Supporting Information:

Individual Ion Activity Coefficients in Aqueous Electrolytes from Explicit-Water Molecular Dynamics Simulations

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I. Supplementary Figures

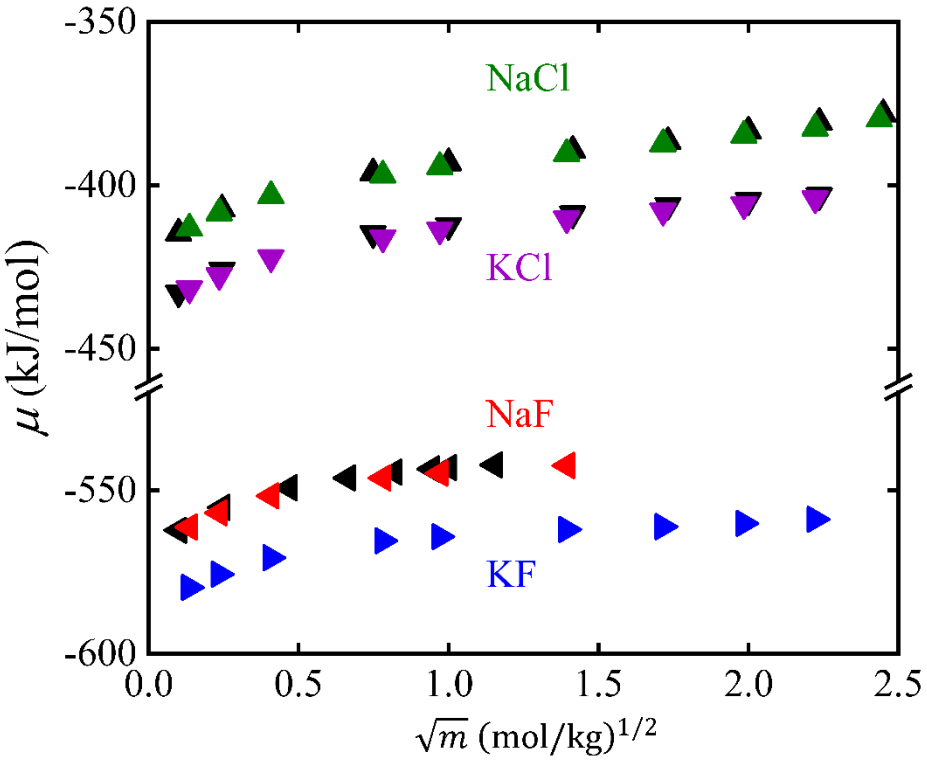

Figure S1. Simulation results for the total chemical potentials, μ (kJ/mol), in aqueous salt solutions at 298.15 K and 1 bar versus $\sqrt{m}$, where m is the molality (mol salt / kg H_2O). Symbols denote the salt solution, NaCl (▲), KCl (▼), NaF (◀), and KF (▶). Black triangles denote simulation results from Mester and Panagiotopoulos[1, 2]. Uncertainties are smaller than symbol size.

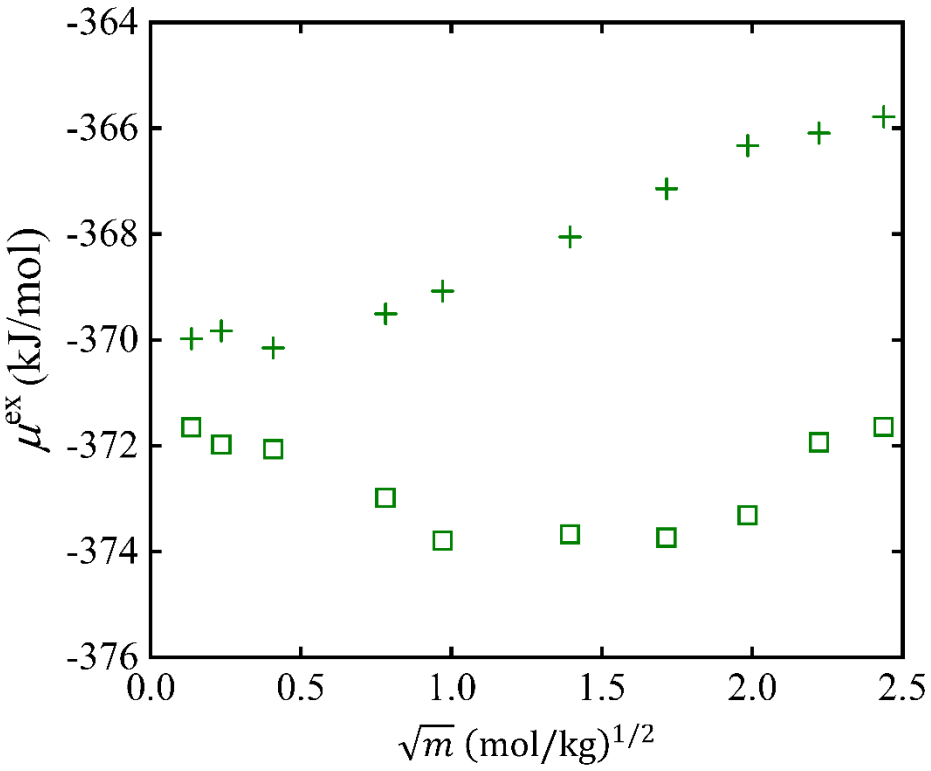

Figure S2. Simulation results for the excess chemical potentials, μ^{ex} (kJ/mol), of adding a single Na^+ (denoted by symbol +) or Cl^- (denoted by symbol □) into aqueous NaCl solutions at 298.15 K and 1 bar versus $\sqrt{m}$, where m is the molality (mol salt / kg H_2O).

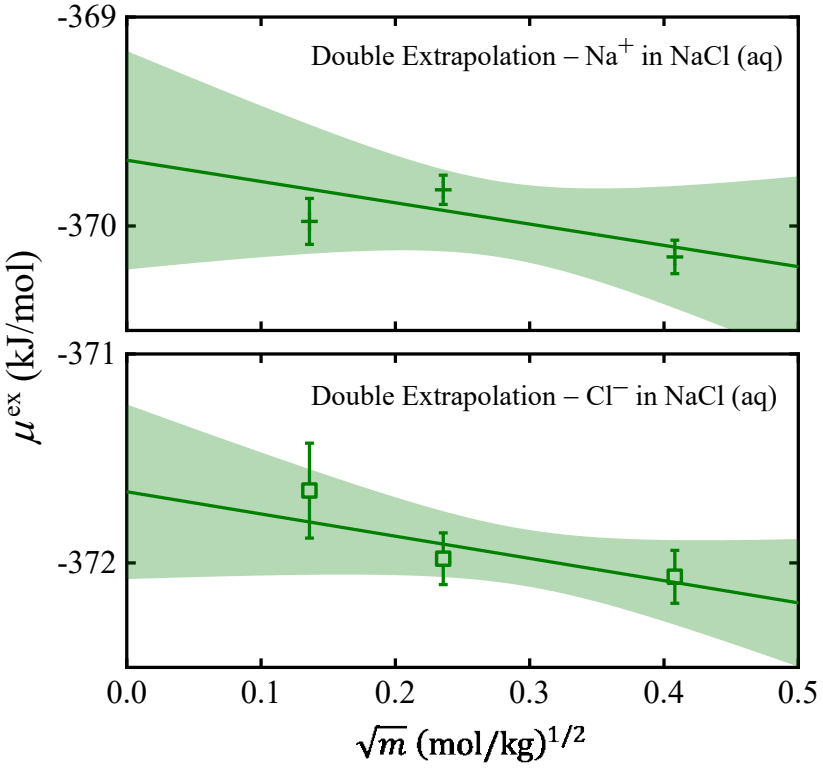

Figure S3. Simulation results extrapolated to infinite dilution for the excess chemical potentials, μ^{ex}, at three lowest concentrations, 0.019 m, 0.056 m, and 0.167 m. Top panel for adding a single Na^+ and bottom panel for adding a single Cl^- into aqueous NaCl solutions. Solid lines represent linear fits and shadings denote one standard deviation for the fitted lines. The error bar for each symbol denotes one standard deviation from extrapolation to infinite system size at each concentration.

II. Tables

Table S1. Model parameters for Joung and Cheatham[3] and SPC/E water[4].

Atom	ϵ (kJ/mol)	σ (nm)	Charge
Na^+	1.4755	0.2159	+1.0
K^+	1.7978	0.2838	+1.0
F^-	0.0310	0.4021	–1.0
Cl^-	0.0535	0.4830	–1.0
O	0.6502	0.3165	–0.8476
H	-	-	0.4238
Geometry of SPC/E water[4]			
$R_{OH} = 0.1$ nm		$\angle HOH = 109.47°$	

Aqueous NaCl Solution:

Table S2. Chemical potentials and extrapolation to infinite system size for added Na^+ or Cl^- in aqueous NaCl solutions at m = 0.019 mol salt / kg H_2O.

	N_{salt}	N_{water}	$\mu^{ex,LJ}$ (kJ/mol)	$\mu^{ex,Coul}$ (kJ/mol)	μ^{ex} (kJ/mol)	μ^{IG} (kJ/mol)	μ (kJ/mol)
Na^+	0	1500	2.22 ± 0.03	-372.98 ± 0.05	-370.76 ± 0.06	572.38	201.62 ± 0.06
	1	4500	2.19 ± 0.02	-372.67 ± 0.03	-370.48 ± 0.04	572.38	201.90 ± 0.04
	2	7500	2.16 ± 0.01	-372.60 ± 0.03	-370.44 ± 0.03	572.38	201.94 ± 0.03
		∞			-369.98 ± 0.11		202.40 ± 0.11
Cl^-	0	1500	27.58 ± 0.03	-400.20 ± 0.15	-372.62 ± 0.15	-242.11	-614.73 ± 0.15
	1	4500	27.64 ± 0.03	-399.94 ± 0.02	-372.30 ± 0.04	-242.10	-614.40 ± 0.04
	2	7500	27.68 ± 0.02	-399.89 ± 0.04	-372.21 ± 0.04	-242.10	-614.31 ± 0.04
		∞			-371.65 ± 0.23		-613.76 ± 0.23

Table S3. Chemical potentials and extrapolation to infinite system size for added Na^+ or Cl^- in aqueous NaCl solutions at m = 0.056 mol salt / kg H_2O.

	N_{salt}	N_{water}	$\mu^{ex,LJ}$ (kJ/mol)	$\mu^{ex,Coul}$ (kJ/mol)	μ^{ex} (kJ/mol)	μ^{IG} (kJ/mol)	μ (kJ/mol)
Na^+	0	500	2.19 ± 0.04	-373.40 ± 0.02	-371.22 ± 0.04	575.10	203.88 ± 0.04
	1	1500	2.18 ± 0.01	-373.00 ± 0.03	-370.82 ± 0.03	575.10	204.28 ± 0.03
	2	2500	2.17 ± 0.01	-372.80 ± 0.03	-370.63 ± 0.03	575.10	204.47 ± 0.03
	5	5500	2.20 ± 0.01	-372.66 ± 0.04	-370.46 ± 0.04	575.10	204.64 ± 0.04
		∞			-369.83 ± 0.07		205.28 ± 0.07
Cl^-	0	500	27.89 ± 0.07	-400.82 ± 0.06	-372.93 ± 0.09	-239.39	-612.32 ± 0.09
	1	1500	27.69 ± 0.05	-400.37 ± 0.05	-372.68 ± 0.07	-239.38	-612.06 ± 0.07
	2	2500	27.71 ± 0.04	-400.30 ± 0.04	-372.59 ± 0.06	-239.38	-611.97 ± 0.06
	5	5500	27.75 ± 0.05	-400.14 ± 0.03	-372.39 ± 0.06	-239.38	-611.77 ± 0.06
		∞			-371.98 ± 0.12		-611.36 ± 0.12

Table S4. Chemical potentials and extrapolation to infinite system size for added Na^+ or Cl^- in aqueous NaCl solutions at m = 0.167 mol salt / kg H_2O.

	N_{salt}	N_{water}	$\mu^{ex,LJ}$ (kJ/mol)	$\mu^{ex,Coul}$ (kJ/mol)	μ^{ex} (kJ/mol)	μ^{IG} (kJ/mol)	μ (kJ/mol)
Na^+	1	500	2.24 ± 0.02	-373.26 ± 0.07	-371.02 ± 0.07	577.83	206.81 ± 0.07
	4	1500	2.27 ± 0.01	-372.84 ± 0.04	-370.59 ± 0.04	577.82	207.25 ± 0.05
	7	2500	2.25 ± 0.01	-372.84 ± 0.04	-370.59 ± 0.04	577.82	207.23 ± 0.04
	16	5500	2.27 ± 0.00	-372.81 ± 0.03	-370.54 ± 0.03	577.82	207.28 ± 0.03
		∞			-370.19 ± 0.08		207.63 ± 0.08
Cl^-	1	500	27.88 ± 0.06	-401.21 ± 0.08	-373.33 ± 0.10	-236.66	-609.99 ± 0.10
	4	1500	27.80 ± 0.06	-400.86 ± 0.06	-373.06 ± 0.08	-236.66	-609.72 ± 0.08
	7	2500	27.92 ± 0.03	-400.76 ± 0.05	-372.84 ± 0.06	-236.66	-609.50 ± 0.06
	16	5500	27.96 ± 0.04	-400.61 ± 0.04	-372.65 ± 0.06	-236.66	-609.31 ± 0.06
		∞			-372.09 ± 0.13		-608.75 ± 0.13

Table S5. Chemical potentials and extrapolation to infinite system size for added Na^+ or Cl^- in aqueous NaCl solutions at m = 0.611 mol salt / kg H_2O.

	N_{salt}	N_{water}	$\mu^{ex,LJ}$ (kJ/mol)	$\mu^{ex,Coul}$ (kJ/mol)	μ^{ex} (kJ/mol)	μ^{IG} (kJ/mol)	μ (kJ/mol)
Na^+	5	500	2.47 ± 0.02	-372.97 ± 0.08	-370.50 ± 0.08	581.03	210.53 ± 0.08
	16	1500	2.53 ± 0.02	-372.53 ± 0.06	-370.00 ± 0.06	581.03	211.03 ± 0.06
	27	2500	2.54 ± 0.01	-372.50 ± 0.05	-369.96 ± 0.05	581.03	211.07 ± 0.05
	60	5500	2.50 ± 0.01	-372.43 ± 0.03	-369.93 ± 0.03	581.03	211.10 ± 0.03
		∞			-369.50 ± 0.09		211.53 ± 0.09
Cl^-	5	500	28.41 ± 0.06	-402.38 ± 0.10	-373.97 ± 0.12	-233.46	-607.43 ± 0.12
	16	1500	28.48 ± 0.06	-402.00 ± 0.09	-373.52 ± 0.11	-233.46	-606.98 ± 0.11
	27	2500	28.48 ± 0.03	-402.08 ± 0.08	-373.60 ± 0.09	-233.46	-607.06 ± 0.09
	60	5500	28.61 ± 0.05	-402.02 ± 0.02	-373.41 ± 0.05	-233.46	-606.87 ± 0.05
		∞			-372.98 ± 0.12		-606.44 ± 0.13

Table S6. Chemical potentials and extrapolation to infinite system size for added Na^+ or Cl^- in aqueous NaCl solutions at m = 0.944 mol salt / kg H_2O.

	N_{salt}	N_{water}	$\mu^{\mathrm{ex,LJ}}$ (kJ/mol)	$\mu^{\mathrm{ex,Coul}}$ (kJ/mol)	μ^{ex} (kJ/mol)	μ^{IG} (kJ/mol)	μ (kJ/mol)
Na^+	8	500	2.62 ± 0.04	-372.71 ± 0.10	-370.09 ± 0.11	582.10	212.01 ± 0.11
	25	1500	2.67 ± 0.01	-372.26 ± 0.03	-369.59 ± 0.03	582.10	212.51 ± 0.03
	42	2500	2.67 ± 0.01	-372.29 ± 0.03	-369.62 ± 0.03	582.10	212.48 ± 0.03
	93	5500	2.68 ± 0.02	-372.10 ± 0.07	-369.42 ± 0.07	582.10	212.68 ± 0.07
		∞			-369.07 ± 0.13		213.02 ± 0.13
Cl^-	8	500	29.25 ± 0.07	-403.15 ± 0.06	-373.90 ± 0.09	-232.40	-606.30 ± 0.09
	25	1500	28.93 ± 0.06	-402.89 ± 0.04	-373.96 ± 0.07	-232.39	-606.35 ± 0.07
	42	2500	28.95 ± 0.05	-402.90 ± 0.09	-373.95 ± 0.10	-232.39	-606.34 ± 0.10
	93	5500	29.00 ± 0.04	-402.81 ± 0.06	-373.81 ± 0.07	-232.39	-606.20 ± 0.07
		∞			-373.79 ± 0.14		-606.19 ± 0.14

Table S7. Chemical potentials and extrapolation to infinite system size for added Na^+ or Cl^- in aqueous NaCl solutions at m = 1.943 mol salt / kg H_2O.

	N_{salt}	N_{water}	$\mu^{\mathrm{ex,LJ}}$ (kJ/mol)	$\mu^{\mathrm{ex,Coul}}$ (kJ/mol)	μ^{ex} (kJ/mol)	μ^{IG} (kJ/mol)	μ (kJ/mol)
Na^+	17	500	3.22 ± 0.02	-371.98 ± 0.03	-368.76 ± 0.04	583.85	215.09 ± 0.04
	52	1500	3.23 ± 0.02	-371.60 ± 0.06	-368.37 ± 0.06	583.84	215.47 ± 0.06
	87	2500	3.22 ± 0.03	-371.69 ± 0.09	-368.47 ± 0.09	583.84	215.37 ± 0.09
	192	5500	3.25 ± 0.02	-371.67 ± 0.05	-368.42 ± 0.05	583.84	215.42 ± 0.05
		∞			-368.05 ± 0.09		215.79 ± 0.09
Cl^-	17	500	30.34 ± 0.09	-405.09 ± 0.05	-374.75 ± 0.10	-230.64	-605.39 ± 0.10
	52	1500	30.47 ± 0.07	-404.76 ± 0.03	-374.29 ± 0.08	-230.64	-604.93 ± 0.08
	87	2500	30.50 ± 0.03	-404.70 ± 0.03	-374.20 ± 0.04	-230.64	-604.84 ± 0.04
	192	5500	30.39 ± 0.07	-404.76 ± 0.08	-374.37 ± 0.11	-230.64	-605.01 ± 0.11
		∞			-373.68 ± 0.16		-604.32 ± 0.16

Table S8. Chemical potentials and extrapolation to infinite system size for added Na^+ or Cl^- in aqueous NaCl solutions at m = 2.942 mol salt / kg H_2O.

	N_{salt}	N_{water}	$\mu^{\mathrm{ex,LJ}}$ (kJ/mol)	$\mu^{\mathrm{ex,Coul}}$ (kJ/mol)	μ^{ex} (kJ/mol)	μ^{IG} (kJ/mol)	μ (kJ/mol)
Na^+	26	500	3.77 ± 0.02	-371.67 ± 0.09	-367.90 ± 0.09	584.83	216.93 ± 0.09
	79	1500	3.74 ± 0.01	-371.37 ± 0.10	-367.63 ± 0.10	584.83	217.20 ± 0.10
	132	2500	3.75 ± 0.01	-371.31 ± 0.07	-367.56 ± 0.07	584.83	217.27 ± 0.07
	291	5500	3.77 ± 0.02	-371.26 ± 0.06	-367.49 ± 0.06	584.83	217.34 ± 0.06
		∞			-367.14 ± 0.13		217.69 ± 0.13
Cl^-	26	500	31.87 ± 0.14	-406.42 ± 0.12	-374.55 ± 0.18	-229.66	-604.21 ± 0.18
	79	1500	31.77 ± 0.12	-405.95 ± 0.12	-374.18 ± 0.17	-229.66	-603.84 ± 0.17
	132	2500	31.75 ± 0.11	-405.96 ± 0.07	-374.21 ± 0.13	-229.66	-603.87 ± 0.13
	291	5500	31.88 ± 0.03	-405.97 ± 0.05	-374.09 ± 0.06	-229.66	-603.75 ± 0.06
		∞			-373.74 ± 0.18		-603.40 ± 0.18

Table S9. Chemical potentials and extrapolation to infinite system size for added Na^+ or Cl^- in aqueous NaCl solutions at m = 3.941 mol salt / kg H_2O.

	N_{salt}	N_{water}	$\mu^{\mathrm{ex,LJ}}$ (kJ/mol)	$\mu^{\mathrm{ex,Coul}}$ (kJ/mol)	μ^{ex} (kJ/mol)	μ^{IG} (kJ/mol)	μ (kJ/mol)
Na^+	35	500	4.33 ± 0.02	-371.62 ± 0.10	-367.29 ± 0.12	585.50	218.21 ± 0.12
	106	1500	4.30 ± 0.01	-371.42 ± 0.09	-367.12 ± 0.10	585.50	218.38 ± 0.10
	177	2500	4.31 ± 0.01	-371.16 ± 0.10	-366.85 ± 0.11	585.50	218.65 ± 0.11
	390	5500	4.30 ± 0.02	-371.06 ± 0.11	-366.76 ± 0.13	585.50	218.74 ± 0.13
		∞			-366.33 ± 0.21		219.17 ± 0.21
Cl^-	35	500	33.01 ± 0.06	-406.92 ± 0.10	-373.91 ± 0.12	-228.98	-602.89 ± 0.12
	106	1500	33.17 ± 0.18	-406.86 ± 0.06	-373.69 ± 0.19	-228.98	-602.67 ± 0.19
	177	2500	33.07 ± 0.10	-406.74 ± 0.05	-373.67 ± 0.11	-228.98	-602.65 ± 0.11
	390	5500	33.08 ± 0.08	-406.66 ± 0.07	-373.58 ± 0.11	-228.98	-602.56 ± 0.11
		∞			-373.31 ± 0.19		-602.30 ± 0.19

Table S10. Chemical potentials and extrapolation to infinite system size for added Na^+ or Cl^- in aqueous NaCl solutions at m = 4.940 mol salt / kg H_2O.

	N_{salt}	N_{water}	$\mu^{\mathrm{ex,LJ}}$ (kJ/mol)	$\mu^{\mathrm{ex,Coul}}$ (kJ/mol)	μ^{ex} (kJ/mol)	μ^{IG} (kJ/mol)	μ (kJ/mol)
Na^+	44	500	4.80 ± 0.02	-371.65 ± 0.07	-366.85 ± 0.09	586.01	219.16 ± 0.09
	133	1500	4.79 ± 0.02	-371.35 ± 0.06	-366.56 ± 0.08	586.01	219.45 ± 0.08
	222	2500	4.79 ± 0.02	-371.39 ± 0.10	-366.60 ± 0.12	586.01	219.41 ± 0.12
	489	5500	4.76 ± 0.02	-371.23 ± 0.08	-366.47 ± 0.10	586.01	219.54 ± 0.10
		∞			-366.14 ± 0.17		219.87 ± 0.17
Cl^-	44	500	34.10 ± 0.16	-407.29 ± 0.15	-373.19 ± 0.22	-228.48	-601.67 ± 0.22
	133	1500	34.49 ± 0.11	-406.93 ± 0.11	-372.44 ± 0.16	-228.48	-600.92 ± 0.16
	222	2500	34.37 ± 0.04	-407.00 ± 0.06	-372.63 ± 0.07	-228.48	-601.11 ± 0.07
	489	5500	34.38 ± 0.08	-406.89 ± 0.07	-372.51 ± 0.11	-228.48	-600.99 ± 0.11
		∞			-372.05 ± 0.25		-600.52 ± 0.25

Table S11. Chemical potentials and extrapolation to infinite system size for added Na^+ or Cl^- in aqueous NaCl solutions at m = 5.939 mol salt / kg H_2O.

	N_{salt}	N_{water}	$\mu^{\mathrm{ex,LJ}}$ (kJ/mol)	$\mu^{\mathrm{ex,Coul}}$ (kJ/mol)	μ^{ex} (kJ/mol)	μ^{IG} (kJ/mol)	μ (kJ/mol)
Na^+	53	500	5.26 ± 0.05	-371.78 ± 0.07	-366.52 ± 0.09	586.41	219.89 ± 0.09
	160	1500	5.23 ± 0.04	-371.43 ± 0.10	-366.20 ± 0.11	586.41	220.21 ± 0.11
	267	2500	5.25 ± 0.04	-371.54 ± 0.12	-366.29 ± 0.13	586.41	220.12 ± 0.13
	588	5500	5.25 ± 0.02	-371.42 ± 0.10	-366.17 ± 0.10	586.41	220.24 ± 0.10
		∞			-365.86 ± 0.17		220.55 ± 0.17
Cl^-	53	500	35.35 ± 0.14	-406.92 ± 0.31	-371.57 ± 0.34	-228.08	-599.65 ± 0.34
	160	1500	35.45 ± 0.08	-406.88 ± 0.20	-371.43 ± 0.22	-228.07	-599.50 ± 0.22
	267	2500	35.22 ± 0.10	-406.81 ± 0.09	-371.59 ± 0.13	-228.07	-599.66 ± 0.13
	588	5500	35.42 ± 0.10	-406.60 ± 0.07	-371.18 ± 0.12	-228.07	-599.25 ± 0.12
		∞			-370.87± 0.34		-598.95 ± 0.34

Aqueous KCl Solution:

Table S12. Chemical potentials and extrapolation to infinite system size for added K^+ or Cl^- in aqueous KCl solutions at m = 0.019 mol salt / kg H_2O.

	N_{salt}	N_{water}	$\mu^{ex,LJ}$ (kJ/mol)	$\mu^{ex,Coul}$ (kJ/mol)	μ^{ex} (kJ/mol)	μ^{IG} (kJ/mol)	μ (kJ/mol)
K^+	0	1500	3.05 ± 0.03	-299.05 ± 0.04	-296.00 ± 0.05	479.01	183.01 ± 0.05
	1	4500	3.08 ± 0.03	-298.72 ± 0.04	-295.64 ± 0.05	479.01	183.37 ± 0.05
	2	7500	3.11 ± 0.01	-298.65 ± 0.02	-295.54 ± 0.02	479.01	183.47 ± 0.02
		∞			-294.88 ± 0.22		184.13 ± 0.22
Cl^-	0	1500	27.66 ± 0.04	-400.21 ± 0.05	-372.55 ± 0.06	-242.10	-614.65 ± 0.06
	1	4500	27.64 ± 0.06	-399.86 ± 0.03	-372.22 ± 0.07	-242.10	-614.32 ± 0.07
	2	7500	27.62 ± 0.02	-399.82 ± 0.04	-372.20 ± 0.04	-242.10	-614.30 ± 0.04
		∞			-371.68 ± 0.14		-613.79 ± 0.14

Table S13. Chemical potentials and extrapolation to infinite system size for added K^+ or Cl^- in aqueous KCl solutions at m = 0.056 mol salt / kg H_2O.

	N_{salt}	N_{water}	$\mu^{ex,LJ}$ (kJ/mol)	$\mu^{ex,Coul}$ (kJ/mol)	μ^{ex} (kJ/mol)	μ^{IG} (kJ/mol)	μ (kJ/mol)
K^+	0	500	3.07 ± 0.03	-299.73 ± 0.02	-296.66 ± 0.04	481.73	185.07 ± 0.04
	1	1500	3.05 ± 0.04	-299.10 ± 0.01	-296.05 ± 0.04	481.73	185.68 ± 0.04
	2	2500	3.08 ± 0.02	-299.09 ± 0.03	-296.01 ± 0.04	481.73	185.72 ± 0.04
	5	5500	3.12 ± 0.01	-298.92 ± 0.04	-295.80 ± 0.04	481.73	185.93 ± 0.04
		∞			-295.07 ± 0.07		186.67 ± 0.07
Cl^-	0	500	27.64 ± 0.07	-400.90 ± 0.07	-373.26 ± 0.10	-239.38	-612.64 ± 0.10
	1	1500	27.70 ± 0.05	-400.22 ± 0.05	-372.52 ± 0.07	-239.38	-611.90 ± 0.07
	2	2500	27.73 ± 0.07	-400.21 ± 0.06	-372.48 ± 0.09	-239.38	-611.86 ± 0.09
	5	5500	27.73 ± 0.03	-400.02 ± 0.05	-372.29 ± 0.06	-239.38	-611.67 ± 0.06
		∞			-371.50 ± 0.13		-610.89 ± 0.13

Table S14. Chemical potentials and extrapolation to infinite system size for added K^+ or Cl^- in aqueous KCl solutions at m = 0.167 mol salt / kg H_2O.

	N_{salt}	N_{water}	$\mu^{ex,LJ}$ (kJ/mol)	$\mu^{ex,Coul}$ (kJ/mol)	μ^{ex} (kJ/mol)	μ^{IG} (kJ/mol)	μ (kJ/mol)
K^+	1	500	3.15 ± 0.03	-299.94 ± 0.06	-296.79 ± 0.07	484.45	187.66 ± 0.07
	4	1500	3.21 ± 0.03	-299.54 ± 0.03	-296.33 ± 0.04	484.45	188.12 ± 0.04
	7	2500	3.22 ± 0.03	-299.41 ± 0.07	-296.19 ± 0.08	484.45	188.26 ± 0.08
	16	5500	3.19 ± 0.03	-299.24 ± 0.05	-296.05 ± 0.06	484.45	188.40 ± 0.06
		∞			-295.42 ± 0.11		189.03 ± 0.11
Cl^-	1	500	27.83 ± 0.07	-400.92 ± 0.07	-373.09 ± 0.10	-236.66	-609.75 ± 0.10
	4	1500	27.80 ± 0.04	-400.30 ± 0.04	-372.50 ± 0.06	-236.66	-609.16 ± 0.06
	7	2500	27.84 ± 0.02	-400.30 ± 0.07	-372.46 ± 0.07	-236.66	-609.12 ± 0.07
	16	5500	27.91 ± 0.04	-400.20 ± 0.04	-372.29 ± 0.06	-236.66	-608.95 ± 0.06
		∞			-371.66 ± 0.13		-608.32 ± 0.13

Table S15. Chemical potentials and extrapolation to infinite system size for added K^+ or Cl^- in aqueous KCl solutions at m = 0.611 mol salt / kg H_2O.

	N_{salt}	N_{water}	$\mu^{ex,LJ}$ (kJ/mol)	$\mu^{ex,Coul}$ (kJ/mol)	μ^{ex} (kJ/mol)	μ^{IG} (kJ/mol)	μ (kJ/mol)
K^+	5	500	3.49 ± 0.01	-301.04 ± 0.04	-297.55 ± 0.04	487.65	190.10 ± 0.04
	16	1500	3.49 ± 0.01	-300.68 ± 0.05	-297.19 ± 0.05	487.64	190.45 ± 0.05
	27	2500	3.57 ± 0.02	-300.64 ± 0.06	-297.07 ± 0.06	487.64	190.57 ± 0.06
	60	5500	3.52 ± 0.03	-300.52 ± 0.06	-297.00 ± 0.07	487.64	190.64 ± 0.07
		∞			-296.47 ± 0.10		191.17 ± 0.10
Cl^-	5	500	28.25 ± 0.07	-400.90 ± 0.07	-372.65 ± 0.10	-233.47	-606.12 ± 0.10
	16	1500	28.45 ± 0.10	-400.72 ± 0.05	-372.27 ± 0.11	-233.47	-605.74 ± 0.11
	27	2500	28.47 ± 0.04	-400.43 ± 0.06	-371.96 ± 0.07	-233.47	-605.43 ± 0.07
	60	5500	28.52 ± 0.07	-400.34 ± 0.05	-371.82 ± 0.09	-233.47	-605.29 ± 0.09
		∞			-371.09 ± 0.16		-604.56 ± 0.16

Table S16. Chemical potentials and extrapolation to infinite system size for added K^+ or Cl^- in aqueous KCl solutions at m = 0.944 mol salt / kg H_2O.

	N_{salt}	N_{water}	$\mu^{ex,LJ}$ (kJ/mol)	$\mu^{ex,Coul}$ (kJ/mol)	μ^{ex} (kJ/mol)	μ^{IG} (kJ/mol)	μ (kJ/mol)
K^+	8	500	3.73 ± 0.02	-301.88 ± 0.06	-298.15 ± 0.06	488.71	190.56 ± 0.06
	25	1500	3.80 ± 0.05	-301.56 ± 0.05	-297.76 ± 0.09	488.70	190.94 ± 0.07
	42	2500	3.73 ± 0.02	-301.48 ± 0.05	-297.75 ± 0.08	488.70	190.95 ± 0.05
	93	5500	3.79 ± 0.04	-301.43 ± 0.04	-297.64 ± 0.05	488.70	191.06 ± 0.06
		∞			-297.20 ± 0.10		191.50 ± 0.10
Cl^-	8	500	28.89 ± 0.04	-400.66 ± 0.05	-371.77 ± 0.06	-232.41	-604.18 ± 0.06
	25	1500	28.94 ± 0.04	-400.37 ± 0.08	-371.43 ± 0.09	-232.41	-603.84 ± 0.09
	42	2500	28.92 ± 0.03	-400.19 ± 0.07	-371.27 ± 0.08	-232.41	-603.68 ± 0.08
	93	5500	28.97 ± 0.04	-400.25 ± 0.03	-371.28 ± 0.05	-232.41	-603.69 ± 0.05
		∞			-370.83 ± 0.10		-603.24 ± 0.10

Table S17. Chemical potentials and extrapolation to infinite system size for added K^+ or Cl^- in aqueous KCl solutions at m =1.943 mol salt / kg H_2O.

	N_{salt}	N_{water}	$\mu^{ex,LJ}$ (kJ/mol)	$\mu^{ex,Coul}$ (kJ/mol)	μ^{ex} (kJ/mol)	μ^{IG} (kJ/mol)	μ (kJ/mol)
K^+	17	500	4.43 ± 0.02	-304.63 ± 0.09	-300.20 ± 0.09	490.43	190.23 ± 0.09
	52	1500	4.41 ± 0.06	-304.10 ± 0.08	-299.69 ± 0.10	490.43	190.74 ± 0.10
	87	2500	4.44 ± 0.02	-303.97 ± 0.03	-299.53 ± 0.04	490.43	190.90 ± 0.04
	192	5500	4.47 ± 0.02	-303.97 ± 0.05	-299.50 ± 0.05	490.43	190.93 ± 0.05
		∞			-298.84 ± 0.12		191.59 ± 0.12
Cl^-	17	500	30.15 ± 0.10	-399.93 ± 0.05	-369.78 ± 0.11	-230.69	-600.47 ± 0.11
	52	1500	30.16 ± 0.08	-399.73 ± 0.05	-369.57 ± 0.09	-230.69	-600.26 ± 0.09
	87	2500	30.16 ± 0.07	-399.57 ± 0.08	-369.41 ± 0.11	-230.69	-600.10 ± 0.11
	192	5500	30.14 ± 0.07	-399.55 ± 0.05	-369.41 ± 0.09	-230.69	-600.10 ± 0.09
		∞			-369.07 ± 0.17		-599.75 ± 0.17

Table S18. Chemical potentials and extrapolation to infinite system size for added K^+ or Cl^- in aqueous KCl solutions at m =2.942 mol salt / kg H_2O.

	N_{salt}	N_{water}	$\mu^{\mathrm{ex,LJ}}$ (kJ/mol)	$\mu^{\mathrm{ex,Coul}}$ (kJ/mol)	μ^{ex} (kJ/mol)	μ^{IG} (kJ/mol)	μ (kJ/mol)
K^+	26	500	5.08 ± 0.06	-307.00 ± 0.03	-301.92 ± 0.07	491.39	189.47 ± 0.07
	79	1500	5.09 ± 0.06	-306.67 ± 0.07	-301.58 ± 0.09	491.39	189.81 ± 0.09
	132	2500	5.12 ± 0.03	-306.55 ± 0.03	-301.43± 0.04	491.39	189.96 ± 0.04
	291	5500	5.09 ± 0.01	-306.50 ± 0.07	-301.41 ± 0.07	491.39	189.98 ± 0.07
		∞			-300.87 ± 0.11		190.52 ± 0.11
Cl^-	26	500	31.26 ± 0.09	-398.84 ± 0.06	-367.58 ± 0.11	-229.73	-597.31 ± 0.11
	79	1500	31.46 ± 0.06	-398.45 ± 0.03	-366.99 ± 0.07	-229.73	-596.72 ± 0.07
	132	2500	31.46 ± 0.12	-398.36 ± 0.03	-366.90 ± 0.12	-229.73	-596.63 ± 0.12
	291	5500	31.34 ± 0.04	-398.31 ± 0.03	-366.97 ± 0.05	-229.73	-596.70 ± 0.05
		∞			-366.51 ± 0.12		-596.24 ± 0.12

Table S19. Chemical potentials and extrapolation to infinite system size for added K^+ or Cl^- in aqueous KCl solutions at m =3.941 mol salt / kg H_2O.

	N_{salt}	N_{water}	$\mu^{\mathrm{ex,LJ}}$ (kJ/mol)	$\mu^{\mathrm{ex,Coul}}$ (kJ/mol)	μ^{ex} (kJ/mol)	μ^{IG} (kJ/mol)	μ (kJ/mol)
K^+	35	500	5.65 ± 0.06	-309.61 ± 0.07	-303.96 ± 0.13	492.04	188.08 ± 0.13
	106	1500	5.77 ± 0.02	-309.26 ± 0.06	-303.49 ± 0.08	492.04	188.55 ± 0.08
	177	2500	5.70 ± 0.03	-309.13 ± 0.04	-303.43 ± 0.07	492.04	188.61 ± 0.07
	390	5500	5.71 ± 0.03	-309.07 ± 0.03	-303.36 ± 0.06	492.04	188.68 ± 0.06
		∞			-302.89 ± 0.15		189.15 ± 0.15
Cl^-	35	500	32.47 ± 0.16	-397.36 ± 0.09	-364.89 ± 0.18	-229.07	-593.96 ± 0.18
	106	1500	32.39 ± 0.05	-396.96 ± 0.08	-364.57 ± 0.19	-229.07	-593.64 ± 0.09
	177	2500	32.27 ± 0.07	-397.03 ± 0.09	-364.76 ± 0.11	-229.07	-593.83 ± 0.11
	390	5500	32.30 ± 0.07	-396.92 ± 0.05	-364.62 ± 0.09	-229.07	-593.69 ± 0.09
		∞			-364.49 ± 0.21		-593.56 ± 0.21

Table S20. Chemical potentials and extrapolation to infinite system size for added K^+ or Cl^- in aqueous KCl solutions at m =4.940 mol salt / kg H_2O.

	N_{salt}	N_{water}	$\mu^{\mathrm{ex,LJ}}$ (kJ/mol)	$\mu^{\mathrm{ex,Coul}}$ (kJ/mol)	μ^{ex} (kJ/mol)	μ^{IG} (kJ/mol)	μ (kJ/mol)
K^+	44	500	6.42 ± 0.09	-311.93 ± 0.05	-305.51 ± 0.14	492.53	187.02 ± 0.14
	133	1500	6.38 ± 0.05	-311.65 ± 0.06	-305.27 ± 0.11	492.53	187.26 ± 0.11
	222	2500	6.35 ± 0.03	-311.58 ± 0.03	-305.23 ± 0.06	492.53	187.30 ± 0.06
	489	5500	6.28 ± 0.03	-311.54 ± 0.02	-305.26 ± 0.05	492.53	187.27 ± 0.05
		∞			-305.08 ± 0.14		187.45 ± 0.14
Cl^-	44	500	33.27 ± 0.18	-395.79 ± 0.02	-362.52 ± 0.18	-228.59	-591.11 ± 0.18
	133	1500	33.41 ± 0.05	-395.58 ± 0.13	-362.17 ± 0.14	-228.59	-590.76 ± 0.14
	222	2500	33.24 ± 0.07	-395.53 ± 0.06	-362.29 ± 0.09	-228.59	-590.88 ± 0.09
	489	5500	33.47 ± 0.06	-395.45 ± 0.09	-361.98 ± 0.11	-228.59	-590.57 ± 0.11
		∞			-361.66 ± 0.23		-590.25 ± 0.23

Aqueous NaF Solution:

Table S21. Chemical potentials and extrapolation to infinite system size for added Na^+ or F^- in aqueous NaF solutions at m = 0.019 mol salt / kg H_2O.

	N_{salt}	N_{water}	$\mu^{ex,LJ}$ (kJ/mol)	$\mu^{ex,Coul}$ (kJ/mol)	μ^{ex} (kJ/mol)	μ^{IG} (kJ/mol)	μ (kJ/mol)
Na^+	0	1500	2.16 ± 0.02	-372.95 ± 0.05	-370.79 ± 0.05	572.38	201.59 ± 0.05
	1	4500	2.16 ± 0.02	-372.65 ± 0.05	-370.49 ± 0.05	572.38	201.89 ± 0.05
	2	7500	2.16 ± 0.02	-372.64 ± 0.04	-370.48 ± 0.04	572.38	201.90 ± 0.04
		∞			-370.00 ± 0.13		202.38 ± 0.13
F^-	0	1500	19.49 ± 0.04	-518.36 ± 0.05	-498.87 ± 0.07	-263.93	-762.80 ± 0.07
	1	4500	19.55 ± 0.04	-518.15 ± 0.05	-498.60 ± 0.07	-263.93	-762.53 ± 0.07
	2	7500	19.53 ± 0.03	-518.05 ± 0.04	-498.52 ± 0.05	-263.93	-762.45 ± 0.05
		∞			-498.02 ± 0.15		-761.95 ± 0.15

Table S22. Chemical potentials and extrapolation to infinite system size for added Na^+ or F^- in aqueous NaF solutions at m = 0.056 mol salt / kg H_2O.

	N_{salt}	N_{water}	$\mu^{ex,LJ}$ (kJ/mol)	$\mu^{ex,Coul}$ (kJ/mol)	μ^{ex} (kJ/mol)	μ^{IG} (kJ/mol)	μ (kJ/mol)
Na^+	0	500	2.21 ± 0.03	-373.48 ± 0.05	-371.27 ± 0.06	575.11	203.84 ± 0.06
	1	1500	2.19 ± 0.02	-372.98 ± 0.05	-370.79 ± 0.05	575.11	204.32 ± 0.05
	2	2500	2.21 ± 0.02	-372.98 ± 0.05	-370.77 ± 0.06	575.11	204.34 ± 0.06
	5	5500	2.20 ± 0.02	-372.82 ± 0.05	-370.62 ± 0.05	575.11	204.49 ± 0.05
		∞			-370.07 ± 0.10		205.04 ± 0.10
F^-	0	500	19.43 ± 0.05	-518.95 ± 0.07	-499.52 ± 0.08	-261.21	-760.73 ± 0.08
	1	1500	19.55 ± 0.03	-518.59 ± 0.06	-499.04 ± 0.07	-261.21	-760.25 ± 0.08
	2	2500	19.59 ± 0.03	-518.48 ± 0.05	-498.89 ± 0.07	-261.21	-760.10 ± 0.08
	5	5500	19.58 ± 0.02	-518.51 ± 0.04	-498.93 ± 0.05	-261.21	-760.14 ± 0.08
		∞			-498.38 ± 0.12		-759.58 ± 0.12

Table S23. Chemical potentials and extrapolation to infinite system size for added Na^+ or F^- in aqueous NaF solutions at m = 0.167 mol salt / kg H_2O.

	N_{salt}	N_{water}	$\mu^{ex,LJ}$ (kJ/mol)	$\mu^{ex,Coul}$ (kJ/mol)	μ^{ex} (kJ/mol)	μ^{IG} (kJ/mol)	μ (kJ/mol)
Na^+	1	500	2.21 ± 0.04	-373.36 ± 0.06	-371.15 ± 0.07	577.83	206.68 ± 0.07
	4	1500	2.24 ± 0.02	-373.08 ± 0.05	-370.84 ± 0.06	577.84	207.00 ± 0.06
	7	2500	2.26 ± 0.03	-373.08 ± 0.04	-370.82 ± 0.05	577.84	207.02 ± 0.05
	16	5500	2.27 ± 0.02	-372.98 ± 0.04	-370.71 ± 0.05	577.84	207.13 ± 0.05
		∞			-370.35 ± 0.10		207.48 ± 0.10
F^-	1	500	19.68 ± 0.05	-519.53 ± 0.06	-499.85 ± 0.08	-258.48	-758.33 ± 0.08
	4	1500	19.71 ± 0.04	-519.23 ± 0.06	-499.52 ± 0.07	-258.48	-758.00 ± 0.07
	7	2500	19.74 ± 0.04	-519.13 ± 0.06	-499.39 ± 0.07	-258.48	-757.87 ± 0.07
	16	5500	19.70 ± 0.03	-519.14 ± 0.06	-499.44 ± 0.07	-258.48	-757.92 ± 0.07
		∞			-499.01 ± 0.13		-757.49 ± 0.13

Table S24. Chemical potentials and extrapolation to infinite system size for added Na^+ or F^- in aqueous NaF solutions at m = 0.611 mol salt / kg H_2O.

	N_{salt}	N_{water}	$\mu^{ex,LJ}$ (kJ/mol)	$\mu^{ex,Coul}$ (kJ/mol)	μ^{ex} (kJ/mol)	μ^{IG} (kJ/mol)	μ (kJ/mol)
Na^+	5	500	2.55 ± 0.04	-373.55 ± 0.07	-371.00 ± 0.07	581.07	210.07 ± 0.07
	16	1500	2.55 ± 0.03	-373.28 ± 0.07	-370.73 ± 0.07	581.07	210.34 ± 0.07
	27	2500	2.56 ± 0.03	-373.27 ± 0.07	-370.71 ± 0.07	581.07	210.36 ± 0.07
	60	5500	2.56 ± 0.02	-373.20 ± 0.05	-370.64 ± 0.05	581.07	210.43 ± 0.05
		∞			-370.33 ± 0.11		210.74 ± 0.11
F^-	5	500	20.27 ± 0.05	-521.28 ± 0.07	-501.01 ± 0.08	-255.24	-756.25 ± 0.08
	16	1500	20.32 ± 0.04	-521.19 ± 0.06	-500.87 ± 0.07	-255.24	-756.11 ± 0.07
	27	2500	20.33 ± 0.04	-521.04 ± 0.07	-500.71 ± 0.08	-255.24	-755.95 ± 0.08
	60	5500	20.34 ± 0.03	-521.12 ± 0.05	-500.78 ± 0.06	-255.24	-756.02 ± 0.06
		∞			-500.54 ± 0.13		-755.78 ± 0.13

Table S25. Chemical potentials and extrapolation to infinite system size for added Na^+ or F^- in aqueous NaF solutions at m = 0.944 mol salt / kg H_2O.

	N_{salt}	N_{water}	$\mu^{ex,LJ}$ (kJ/mol)	$\mu^{ex,Coul}$ (kJ/mol)	μ^{ex} (kJ/mol)	μ^{IG} (kJ/mol)	μ (kJ/mol)
Na^+	8	500	2.79 ± 0.03	-373.68 ± 0.06	-370.89 ± 0.07	582.16	211.27 ± 0.07
	25	1500	2.77 ± 0.03	-373.48 ± 0.06	-370.71 ± 0.07	582.16	211.45 ± 0.07
	42	2500	2.76 ± 0.02	-373.44 ± 0.06	-370.68 ± 0.06	582.16	211.48 ± 0.06
	93	5500	2.77 ± 0.02	-373.42 ± 0.05	-370.65 ± 0.05	582.16	211.51 ± 0.05
		∞			-370.44 ± 0.11		211.73 ± 0.11
F^-	8	500	20.77 ± 0.05	-522.64 ± 0.06	-501.87 ± 0.08	-254.15	-756.02 ± 0.08
	25	1500	20.77 ± 0.05	-522.19 ± 0.07	-501.42 ± 0.09	-254.15	-755.57 ± 0.09
	42	2500	20.87 ± 0.04	-522.10 ± 0.05	-501.23 ± 0.07	-254.15	-755.38 ± 0.07
	93	5500	20.77 ± 0.03	-522.17 ± 0.06	-501.40 ± 0.07	-254.15	-755.55 ± 0.07
		∞			-500.84 ± 0.13		-754.98 ± 0.13

Table S26. Chemical potentials and extrapolation to infinite system size for added Na^+ or F^- in aqueous NaF solutions at m = 1.943 mol salt / kg H_2O.

	N_{salt}	N_{water}	$\mu^{ex,LJ}$ (kJ/mol)	$\mu^{ex,Coul}$ (kJ/mol)	μ^{ex} (kJ/mol)	μ^{IG} (kJ/mol)	μ (kJ/mol)
Na^+	17	500	3.40 ± 0.03	-374.39 ± 0.07	-370.99 ± 0.08	583.98	212.99 ± 0.08
	52	1500	3.36 ± 0.03	-373.94 ± 0.07	-370.58 ± 0.08	583.98	213.40 ± 0.08
	87	2500	3.41 ± 0.03	-373.98 ± 0.07	-370.57 ± 0.08	583.98	213.41 ± 0.08
	192	5500	3.43 ± 0.03	-373.93 ± 0.05	-370.50 ± 0.06	583.98	213.48 ± 0.06
		∞			-370.07 ± 0.12		213.91 ± 0.12
F^-	17	500	22.11 ± 0.06	-525.13 ± 0.14	-503.02 ± 0.10	-252.33	-755.35 ± 0.10
	52	1500	22.19 ± 0.05	-524.96 ± 0.10	-502.77 ± 0.09	-252.33	-755.10 ± 0.09
	87	2500	22.24 ± 0.04	-524.77 ± 0.09	-502.53 ± 0.08	-252.33	-754.86 ± 0.08
	192	5500	22.23 ± 0.04	-524.60 ± 0.07	-502.37 ± 0.07	-252.33	-754.70 ± 0.07
		∞			-501.84 ± 0.15		-754.17 ± 0.15

Aqueous KF Solution:

Table S27. Chemical potentials and extrapolation to infinite system size for added K^+ or F^- in aqueous KF solutions at m = 0.019 mol salt / kg H_2O.

	N_{salt}	N_{water}	$\mu^{\text{ex,LJ}}$ (kJ/mol)	$\mu^{\text{ex,Coul}}$ (kJ/mol)	μ^{ex} (kJ/mol)	μ^{IG} (kJ/mol)	μ (kJ/mol)
K^+	0	1500	3.10 ± 0.04	-299.04 ± 0.05	-295.94 ± 0.06	479.01	183.07 ± 0.06
	1	4500	3.05 ± 0.03	-298.78 ± 0.04	-295.73 ± 0.05	479.01	183.28 ± 0.05
	2	7500	3.10 ± 0.02	-298.72 ± 0.04	-295.62 ± 0.04	479.01	183.39 ± 0.04
		∞			-295.18 ± 0.14		183.83 ± 0.14
F^-	0	1500	19.48 ± 0.04	-518.32 ± 0.06	-498.84 ± 0.07	-263.93	-762.77 ± 0.07
	1	4500	19.56 ± 0.04	-518.08 ± 0.06	-498.52 ± 0.07	-263.93	-762.45 ± 0.07
	2	7500	19.50 ± 0.03	-517.99 ± 0.04	-498.49 ± 0.05	-263.93	-762.42 ± 0.05
		∞			-497.97 ± 0.16		-761.91 ± 0.16

Table S28. Chemical potentials and extrapolation to infinite system size for added K^+ or F^- in aqueous KF solutions at m = 0.056 mol salt / kg H_2O.

	N_{salt}	N_{water}	$\mu^{\text{ex,LJ}}$ (kJ/mol)	$\mu^{\text{ex,Coul}}$ (kJ/mol)	μ^{ex} (kJ/mol)	μ^{IG} (kJ/mol)	μ (kJ/mol)
K^+	0	500	3.09 ± 0.04	-299.73 ± 0.06	-296.64 ± 0.07	481.74	185.10 ± 0.07
	1	1500	3.07 ± 0.03	-299.23 ± 0.04	-296.16 ± 0.05	481.74	185.58 ± 0.05
	2	2500	3.11 ± 0.03	-299.08 ± 0.05	-295.97 ± 0.06	481.74	185.77 ± 0.06
	5	5500	3.09 ± 0.03	-298.96 ± 0.04	-295.87 ± 0.05	481.74	185.87 ± 0.05
		∞			-295.20 ± 0.10		186.53 ± 0.10
F^-	0	500	19.53 ± 0.04	-519.06 ± 0.06	-499.53 ± 0.07	-261.21	-760.74 ± 0.07
	1	1500	19.54 ± 0.05	-518.56 ± 0.05	-499.02 ± 0.07	-261.21	-760.23 ± 0.07
	2	2500	19.59 ± 0.04	-518.32 ± 0.05	-498.73 ± 0.07	-261.21	-759.94 ± 0.07
	5	5500	19.57 ± 0.03	-518.34 ± 0.05	-498.77 ± 0.06	-261.21	-759.98 ± 0.06
		∞			-498.01 ± 0.12		-759.22 ± 0.12

Table S29. Chemical potentials and extrapolation to infinite system size for added K^+ or F^- in aqueous KF solutions at m = 0.167 mol salt / kg H_2O.

	N_{salt}	N_{water}	$\mu^{\text{ex,LJ}}$ (kJ/mol)	$\mu^{\text{ex,Coul}}$ (kJ/mol)	μ^{ex} (kJ/mol)	μ^{IG} (kJ/mol)	μ (kJ/mol)
K^+	1	500	3.19 ± 0.04	-300.18 ± 0.05	-296.99 ± 0.07	484.46	187.47 ± 0.07
	4	1500	3.18 ± 0.03	-299.74 ± 0.05	-296.56 ± 0.06	484.46	187.90 ± 0.06
	7	2500	3.23 ± 0.03	-299.69 ± 0.05	-296.46 ± 0.06	484.46	188.00 ± 0.06
	16	5500	3.22 ± 0.03	-299.60 ± 0.05	-296.38 ± 0.06	484.46	188.08 ± 0.06
		∞			-295.83 ± 0.11		188.63 ± 0.11
F^-	1	500	19.65 ± 0.05	-519.27 ± 0.06	-499.62 ± 0.08	-258.48	-758.10 ± 0.08
	4	1500	19.67 ± 0.04	-518.78 ± 0.05	-499.11 ± 0.07	-258.48	-757.59 ± 0.07
	7	2500	19.70 ± 0.04	-518.67 ± 0.06	-498.97 ± 0.07	-258.48	-757.45 ± 0.07
	16	5500	19.74 ± 0.04	-518.75 ± 0.06	-499.01 ± 0.07	-258.48	-757.49 ± 0.07
		∞			-498.38 ± 0.13		-756.86 ± 0.13

Table S30. Chemical potentials and extrapolation to infinite system size for added K^+ or F^- in aqueous KF solutions at m = 0.611 mol salt / kg H_2O.

	N_{salt}	N_{water}	$\mu^{\mathrm{ex,LJ}}$ (kJ/mol)	$\mu^{\mathrm{ex,Coul}}$ (kJ/mol)	μ^{ex} (kJ/mol)	μ^{IG} (kJ/mol)	μ (kJ/mol)
K^+	5	500	3.53 ± 0.04	-301.94 ± 0.06	-298.41 ± 0.07	487.69	189.28 ± 0.07
	16	1500	3.59 ± 0.03	-301.42 ± 0.05	-297.83 ± 0.06	487.69	189.86 ± 0.06
	27	2500	3.63 ± 0.04	-301.44 ± 0.05	-297.81 ± 0.06	487.69	189.88 ± 0.06
	60	5500	3.57 ± 0.03	-301.41 ± 0.05	-297.84 ± 0.06	487.69	189.85 ± 0.06
		∞			-297.24 ± 0.12		190.45 ± 0.12
F^-	5	500	20.35 ± 0.05	-519.80 ± 0.11	-499.45 ± 0.09	-255.25	-754.70 ± 0.09
	16	1500	20.38 ± 0.04	-519.60 ± 0.09	-499.22 ± 0.08	-255.25	-754.47 ± 0.08
	27	2500	20.34 ± 0.04	-519.47 ± 0.07	-499.13 ± 0.07	-255.25	-754.38 ± 0.07
	60	5500	20.34 ± 0.03	-519.35 ± 0.06	-499.01 ± 0.07	-255.25	-754.26 ± 0.07
		∞			-498.66 ± 0.13		-753.91 ± 0.13

Table S31. Chemical potentials and extrapolation to infinite system size for added K^+ or F^- in aqueous KF solutions at m = 0.944 mol salt / kg H_2O.

	N_{salt}	N_{water}	$\mu^{\mathrm{ex,LJ}}$ (kJ/mol)	$\mu^{\mathrm{ex,Coul}}$ (kJ/mol)	μ^{ex} (kJ/mol)	μ^{IG} (kJ/mol)	μ (kJ/mol)
K^+	8	500	3.79 ± 0.04	-303.09 ± 0.07	-299.30 ± 0.08	488.77	189.47 ± 0.08
	25	1500	3.82 ± 0.04	-302.79 ± 0.06	-298.97 ± 0.07	488.77	189.80 ± 0.07
	42	2500	3.84 ± 0.04	-302.67 ± 0.05	-298.83 ± 0.07	488.77	189.94 ± 0.07
	93	5500	3.86 ± 0.03	-302.77 ± 0.06	-298.91 ± 0.07	488.77	189.86 ± 0.07
		∞			-298.47 ± 0.13		190.30 ± 0.13
F^-	8	500	20.76 ± 0.05	-520.19 ± 0.06	-499.43 ± 0.08	-254.17	-753.60 ± 0.08
	25	1500	20.66 ± 0.04	-519.70 ± 0.05	-499.04 ± 0.07	-254.17	-753.21 ± 0.07
	42	2500	20.75 ± 0.05	-519.82 ± 0.06	-499.07 ± 0.08	-254.17	-753.24 ± 0.08
	93	5500	20.73 ± 0.04	-519.66 ± 0.06	-498.93 ± 0.07	-254.17	-753.10 ± 0.07
		∞			-498.51 ± 0.13		-752.68 ± 0.13

Table S32. Chemical potentials and extrapolation to infinite system size for added K^+ or F^- in aqueous KF solutions at m =1.943 mol salt / kg H_2O.

	N_{salt}	N_{water}	$\mu^{\mathrm{ex,LJ}}$ (kJ/mol)	$\mu^{\mathrm{ex,Coul}}$ (kJ/mol)	μ^{ex} (kJ/mol)	μ^{IG} (kJ/mol)	μ (kJ/mol)
K^+	17	500	4.65 ± 0.04	-306.81 ± 0.07	-302.16 ± 0.08	490.56	188.40 ± 0.08
	52	1500	4.76 ± 0.04	-306.51 ± 0.07	-301.75 ± 0.08	490.56	188.81 ± 0.08
	87	2500	4.71 ± 0.04	-306.26 ± 0.06	-301.55 ± 0.07	490.56	189.01 ± 0.07
	192	5500	4.68 ± 0.03	-306.27 ± 0.05	-301.59 ± 0.06	490.56	188.97 ± 0.06
		∞			-301.04 ± 0.12		189.53 ± 0.12
F^-	17	500	22.01 ± 0.05	-520.00 ± 0.08	-497.99 ± 0.10	-252.38	-750.37 ± 0.10
	52	1500	22.09 ± 0.05	-519.65 ± 0.07	-497.56 ± 0.09	-252.38	-749.94 ± 0.09
	87	2500	22.05 ± 0.05	-519.82 ± 0.07	-497.77 ± 0.08	-252.38	-750.15 ± 0.08
	192	5500	22.11 ± 0.05	-519.68 ± 0.05	-497.57 ± 0.07	-252.38	-749.95 ± 0.07
		∞			-497.27 ± 0.15		-749.65 ± 0.15

Table S33. Chemical potentials and extrapolation to infinite system size for added K^+ or F^- in aqueous KF solutions at m =2.942 mol salt / kg H_2O.

	N_{salt}	N_{water}	$\mu^{\mathrm{ex,LJ}}$ (kJ/mol)	$\mu^{\mathrm{ex,Coul}}$ (kJ/mol)	μ^{ex} (kJ/mol)	μ^{IG} (kJ/mol)	μ (kJ/mol)
K^+	26	500	5.53 ± 0.05	-310.57 ± 0.07	-305.04 ± 0.09	491.59	186.55 ± 0.09
	79	1500	5.57 ± 0.04	-310.15 ± 0.07	-304.58 ± 0.09	491.59	187.01 ± 0.07
	132	2500	5.56 ± 0.04	-310.16 ± 0.06	-304.60 ± 0.07	491.59	186.99 ± 0.07
	291	5500	5.52 ± 0.04	-310.00 ± 0.06	-304.48 ± 0.07	491.59	187.11 ± 0.07
					-304.00 ± 0.14		187.58 ± 0.14
F^-	26	500	23.37 ± 0.06	-519.62 ± 0.07	-496.25 ± 0.09	-251.36	-747.61 ± 0.09
	79	1500	23.37 ± 0.05	-519.31 ± 0.08	-495.94 ± 0.09	-251.36	-747.30 ± 0.09
	132	2500	23.35 ± 0.05	-519.06 ± 0.06	-495.71 ± 0.08	-251.36	-747.07 ± 0.08
	291	5500	23.35 ± 0.04	-519.00 ± 0.06	-495.65 ± 0.08	-251.36	-747.01 ± 0.08
					-495.11 ± 0.15		-746.47 ± 0.15

Table S34. Chemical potentials and extrapolation to infinite system size for added K^+ or F^- in aqueous KF solutions at m =3.941 mol salt / kg H_2O.

	N_{salt}	N_{water}	$\mu^{\mathrm{ex,LJ}}$ (kJ/mol)	$\mu^{\mathrm{ex,Coul}}$ (kJ/mol)	μ^{ex} (kJ/mol)	μ^{IG} (kJ/mol)	μ (kJ/mol)
K^+	35	500	6.52 ± 0.05	-314.25 ± 0.08	-307.73 ± 0.09	492.30	184.57 ± 0.09
	106	1500	6.39 ± 0.05	-314.04 ± 0.07	-307.65 ± 0.09	492.30	184.65 ± 0.09
	177	2500	6.44 ± 0.04	-313.79 ± 0.06	-307.35 ± 0.07	492.30	184.95 ± 0.07
	390	5500	6.45 ± 0.04	-313.76 ± 0.06	-307.31 ± 0.07	492.30	184.99 ± 0.07
					-306.93 ± 0.14		185.37 ± 0.14
F^-	35	500	24.77 ± 0.07	-518.80 ± 0.10	-494.03 ± 0.10	-250.65	-744.68 ± 0.10
	106	1500	24.82 ± 0.06	-518.19 ± 0.14	-493.37 ± 0.10	-250.65	-744.02 ± 0.10
	177	2500	24.85 ± 0.07	-518.12 ± 0.12	-493.27 ± 0.10	-250.65	-743.92 ± 0.10
	390	5500	24.77 ± 0.05	-518.10 ± 0.07	-493.33 ± 0.08	-250.65	-743.98 ± 0.08
					-492.64 ± 0.15		-743.29 ± 0.15

Table S35. Chemical potentials and extrapolation to infinite system size for added K^+ or F^- in aqueous KF solutions at m =4.940 mol salt / kg H_2O.

	N_{salt}	N_{water}	$\mu^{\mathrm{ex,LJ}}$ (kJ/mol)	$\mu^{\mathrm{ex,Coul}}$ (kJ/mol)	μ^{ex} (kJ/mol)	μ^{IG} (kJ/mol)	μ (kJ/mol)
K^+	44	500	7.47 ± 0.05	-318.03 ± 0.06	-310.56 ± 0.08	492.84	182.28 ± 0.08
	133	1500	7.52 ± 0.05	-317.82 ± 0.07	-310.30 ± 0.09	492.84	182.54 ± 0.09
	222	2500	7.40 ± 0.05	-317.75 ± 0.07	-310.35 ± 0.08	492.84	182.49 ± 0.08
	489	5500	7.45 ± 0.04	-317.66 ± 0.07	-310.21 ± 0.08	492.84	182.63 ± 0.08
					-309.95 ± 0.14		182.89 ± 0.14
F^-	44	500	26.57 ± 0.08	-517.55 ± 0.08	-490.98 ± 0.12	-250.10	-741.08 ± 0.12
	133	1500	26.41 ± 0.06	-517.02 ± 0.07	-490.61 ± 0.09	-250.10	-740.71 ± 0.09
	222	2500	26.43 ± 0.06	-516.98 ± 0.09	-490.55 ± 0.10	-250.10	-740.65 ± 0.10
	489	5500	26.27 ± 0.05	-517.01 ± 0.07	-490.74 ± 0.09	-250.10	-740.84 ± 0.09
					-490.40 ± 0.18		-740.50 ± 0.18

Table S36. Chemical potential at infinite dilution reference state for individual ions and salt in aqueous NaCl solution.

Source	$\mu^{\dagger}_{Na^+}$ (kJ/mol)	$\mu^{\dagger}_{Cl^-}$ (kJ/mol)	$\mu^{\dagger}_{NaCl}$ (kJ/mol)
This Work (DS[a])	212.73 ± 0.11	-603.43 ± 0.23	-390.69 ± 0.25
This Work (DE[b])	212.59 ± 0.14	-603.87 ± 0.25	-391.28 ± 0.29
Mester and Panagiotopoulos[1]	–	–	-391.60 ± 0.20
Young and Panagiotopoulos[5]	–	–	-390.80 ± 0.90
Moučka et al.[6]	–	–	-391.278
Experiments[7]	–	–	-393.133

[a] by enforcing the Debye-Hückel limiting slope at lowest concentration.
[b] by double extrapolation.

Table S37. Chemical potential at infinite dilution reference state for individual ions and salt in aqueous KCl solution.

Source	$\mu^{\dagger}_{K^+}$ (kJ/mol)	$\mu^{\dagger}_{Cl^-}$ (kJ/mol)	$\mu^{\dagger}_{KCl}$ (kJ/mol)
This Work (DS)	194.46 ± 0.10	-603.45 ± 0.06	-409.00 ± 0.12
This Work (DE)	194.30 ± 0.20	-603.81 ± 0.27	-409.51 ± 0.34
Mester and Panagiotopoulos[2]	–	–	-410.0 ± 0.2
Experiments[7]	–	–	-414.49

Table S38. Chemical potential at infinite dilution reference state for individual ions and salt in aqueous NaF solution.

Source	$\mu^{\dagger}_{Na^+}$ (kJ/mol)	$\mu^{\dagger}_{F^-}$ (kJ/mol)	$\mu^{\dagger}_{NaF}$ (kJ/mol)
This Work (DS)	212.71 ± 0.05	-751.62 ± 0.07	-538.91 ± 0.09
This Work (DE)	212.49 ± 0.17	-751.57 ± 0.20	-539.07 ± 0.27
Mester and Panagiotopoulos[2]	–	–	-539.3 ± 0.2
Experiments[7]	–	–	-540.68

Table S39. Chemical potential at infinite dilution reference state for individual ions and salt in aqueous KF solution.

Source	$\mu^{\dagger}_{K}$ (kJ/mol)	$\mu^{\dagger}_{F}$ (kJ/mol)	$\mu^{\dagger}_{KF}$ (kJ/mol)
This Work (DS)	194.16 ± 0.14	-751.58 ± 0.16	-557.41 ± 0.21
This Work (DE)	194.21 ± 0.18	-751.73 ± 0.21	-557.52 ± 0.28
Experiments[7]	–	–	-562.06

Table S40. Simulation results for individual and mean ionic activity coefficients in aqueous NaCl solution.

m	$\ln \gamma_+$	$\ln \gamma_-$	$\ln \gamma_\pm$
0.019	-0.18 ± 0.06	-0.18 ± 0.13	-0.18 ± 0.14
0.056	-0.12 ± 0.05	-0.31 ± 0.10	-0.21 ± 0.12
0.167	-0.27 ± 0.05	-0.36 ± 0.10	-0.31 ± 0.12
0.611	0.01 ± 0.06	-0.72 ± 0.11	-0.36 ± 0.12
0.944	0.17 ± 0.07	-1.06 ± 0.11	-0.44 ± 0.13
1.943	0.57 ± 0.06	-1.02 ± 0.11	-0.23 ± 0.13
2.942	0.92 ± 0.07	-1.07 ± 0.12	-0.07 ± 0.14
3.941	1.23 ± 0.10	-0.92 ± 0.12	0.15 ± 0.16
4.940	1.28 ± 0.08	-0.43 ± 0.14	0.43 ± 0.16
5.939	1.37 ± 0.08	0.02 ± 0.17	0.70 ± 0.19

Table S41. Simulation results for individual and mean ionic activity coefficients in aqueous KCl solution.

m	$\ln \gamma_+$	$\ln \gamma_-$	$\ln \gamma_\pm$
0.019	-0.18 ± 0.10	-0.18 ± 0.06	-0.18 ± 0.11
0.056	-0.25 ± 0.05	-0.11 ± 0.06	-0.18 ± 0.08
0.167	-0.40 ± 0.06	-0.17 ± 0.06	-0.28 ± 0.08
0.611	-0.83 ± 0.06	0.05 ± 0.07	-0.39 ± 0.09
0.944	-1.14 ± 0.06	0.14 ± 0.05	-0.50 ± 0.08
1.943	-1.82 ± 0.06	0.83 ± 0.07	-0.50 ± 0.10
2.942	-2.67 ± 0.06	1.83 ± 0.06	-0.42 ± 0.08
3.941	-3.51 ± 0.07	2.62 ± 0.09	-0.45 ± 0.11
4.940	-4.42 ± 0.07	3.73 ± 0.10	-0.35 ± 0.12

Table S42. Simulation results for individual and mean ionic activity coefficients in aqueous NaF solution.

m	$\ln \gamma_+$	$\ln \gamma_-$	$\ln \gamma_\pm$
0.019	-0.18 ± 0.06	-0.18 ± 0.07	-0.18 ± 0.09
0.056	-0.20 ± 0.05	-0.32 ± 0.05	-0.26 ± 0.07
0.167	-0.32 ± 0.05	-0.57 ± 0.06	-0.44 ± 0.07
0.611	-0.30 ± 0.05	-1.18 ± 0.06	-0.74 ± 0.08
0.944	-0.34 ± 0.05	-1.30 ± 0.06	-0.82 ± 0.08
1.943	-0.18 ± 0.05	-1.69 ± 0.07	-0.94 ± 0.08

Table S43. Simulation results for individual and mean ionic activity coefficients in aqueous KF solution.

m	$\ln \gamma_+$	$\ln \gamma_-$	$\ln \gamma_\pm$
0.019	-0.18 ± 0.09	-0.18 ± 0.12	-0.18 ± 0.15
0.056	-0.16 ± 0.04	-0.26 ± 0.05	-0.21 ± 0.06
0.167	-0.41 ± 0.04	-0.41 ± 0.05	-0.41 ± 0.07
0.611	-0.98 ± 0.05	-0.52 ± 0.05	-0.75 ± 0.07
0.944	-1.47 ± 0.05	-0.46 ± 0.05	-0.96 ± 0.07
1.943	-2.50 ± 0.05	0.04 ± 0.06	-1.23 ± 0.08
2.942	-3.70 ± 0.06	0.91 ± 0.06	-1.40 ± 0.08
3.941	-4.89 ± 0.06	1.90 ± 0.06	-1.49 ± 0.08
4.940	-6.11 ± 0.06	2.80 ± 0.07	-1.66 ± 0.09

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