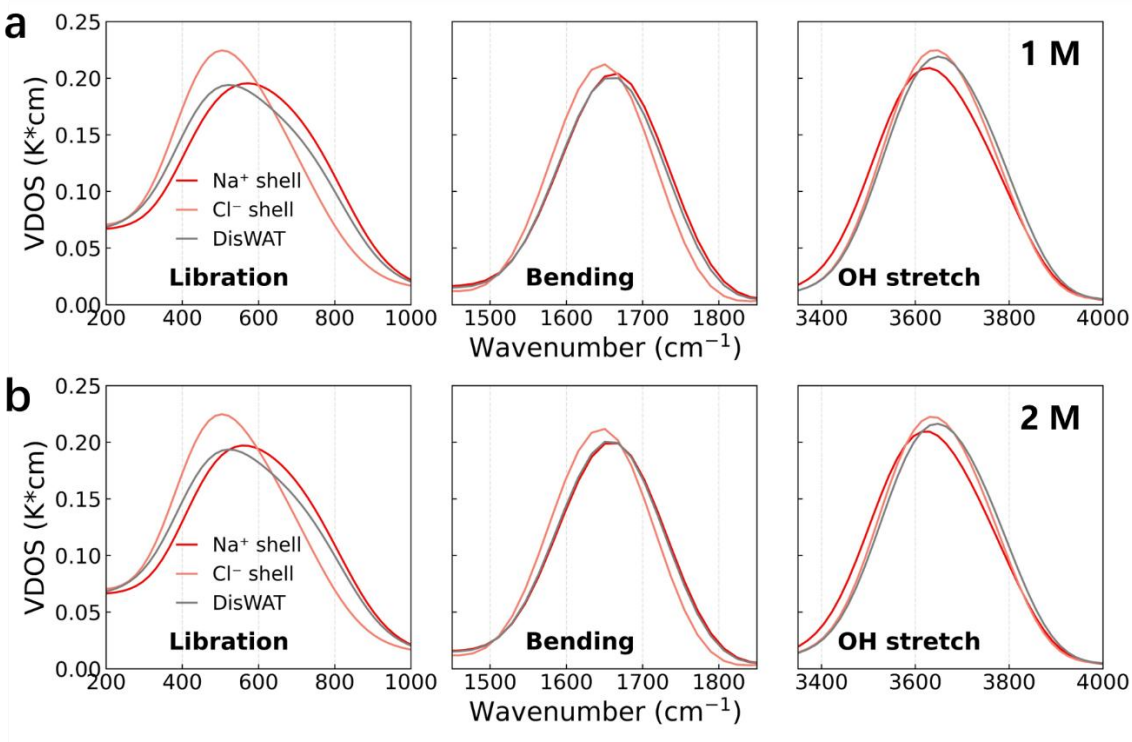
Supplementary Figure 5. EE-GMF-based AIMD simulated g_{OO}, g_{OH}, and g_{HH} RDFs of water in **a.** NaCl and **b.** CsI solutions across various salt concentrations. Each RDF was obtained from 10 ps AIMD simulation.


a
Water shell
Cl⁻ shell
Na⁺ shell
I⁻ shell
Cs⁺ shell
ND Configuration
Percentage
0.030
0.025
0.020
0.015
0.010
0.005
0.000
Pure water
1M NaCl
2M NaCl
3M NaCl
1M CsI
2M CsI
3M CsI
Salt Solutions
b
Water shell
Cl⁻ shell
Na⁺ shell
I⁻ shell
Cs⁺ shell
DA Configuration
Percentage
0.700
0.675
0.650
0.625
0.600
0.575
0.550
0.525
Pure water
1M NaCl
2M NaCl
3M NaCl
1M CsI
2M CsI
3M CsI
Salt Solutions
c
Water shell
Cl⁻ shell
Na⁺ shell
I⁻ shell
Cs⁺ shell
SA Configuration
Percentage
0.40
0.35
0.30
0.25
0.20
0.15
0.10
Pure water
1M NaCl
2M NaCl
3M NaCl
1M CsI
2M CsI
3M CsI
Salt Solutions
d
Water shell
Cl⁻ shell
Na⁺ shell
I⁻ shell
Cs⁺ shell
NA Configuration
Percentage
0.025
0.020
0.015
0.010
0.005
0.000
Pure water
1M NaCl
2M NaCl
3M NaCl
1M CsI
2M CsI
3M CsI
Salt Solutions

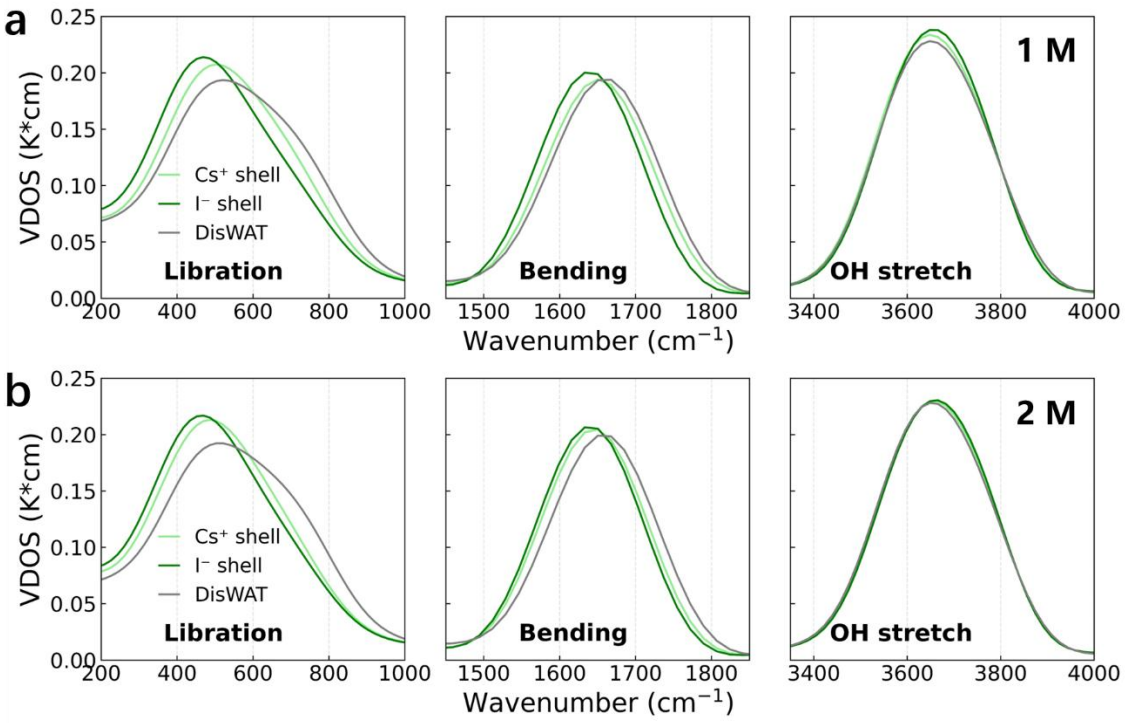
Supplementary Figure 6. Comparison of percentage of hydrogen bond non-donor (ND), double acceptor (DA), single acceptor (SA), and non-acceptor (NA) configurations in pure water and ionic solutions. All of the results were obtained from classical simulations using the salt-specific DP-MP2 potentials.



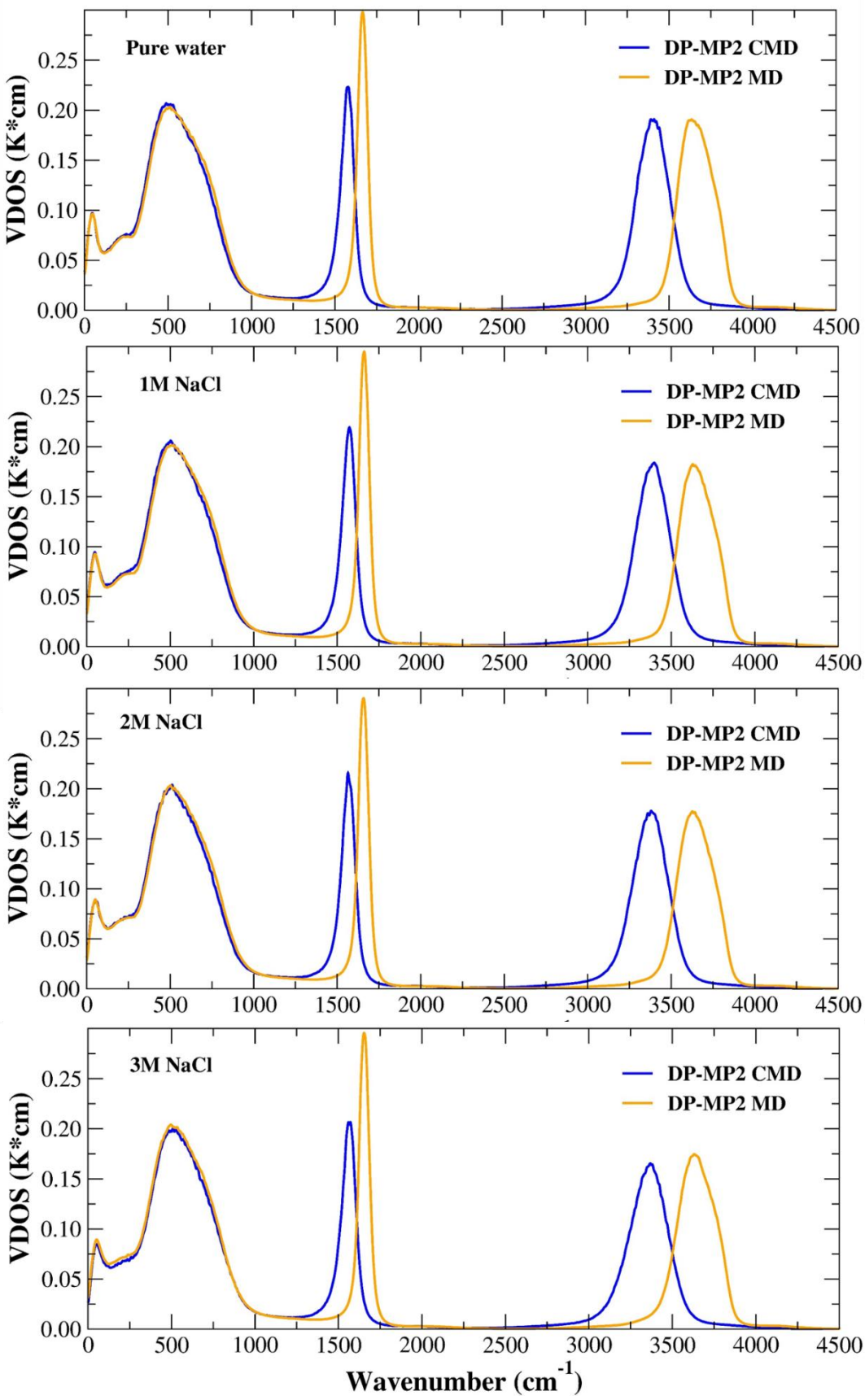
Supplementary Figure 7. Classical simulated distribution of water hydrogen bond angles for ion-hydrated and non-hydrated water molecules in NaCl and CsI solutions across various salt concentrations by using the salt-specific DP-MP2 potentials.



Supplementary Figure 8. Classical simulated vibrational density of states of ion-hydrated and non-hydrated water molecules in 1 M and 2 M NaCl solutions by using the salt-specific DP-MP2 potentials.



Supplementary Figure 9. Classical simulated vibrational density of states of ion-hydrated and non-hydrated water molecules in 1 M and 2 M CsI solutions by using the salt-specific DP-MP2 potentials.



Supplementary Figure 10. Calculated vibrational density of states of water in NaCl

solutions across various salt concentrations from the DP-MP2 simulations.

References:

1. Skinner, L.B. et al. Benchmark oxygen-oxygen pair-distribution function of ambient water from x-ray diffraction measurements with a wide Q-range. *J. Chem. Phys.* **138**, 074506 (2013).
2. Soper, A.K. & Benmore, C.J. Quantum differences between heavy and light water. *Phys. Rev. Lett.* **101**, 065502 (2008).