Supporting Information: The Effect of Atmospherically Relevant Aminium Salts on Water Uptake

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S1 Parametrization for Electrolytes

A new parametrization for the COSMO*therm* program, BP-TZVP-ELYTE-21, was recently developed for property calculations of small, spherical ions. This electrolyte parametrization can be used with cosmo-files calculated at the BP/def-TZVP level of theory. In order to find the best agreement between experimental and COSMO*therm*-derived water activities, three different parametrizations were tested: BP_TZVP_21 (abbreviated TZVP), BP_TZVP_ELYTE_21 (abbreviated ELYTE) and BP_TZVPD_FINE_21 (abbreviated FINE).

Water activities in sulfate solutions have been derived experimentally by Clegg et al.,[S1] Sauerwein et al.[S2] and Rovelli et al.[S3] The comparison between these experiments and COSMO*therm* calculations is shown in Figure S1. The experimental water activities of Sauerwein et al.[S2] were taken only from experiments of that had close to 2:1 ratio (1.92447:1 to 2.0979:1) of amines and sulfuric acid, corresponding to sulfate as opposed to bisulfate solutions. The experiments of Rovelli et al.[S3] were conducted in 293 K, which is 5 K lower than the temperature of the calculations. However, such a small temperature difference should lead to only minor differences in water activities.

It is clear that the FINE parametrization is not able to capture even the general trend of experimental water activity in ammonium sulfate solutions. This result can be expected, as both ammonium and sulfate are small spherical ions. The FINE parametrization predicts water activities higher than 1 when the salt mole fraction is higher than 0.015, which is well below the solubility limit of AS in water at 298.15 K. Water activities calculated using the lower level of theory TZVP parametrization agree with the experiments much better than the FINE parametrization, which uses a higher level of theory DFT input. Additionally, there is no significant difference between the TZVP and ELYTE parametrizations. For the alkyl aminium sulfates, TZVP clearly underestimates the experimental water activities, while ELYTE and FINE lead to quite equal agreement with experiments.

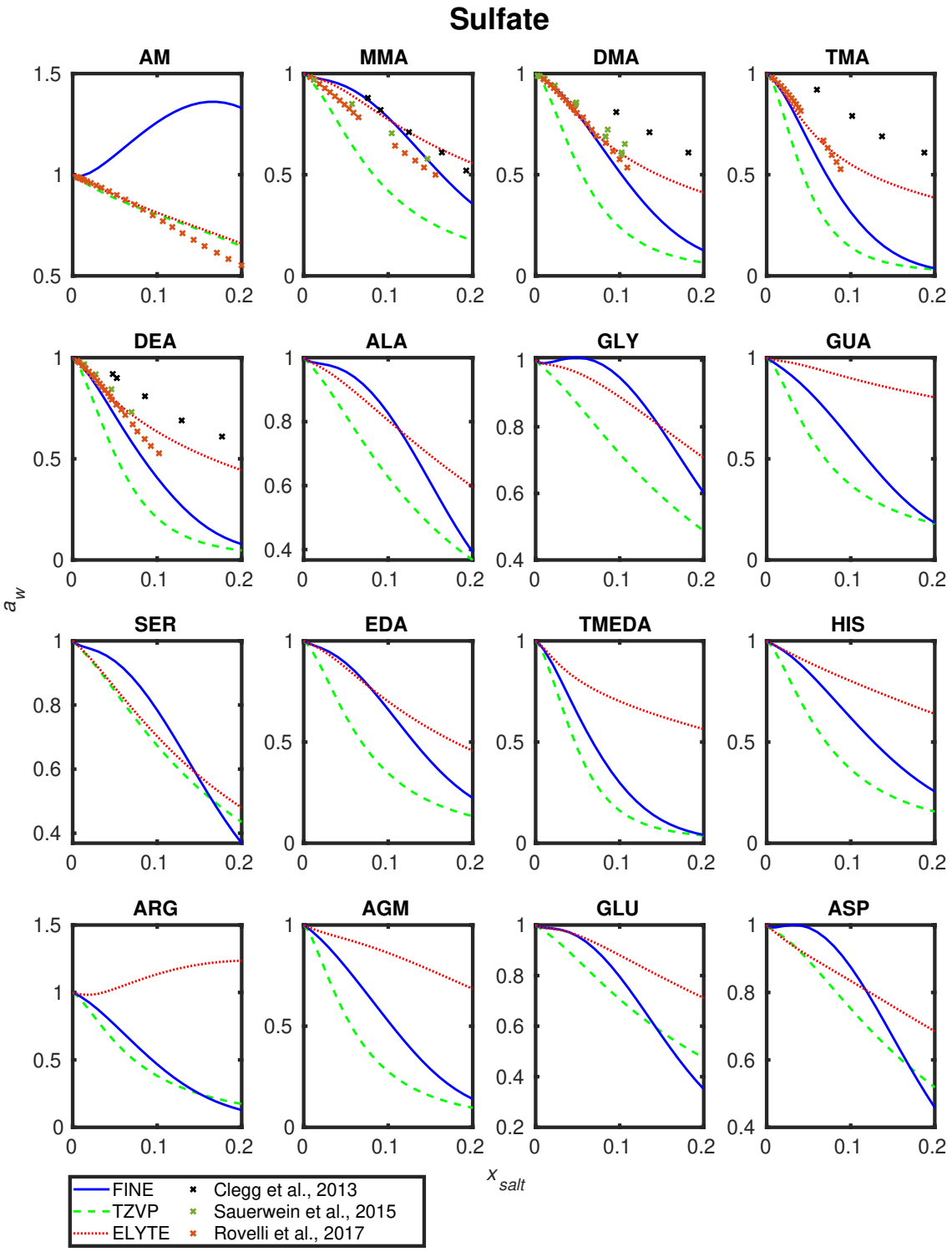

Figure S1: Comparison between experiments[S1–S3] and different COSMO*therm* parametrizations in aqueous sulfate salt solutions at 298.15 K.

The experimentally derived water activities of alkylaminium sulfates by Clegg et al.[S1]

deviate from the other experiments and all of the COSMO*therm* parametrizations (Figure S1). On the other hand, the water activities of Clegg et al.[S1] are similar to those measured by Sauerwein et al.[S2] for corresponding bisulfates (0.861:1 to 1.0405:1 mole ratio of amines and sulfuric acid). As was hypothesized by Sauerwein et al.,[S2] some of the amines may have evaporated during the experiments, leading to bisulfate solutions.

The comparison between the experiments and COSMO*therm* calculations is shown in Figure S2. The best agreement with experiments is found using the FINE parametrization.

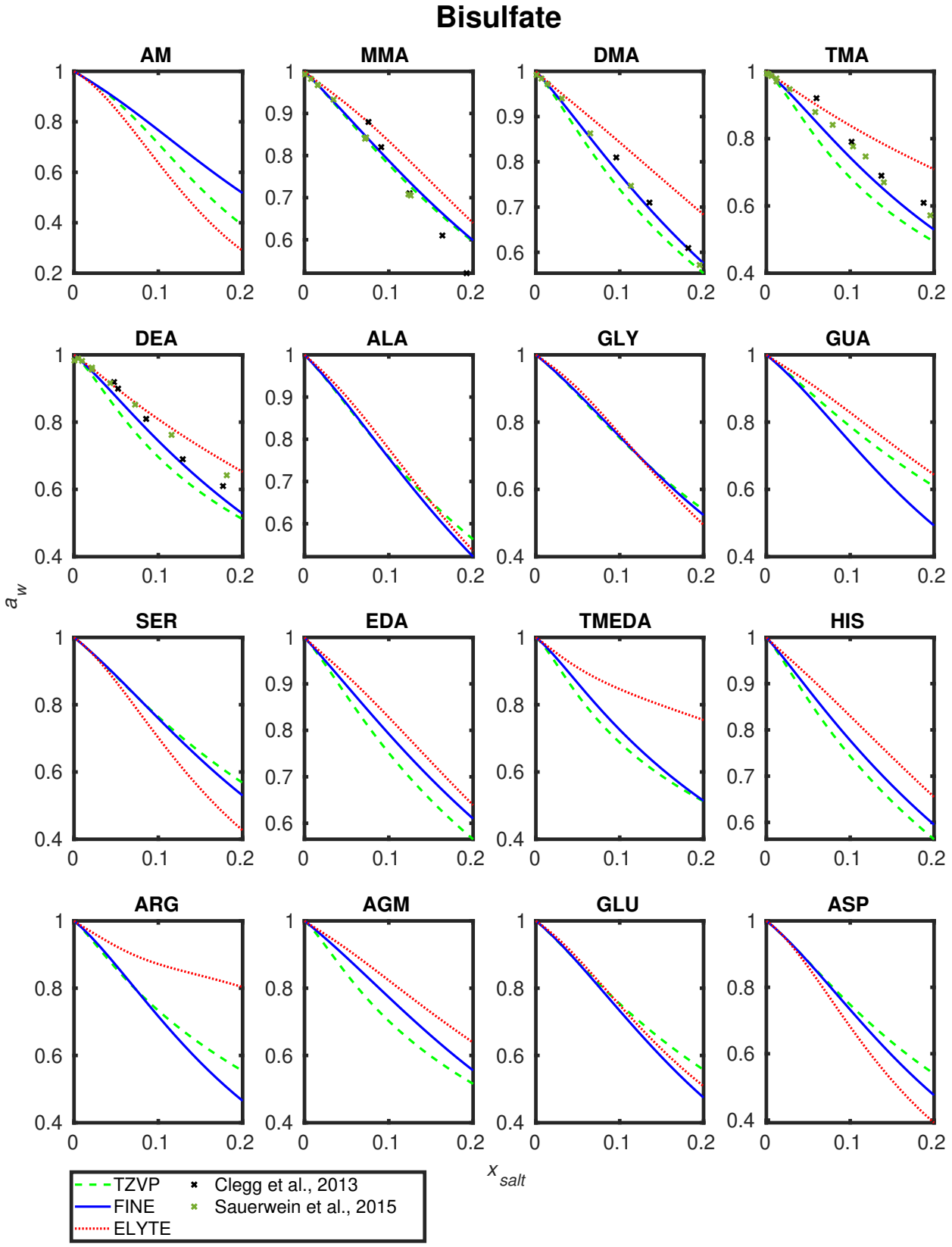

Figure S2: Comparison between experiments[S1,S2] and different COSMO*therm* parametrizations in aqueous bisulfate salt solutions at 298.15 K.

Bonner[S4] measured osmotic coefficients of MMA, DMA and TMA nitrate solutions at 298.15 K. The osmotic coefficients were converted to water activities using the following equation:

$$\ln a_{\mathrm{w}} = -\frac{\phi \nu m}{s} \tag{1}$$

where ϕ is the osmotic coefficient, ν is the number of moles of dissolved ions from 1 mole of salt, m is the molality of the solution and s is the number of moles in 1 kg of water.[S5]

A comparison between the experimentally derived water activities and the three COSMO-*therm* parametrizations is shown in Figure S3.

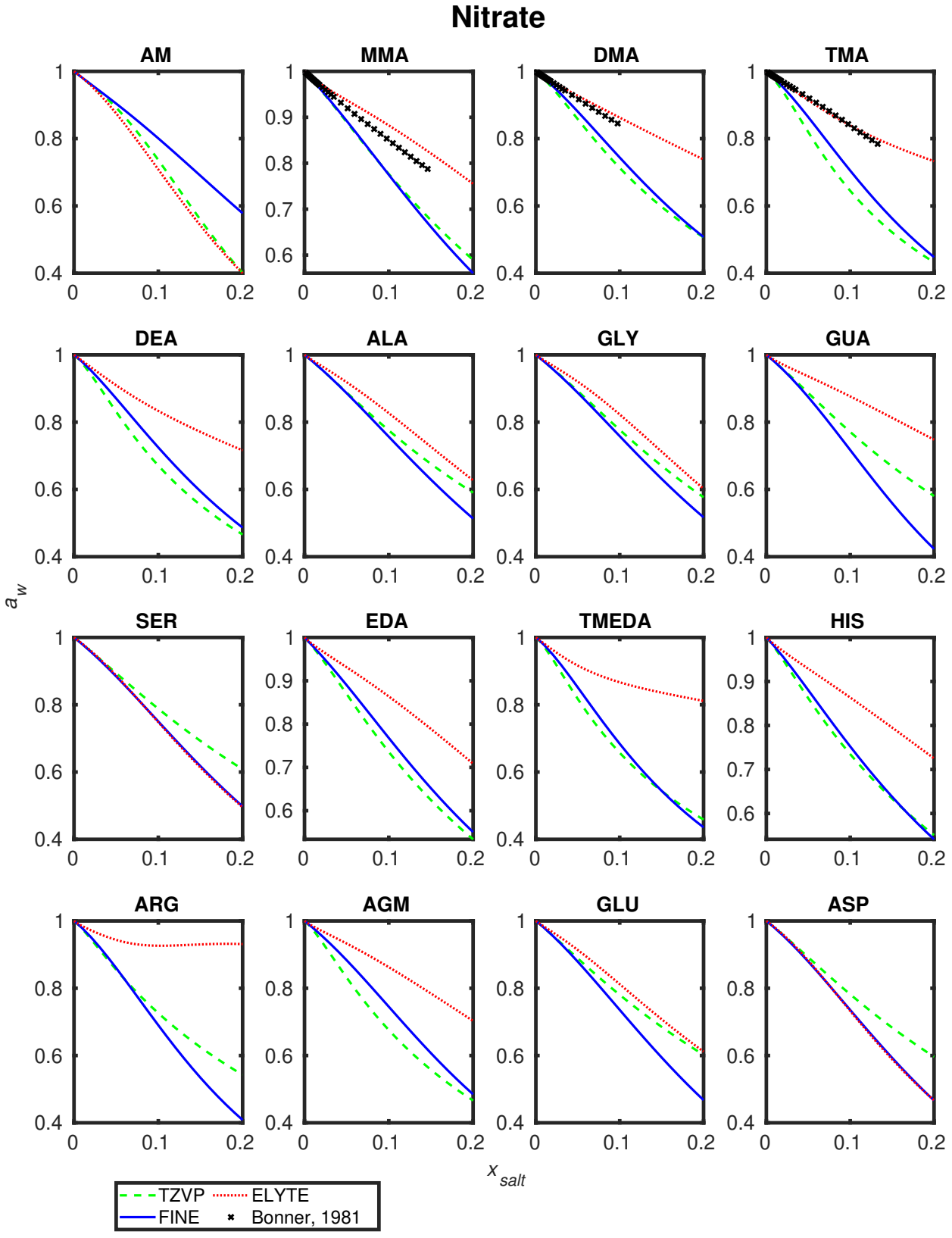

Figure S3: Comparison between COSMO*therm* parametrizations in aqueous nitrate salt solutions at 298.15 K. Experimental water activities are calculated from osmotic coefficients of Bonner.[S4]

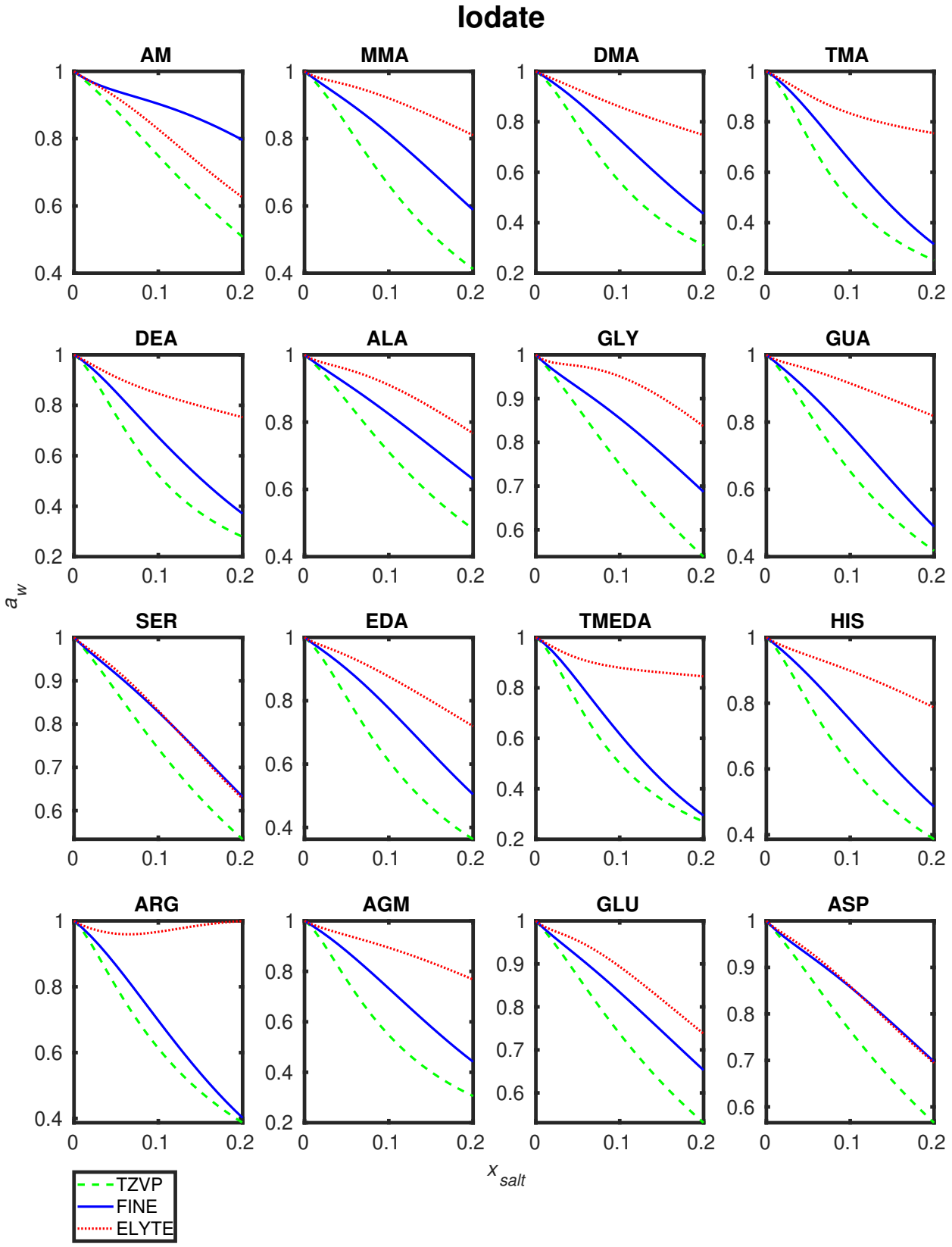

Figure S4: Comparison between COSMO*therm* parametrizations in aqueous iodate salt solutions at 298.15 K.

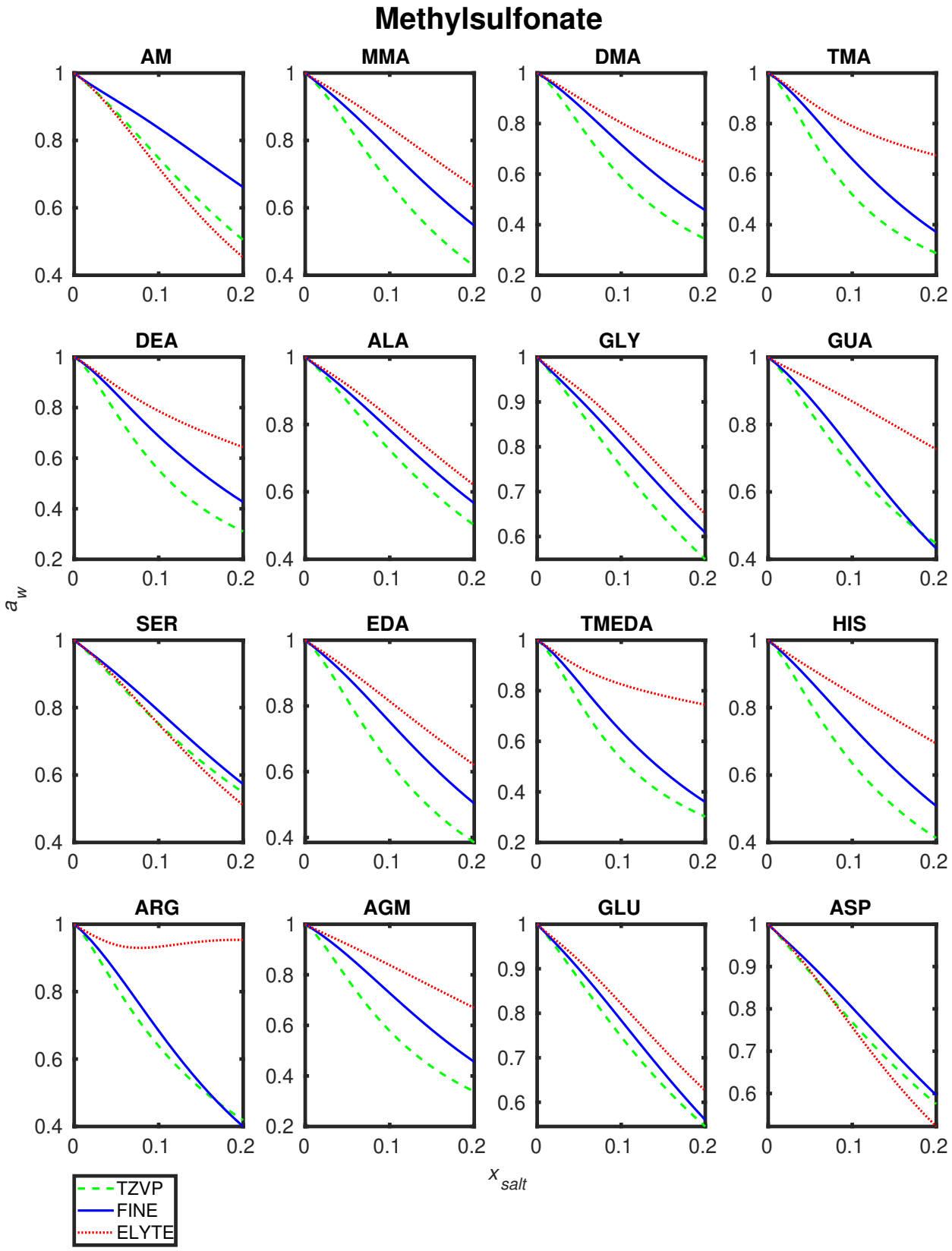

Figure S5: Comparison between COSMO*therm* parametrizations in aqueous methylsulfonate salt solutions at 298.15 K.

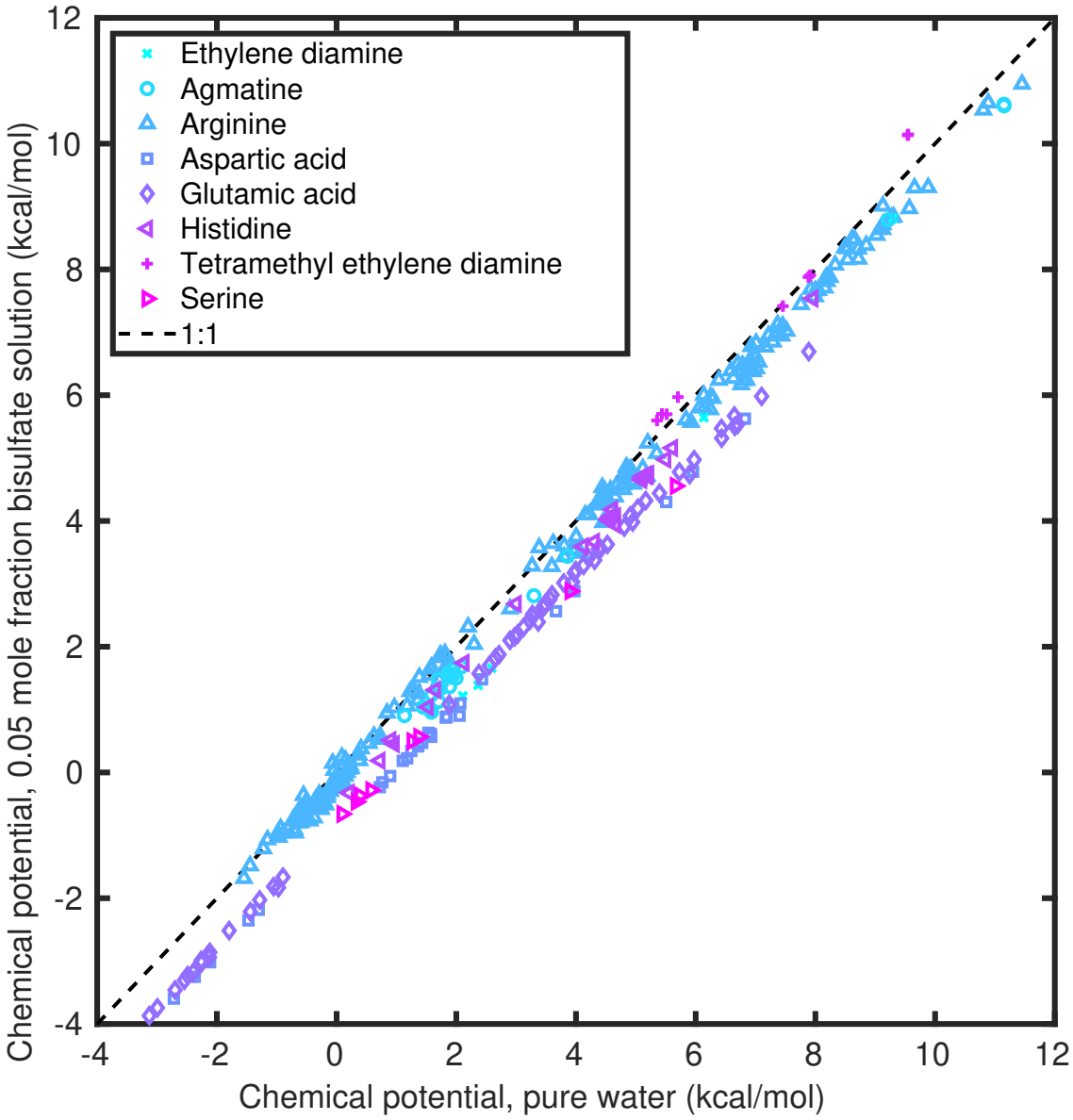

Figure S6: Pseudo-chemical potentials of selected aminium cations in the infinite dilution in water (x-axis) and in 0.05 mole fraction aqueous aminium bisulfate at 298.15 K.

Table S1: COSMO*therm*-derived water activities (a_w) in binary aqueous AM salt solutions (x_{salt} mole fraction of salt) at 298.15 K.

x_{salt}	SO_4^{2-}	HSO_4^-	NO_3^-	IO_3^-	$CH_3SO_3^-$
0.005	0.988937	0.989959	0.990170	0.990789	0.990410
0.01	0.978529	0.979806	0.980547	0.982836	0.981457
0.02	0.957945	0.959041	0.961579	0.969610	0.964750
0.03	0.937696	0.937509	0.942670	0.958851	0.948924
0.04	0.917970	0.915136	0.923577	0.949668	0.933443
0.05	0.898882	0.891944	0.904157	0.941456	0.917986
0.06	0.880468	0.868020	0.884335	0.933786	0.902362
0.07	0.862709	0.843471	0.864076	0.926346	0.886467
0.08	0.845553	0.818399	0.843385	0.918906	0.870251
0.09	0.828933	0.792908	0.822279	0.911293	0.853700
0.1	0.812770	0.767115	0.800793	0.903378	0.836829
0.11	0.796985	0.741141	0.778971	0.895062	0.819671
0.12	0.781497	0.715110	0.756870	0.886274	0.802270
0.13	0.766230	0.689146	0.734553	0.876964	0.784679
0.14	0.751111	0.663362	0.712089	0.867096	0.766955
0.15	0.736074	0.637861	0.689551	0.856653	0.749160
0.16	0.721055	0.612734	0.667010	0.845626	0.731356
0.17	0.705996	0.588062	0.644538	0.834022	0.713602
0.18	0.690846	0.563913	0.622202	0.821850	0.695956
0.19	0.675556	0.540345	0.600068	0.809132	0.678470
0.2	0.660084	0.517401	0.578194	0.795891	0.661190

Table S2: COSMO*therm*-derived water activities (a_w) in binary aqueous MMA salt solutions (x_{salt} mole fraction of salt) at 298.15 K.

x_{salt}	SO_4^{2-}	HSO_4^-	NO_3^-	IO_3^-	$CH_3SO_3^-$
0.005	0.989351	0.989939	0.989965	0.990348	0.990057
0.01	0.982673	0.979757	0.979791	0.981185	0.980126
0.02	0.973589	0.959053	0.958911	0.963629	0.959945
0.03	0.964555	0.937960	0.937324	0.946444	0.939041
0.04	0.952444	0.916572	0.915100	0.929158	0.917290
0.05	0.935895	0.894998	0.892341	0.911487	0.894704
0.06	0.914393	0.873347	0.869169	0.893278	0.871386
0.07	0.887906	0.851717	0.845710	0.874448	0.847477
0.08	0.856701	0.830201	0.822092	0.854964	0.823138
0.09	0.821246	0.808874	0.798437	0.834837	0.798536
0.1	0.782148	0.787804	0.774862	0.814099	0.773839
0.11	0.740105	0.767042	0.751459	0.792804	0.749201
0.12	0.695879	0.746628	0.728336	0.771014	0.724764
0.13	0.650268	0.726593	0.705569	0.748804	0.700650
0.14	0.604085	0.706959	0.683226	0.726251	0.676963
0.15	0.558128	0.687748	0.661363	0.703439	0.653790
0.16	0.513150	0.668972	0.640028	0.680449	0.631196
0.17	0.469826	0.650634	0.619253	0.657367	0.609232
0.18	0.428721	0.632736	0.599065	0.634274	0.587930
0.19	0.390263	0.615276	0.579480	0.611250	0.567311
0.2	0.354732	0.598254	0.560506	0.588371	0.547383

Table S3: COSMO*therm*-derived water activities (a_w) in binary aqueous DMA salt solutions (x_{salt} mole fraction of salt) at 298.15 K.

x_{salt}	SO_4^{2-}	HSO_4^-	NO_3^-	IO_3^-	$CH_3SO_3^-$
0.005	0.986419	0.989802	0.989774	0.989847	0.989666
0.01	0.972624	0.979248	0.979079	0.979305	0.978659
0.02	0.940601	0.957297	0.956364	0.956871	0.954697
0.03	0.900923	0.934537	0.932129	0.932634	0.928431
0.04	0.854075	0.911283	0.906685	0.906746	0.900310
0.05	0.801656	0.887782	0.880332	0.879439	0.870823
0.06	0.745481	0.864238	0.853350	0.850952	0.840426
0.07	0.687220	0.840805	0.826000	0.821530	0.809532
0.08	0.628275	0.817603	0.798514	0.791399	0.778497
0.09	0.569772	0.794730	0.771094	0.760766	0.747617
0.1	0.512607	0.772256	0.743920	0.729821	0.717127
0.11	0.457514	0.750236	0.717142	0.698737	0.687213
0.12	0.405112	0.728704	0.690886	0.667674	0.658014
0.13	0.355939	0.707690	0.665251	0.636778	0.629628
0.14	0.310454	0.687208	0.640317	0.606186	0.602126
0.15	0.269023	0.667268	0.616142	0.576026	0.575549
0.16	0.231888	0.647873	0.592766	0.546414	0.549921
0.17	0.199128	0.629019	0.570214	0.517458	0.525251
0.18	0.170650	0.610701	0.548499	0.489255	0.501535
0.19	0.146196	0.592910	0.527623	0.461890	0.478761
0.2	0.125386	0.575636	0.507578	0.435437	0.456914

Table S4: COSMO*therm*-derived water activities (a_w) in binary aqueous TMA salt solutions (x_{salt} mole fraction of salt) at 298.15 K.

x_{salt}	SO_4^{2-}	HSO_4^-	NO_3^-	IO_3^-	$CH_3SO_3^-$
0.005	0.982559	0.989580	0.989501	0.989249	0.989198
0.01	0.959628	0.978420	0.978062	0.977065	0.976900
0.02	0.899598	0.954404	0.952756	0.948892	0.948479
0.03	0.825962	0.928828	0.924856	0.916570	0.916076
0.04	0.745349	0.902337	0.895063	0.881169	0.880956
0.05	0.663194	0.875416	0.863993	0.843613	0.844190
0.06	0.583208	0.848412	0.832166	0.804676	0.806646
0.07	0.507596	0.821584	0.800021	0.764978	0.768993
0.08	0.437508	0.795122	0.767923	0.725010	0.731717
0.09	0.373481	0.769163	0.736170	0.685164	0.695162
0.1	0.315652	0.743803	0.704997	0.645734	0.659540
0.11	0.264035	0.719108	0.674590	0.606980	0.625011
0.12	0.218550	0.695114	0.645084	0.569118	0.591672
0.13	0.179037	0.671849	0.616587	0.532326	0.559576
0.14	0.145255	0.649325	0.589152	0.496754	0.528755
0.15	0.116869	0.627543	0.562819	0.462539	0.499230
0.16	0.093427	0.606494	0.537606	0.429791	0.471004
0.17	0.074366	0.586167	0.513512	0.398597	0.444070
0.18	0.059061	0.566548	0.490524	0.369020	0.418417
0.19	0.046882	0.547617	0.468616	0.341102	0.394021
0.2	0.037249	0.529355	0.447756	0.314863	0.370856

Table S5: COSMO*therm*-derived water activities (a_w) in binary aqueous DEA salt solutions (x_{salt} mole fraction of salt) at 298.15 K.

x_{salt}	SO_4^{2-}	HSO_4^-	NO_3^-	IO_3^-	$CH_3SO_3^-$
0.005	0.983374	0.989580	0.989578	0.989357	0.989352
0.01	0.962510	0.978413	0.978358	0.977465	0.977475
0.02	0.910198	0.954350	0.953884	0.950359	0.950520
0.03	0.848215	0.928700	0.927306	0.919697	0.920210
0.04	0.781431	0.902151	0.899279	0.886527	0.887629
0.05	0.713399	0.875215	0.870343	0.851761	0.853745
0.06	0.646385	0.848261	0.840944	0.816134	0.819356
0.07	0.581674	0.821541	0.811436	0.780202	0.785080
0.08	0.519911	0.795229	0.782096	0.744358	0.751366
0.09	0.461379	0.769438	0.753142	0.708865	0.718508
0.1	0.406224	0.744238	0.724743	0.673899	0.686678
0.11	0.354591	0.719668	0.697025	0.639571	0.655955
0.12	0.306695	0.695752	0.670080	0.605963	0.626354
0.13	0.262818	0.672496	0.643974	0.573133	0.597847
0.14	0.223261	0.649900	0.618751	0.541145	0.570402
0.15	0.188265	0.627960	0.594435	0.510063	0.543982
0.16	0.157921	0.606666	0.571037	0.479953	0.518550
0.17	0.132113	0.586005	0.548557	0.450881	0.494080
0.18	0.110528	0.565967	0.526984	0.422910	0.470554
0.19	0.092697	0.546538	0.506301	0.396101	0.447959
0.2	0.078076	0.527706	0.486487	0.370503	0.426286

Table S6: COSMO*therm*-derived water activities (a_w) in binary aqueous EDA salt solutions (x_{salt} mole fraction of salt) at 298.15 K.

x_{salt}	SO_4^{2-}	HSO_4^-	NO_3^-	IO_3^-	$CH_3SO_3^-$
0.005	0.988960	0.989964	0.989946	0.990243	0.989991
0.01	0.980908	0.979840	0.979697	0.980739	0.979839
0.02	0.965678	0.959324	0.958505	0.961763	0.958726
0.03	0.946488	0.938501	0.936396	0.942203	0.936281
0.04	0.921232	0.917463	0.913474	0.921673	0.912468
0.05	0.889610	0.896317	0.889888	0.900014	0.887418
0.06	0.852069	0.875167	0.865809	0.877204	0.861350
0.07	0.809411	0.854107	0.841410	0.853289	0.834508
0.08	0.762599	0.833219	0.816860	0.828371	0.807147
0.09	0.712670	0.812567	0.792315	0.802579	0.779517
0.1	0.660688	0.792208	0.767918	0.776060	0.751843
0.11	0.607716	0.772182	0.743791	0.748971	0.724328
0.12	0.554788	0.752520	0.720043	0.721470	0.697146
0.13	0.502884	0.733245	0.696748	0.693718	0.670440
0.14	0.452893	0.714371	0.673987	0.665867	0.644328
0.15	0.405576	0.695909	0.651809	0.638067	0.618898
0.16	0.361529	0.677864	0.630256	0.610458	0.594216
0.17	0.321154	0.660235	0.609355	0.583168	0.570326
0.18	0.284655	0.643021	0.589120	0.556315	0.547256
0.19	0.252044	0.626219	0.569557	0.530005	0.525017
0.2	0.223182	0.609823	0.550666	0.504330	0.503611

Table S7: COSMO*therm*-derived water activities (a_w) in binary aqueous TMEDA salt solutions (x_{salt} mole fraction of salt) at 298.15 K.

x_{salt}	SO_4^{2-}	HSO_4^-	NO_3^-	IO_3^-	$CH_3SO_3^-$
0.005	0.981153	0.989337	0.989265	0.988914	0.988960
0.01	0.954851	0.977513	0.977169	0.975787	0.975984
0.02	0.886056	0.951296	0.949613	0.944366	0.945192
0.03	0.805204	0.922920	0.918725	0.907750	0.909633
0.04	0.720666	0.893539	0.885707	0.867793	0.871168
0.05	0.637427	0.863953	0.851532	0.825963	0.831300
0.06	0.558142	0.834682	0.816952	0.783347	0.791163
0.07	0.484119	0.806048	0.782539	0.740694	0.751547
0.08	0.415976	0.778240	0.748712	0.698504	0.712956
0.09	0.354064	0.751354	0.715776	0.657103	0.675677
0.1	0.298571	0.725429	0.683938	0.616718	0.639855
0.11	0.249564	0.700465	0.653330	0.577511	0.605537
0.12	0.206961	0.676442	0.624028	0.539611	0.572718
0.13	0.170475	0.653333	0.596063	0.503130	0.541368
0.14	0.139709	0.631103	0.569435	0.468170	0.511446
0.15	0.114075	0.609712	0.544122	0.434809	0.482909
0.16	0.092938	0.589124	0.520086	0.403130	0.455730
0.17	0.075642	0.569303	0.497277	0.373183	0.429870
0.18	0.061561	0.550214	0.475639	0.345000	0.405298
0.19	0.050138	0.531823	0.455112	0.318591	0.381977
0.2	0.040894	0.514102	0.435635	0.293943	0.359870

Table S8: COSMO*therm*-derived water activities (a_w) in binary aqueous GUA salt solutions (x_{salt} mole fraction of salt) at 298.15 K.

x_{salt}	SO_4^{2-}	HSO_4^-	NO_3^-	IO_3^-	$CH_3SO_3^-$
0.005	0.985858	0.989748	0.989723	0.989992	0.989781
0.01	0.971851	0.979010	0.978885	0.979906	0.979105
0.02	0.942535	0.956212	0.955578	0.959268	0.956345
0.03	0.910145	0.931877	0.930249	0.937811	0.931745
0.04	0.874087	0.906283	0.903126	0.915421	0.905434
0.05	0.834376	0.879702	0.874457	0.892068	0.877603
0.06	0.791338	0.852398	0.844504	0.867776	0.848472
0.07	0.745466	0.824616	0.813533	0.842606	0.818283
0.08	0.697351	0.796580	0.781813	0.816647	0.787288
0.09	0.647639	0.768497	0.749607	0.790008	0.755739
0.1	0.597011	0.740544	0.717169	0.762810	0.723888
0.11	0.546171	0.712880	0.684741	0.735184	0.691973
0.12	0.495827	0.685639	0.652549	0.707266	0.660221
0.13	0.446687	0.658933	0.620798	0.679194	0.628838
0.14	0.399431	0.632850	0.589672	0.651104	0.598011
0.15	0.354695	0.607467	0.559329	0.623130	0.567902
0.16	0.313031	0.582839	0.529905	0.595400	0.538651
0.17	0.274874	0.559007	0.501508	0.568034	0.510372
0.18	0.240502	0.535998	0.474222	0.541144	0.483154
0.19	0.210022	0.513828	0.448110	0.514831	0.457062
0.2	0.183361	0.492503	0.423210	0.489183	0.432142

Table S9: COSMO*therm*-derived water activities (a_w) in binary aqueous AGM salt solutions (x_{salt} mole fraction of salt) at 298.15 K.

x_{salt}	SO_4^{2-}	HSO_4^-	NO_3^-	IO_3^-	$CH_3SO_3^-$
0.005	0.984768	0.989914	0.989840	0.989830	0.989777
0.01	0.968073	0.979628	0.979327	0.979257	0.979062
0.02	0.930187	0.958449	0.957227	0.956773	0.956109
0.03	0.887240	0.936527	0.933763	0.932545	0.931237
0.04	0.840433	0.913987	0.909071	0.906758	0.904706
0.05	0.790756	0.890967	0.883316	0.879654	0.876829
0.06	0.739021	0.867602	0.856682	0.851482	0.847924
0.07	0.685932	0.844015	0.829355	0.822462	0.818289
0.08	0.632136	0.820322	0.801524	0.792792	0.788194
0.09	0.578250	0.796624	0.773376	0.762655	0.757888
0.1	0.524892	0.773015	0.745089	0.732218	0.727591
0.11	0.472685	0.749575	0.716834	0.701633	0.697494
0.12	0.422262	0.726375	0.688769	0.671044	0.667763
0.13	0.374245	0.703479	0.661038	0.640593	0.638540
0.14	0.329221	0.680942	0.633766	0.610413	0.609942
0.15	0.287699	0.658812	0.607066	0.580632	0.582066
0.16	0.250064	0.637128	0.581029	0.551374	0.554990
0.17	0.216534	0.615923	0.555734	0.522755	0.528777
0.18	0.187142	0.595223	0.531238	0.494879	0.503472
0.19	0.161733	0.575049	0.507589	0.467840	0.479108
0.2	0.140008	0.555416	0.484815	0.441719	0.455706

Table S10: COSMO*therm*-derived water activities (a_w) in binary aqueous ARG salt solutions (x_{salt} mole fraction of salt) at 298.15 K.

x_{salt}	SO_4^{2-}	HSO_4^-	NO_3^-	IO_3^-	$CH_3SO_3^-$
0.005	0.984434	0.989503	0.989454	0.989567	0.989434
0.01	0.966477	0.978105	0.977883	0.978278	0.977790
0.02	0.923574	0.953131	0.952094	0.953353	0.951669
0.03	0.873148	0.925954	0.923437	0.925754	0.922445
0.04	0.817544	0.897244	0.892595	0.896031	0.890876
0.05	0.758810	0.867531	0.860160	0.864685	0.857629
0.06	0.698627	0.837240	0.826649	0.832162	0.823285
0.07	0.638366	0.806720	0.792513	0.798852	0.788344
0.08	0.579139	0.776252	0.758149	0.765090	0.753228
0.09	0.521851	0.746068	0.723900	0.731175	0.718294
0.1	0.467226	0.716356	0.690058	0.697366	0.683843
0.11	0.415830	0.687265	0.656869	0.663891	0.650123
0.12	0.368088	0.658912	0.624531	0.630948	0.617331
0.13	0.324292	0.631387	0.593202	0.598708	0.585622
0.14	0.284609	0.604753	0.563003	0.567314	0.555109
0.15	0.249083	0.579057	0.534018	0.536883	0.525874
0.16	0.217642	0.554326	0.506304	0.507511	0.497964
0.17	0.190111	0.530572	0.479891	0.479271	0.471402
0.18	0.166221	0.507797	0.454787	0.452214	0.446190
0.19	0.145643	0.485992	0.430984	0.426375	0.422312
0.2	0.128009	0.465140	0.408457	0.401771	0.399738

Table S11: COSMO*therm*-derived water activities (a_w) in binary aqueous ALA salt solutions (x_{salt} mole fraction of salt) at 298.15 K.

x_{salt}	SO_4^{2-}	HSO_4^-	NO_3^-	IO_3^-	$CH_3SO_3^-$
0.005	0.990560	0.989785	0.989887	0.990390	0.990113
0.01	0.985955	0.979167	0.979461	0.981294	0.980317
0.02	0.981178	0.956877	0.957647	0.963931	0.960564
0.03	0.976273	0.933426	0.934684	0.947073	0.940282
0.04	0.968329	0.909079	0.910757	0.930306	0.919337
0.05	0.956190	0.884076	0.886056	0.913398	0.897739
0.06	0.939399	0.858618	0.860750	0.896223	0.875564
0.07	0.917843	0.832897	0.835007	0.878719	0.852918
0.08	0.891608	0.807062	0.808977	0.860862	0.829929
0.09	0.860919	0.781253	0.782805	0.842657	0.806729
0.1	0.826110	0.755585	0.756620	0.824122	0.783434
0.11	0.787618	0.730156	0.730533	0.805293	0.760164
0.12	0.745983	0.705051	0.704649	0.786208	0.737028
0.13	0.701850	0.680337	0.679058	0.766914	0.714128
0.14	0.655965	0.656069	0.653840	0.747461	0.691551
0.15	0.609164	0.632296	0.629063	0.727899	0.669373
0.16	0.562345	0.609055	0.604783	0.708279	0.647657
0.17	0.516415	0.586375	0.581047	0.688651	0.626454
0.18	0.472230	0.564278	0.557893	0.669064	0.605804
0.19	0.430519	0.542782	0.535352	0.649564	0.585736
0.2	0.391824	0.521899	0.513445	0.630195	0.566269

Table S12: COSMO*therm*-derived water activities (a_w) in binary aqueous GLY salt solutions (x_{salt} mole fraction of salt) at 298.15 K.

x_{salt}	SO_4^{2-}	HSO_4^-	NO_3^-	IO_3^-	$CH_3SO_3^-$
0.005	0.992018	0.989830	0.989923	0.990612	0.990277
0.01	0.990540	0.979325	0.979580	0.982103	0.980921
0.02	0.994755	0.957370	0.958008	0.966725	0.962671
0.03	1.001150	0.934281	0.935319	0.952609	0.944484
0.04	1.005936	0.910248	0.911650	0.939071	0.926027
0.05	1.007407	0.885459	0.887165	0.925694	0.907162
0.06	1.004718	0.860137	0.862034	0.912217	0.887853
0.07	0.997470	0.834478	0.836424	0.898478	0.868139
0.08	0.985535	0.808652	0.810504	0.884380	0.848071
0.09	0.968974	0.782814	0.784423	0.869867	0.827727
0.1	0.947989	0.757091	0.758320	0.854917	0.807195
0.11	0.922901	0.731592	0.732319	0.839532	0.786562
0.12	0.894127	0.706412	0.706533	0.823726	0.765917
0.13	0.862173	0.681628	0.681060	0.807530	0.745343
0.14	0.827612	0.657311	0.655984	0.790978	0.724915
0.15	0.791073	0.633515	0.631377	0.774112	0.704701
0.16	0.753215	0.610284	0.607299	0.756979	0.684761
0.17	0.714701	0.587656	0.583798	0.739626	0.665146
0.18	0.676173	0.565657	0.560912	0.722100	0.645901
0.19	0.638218	0.544306	0.538670	0.704451	0.627060
0.2	0.601343	0.523614	0.517092	0.686725	0.608651

Table S13: COSMO*therm*-derived water activities (a_w) in binary aqueous SER salt solutions (x_{salt} mole fraction of salt) at 298.15 K.

x_{salt}	SO_4^{2-}	HSO_4^-	NO_3^-	IO_3^-	$CH_3SO_3^-$
0.005	0.990017	0.989821	0.989857	0.990420	0.990148
0.01	0.984274	0.979297	0.979369	0.981437	0.980467
0.02	0.976147	0.957317	0.957338	0.964485	0.961157
0.03	0.966953	0.934271	0.934023	0.948168	0.941534
0.04	0.954109	0.910370	0.909605	0.931978	0.921385
0.05	0.936721	0.885819	0.884276	0.915626	0.900654
0.06	0.914589	0.860811	0.858223	0.898949	0.879362
0.07	0.887857	0.835516	0.831628	0.881863	0.857578
0.08	0.856864	0.810095	0.804664	0.864335	0.835395
0.09	0.822075	0.784685	0.777485	0.846364	0.812915
0.1	0.784039	0.759406	0.750248	0.827974	0.790246
0.11	0.743365	0.734363	0.723083	0.809202	0.767493
0.12	0.700714	0.709641	0.696116	0.790098	0.744756
0.13	0.656774	0.685314	0.669454	0.770715	0.722129
0.14	0.612252	0.661445	0.643192	0.751111	0.699705
0.15	0.567853	0.638081	0.617413	0.731347	0.677552
0.16	0.524253	0.615261	0.592188	0.711481	0.655738
0.17	0.482073	0.593017	0.567577	0.691574	0.634319
0.18	0.441850	0.571374	0.543625	0.671680	0.613343
0.19	0.404011	0.550347	0.520369	0.651855	0.592848
0.2	0.368853	0.529947	0.497839	0.632153	0.572864

Table S14: COSMO*therm*-derived water activities (a_w) in binary aqueous ASP salt solutions (x_{salt} mole fraction of salt) at 298.15 K.

x_{salt}	SO_4^{2-}	HSO_4^-	NO_3^-	IO_3^-	$CH_3SO_3^-$
0.005	0.993241	0.989690	0.989812	0.990687	0.990293
0.01	0.992962	0.978801	0.979144	0.982335	0.980952
0.02	0.997137	0.955496	0.956411	0.967347	0.962652
0.03	1.000232	0.930458	0.932023	0.953614	0.944278
0.04	0.998988	0.904021	0.906233	0.940411	0.925496
0.05	0.992245	0.876500	0.879290	0.927321	0.906186
0.06	0.979669	0.848206	0.851430	0.914100	0.886341
0.07	0.961332	0.819422	0.822895	0.900602	0.866002
0.08	0.937544	0.790395	0.793908	0.886753	0.845252
0.09	0.908773	0.761339	0.764674	0.872520	0.824186
0.1	0.875598	0.732427	0.735380	0.857898	0.802903
0.11	0.838679	0.703812	0.706195	0.842903	0.781495
0.12	0.798739	0.675623	0.677263	0.827564	0.760057
0.13	0.756535	0.647970	0.648708	0.811916	0.738672
0.14	0.712841	0.620947	0.620635	0.796000	0.717420
0.15	0.668426	0.594625	0.593131	0.779859	0.696370
0.16	0.624019	0.569061	0.566268	0.763539	0.675582
0.17	0.580295	0.544298	0.540103	0.747083	0.655109
0.18	0.537844	0.520365	0.514681	0.730533	0.634994
0.19	0.497152	0.497277	0.490036	0.713931	0.615275
0.2	0.458595	0.475044	0.466193	0.697315	0.595978

Table S15: COSMO*therm*-derived water activities (a_w) in binary aqueous GLU salt solutions (x_{salt} mole fraction of salt) at 298.15 K.

x_{salt}	SO_4^{2-}	HSO_4^-	NO_3^-	IO_3^-	$CH_3SO_3^-$
0.005	0.992005	0.989661	0.989782	0.990506	0.990156
0.01	0.989215	0.978706	0.979050	0.981677	0.980450
0.02	0.986542	0.955249	0.956154	0.965100	0.960921
0.03	0.981446	0.930116	0.931627	0.949204	0.940858
0.04	0.971315	0.903678	0.905766	0.933482	0.920097
0.05	0.955383	0.876231	0.878829	0.917660	0.898628
0.06	0.933585	0.848060	0.851045	0.901595	0.876525
0.07	0.906198	0.819423	0.822639	0.885215	0.853893
0.08	0.873689	0.790547	0.793819	0.868492	0.830845
0.09	0.836651	0.761637	0.764771	0.851432	0.807505
0.1	0.795773	0.732863	0.735674	0.834060	0.783997
0.11	0.751814	0.704370	0.706681	0.816412	0.760435
0.12	0.705588	0.676277	0.677929	0.798532	0.736928
0.13	0.657946	0.648682	0.649536	0.780468	0.713573
0.14	0.609751	0.621670	0.621604	0.762269	0.690456
0.15	0.561851	0.595304	0.594217	0.743984	0.667652
0.16	0.515045	0.569638	0.567447	0.725663	0.645226
0.17	0.470040	0.544713	0.541348	0.707352	0.623232
0.18	0.427420	0.520561	0.515967	0.689094	0.601714
0.19	0.387618	0.497204	0.491338	0.670930	0.580706
0.2	0.350904	0.474655	0.467488	0.652898	0.560236

Table S16: COSMO*therm*-derived water activities (a_w) in binary aqueous HIS salt solutions (x_{salt} mole fraction of salt) at 298.15 K.

x_{salt}	SO_4^{2-}	HSO_4^-	NO_3^-	IO_3^-	$CH_3SO_3^-$
0.005	0.987957	0.989747	0.989709	0.989929	0.989780
0.01	0.976969	0.979066	0.978866	0.979619	0.979098
0.02	0.951820	0.956817	0.955805	0.958024	0.956358
0.03	0.920836	0.933883	0.931374	0.935054	0.932005
0.04	0.884685	0.910694	0.906048	0.910810	0.906397
0.05	0.844599	0.887539	0.880216	0.885476	0.879897
0.06	0.801752	0.864613	0.854187	0.859259	0.852838
0.07	0.757130	0.842047	0.828209	0.832354	0.825502
0.08	0.711547	0.819923	0.802474	0.804945	0.798124
0.09	0.665672	0.798294	0.777133	0.777198	0.770893
0.1	0.620066	0.777190	0.752298	0.749266	0.743965
0.11	0.575208	0.756626	0.728052	0.721290	0.717462
0.12	0.531511	0.736606	0.704455	0.693395	0.691480
0.13	0.489337	0.717128	0.681548	0.665698	0.666090
0.14	0.448989	0.698182	0.659355	0.638306	0.641348
0.15	0.410723	0.679759	0.637887	0.611315	0.617293
0.16	0.374735	0.661844	0.617146	0.584811	0.593950
0.17	0.341164	0.644421	0.597126	0.558871	0.571334
0.18	0.310087	0.627477	0.577814	0.533565	0.549452
0.19	0.281523	0.610995	0.559195	0.508951	0.528305
0.2	0.255437	0.594960	0.541248	0.485077	0.507886

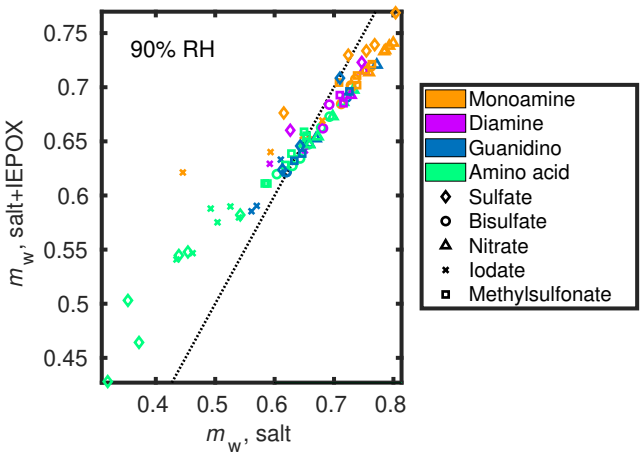

Figure S7: COSMO*therm*-derived equilibrium water contents (m_w in mass fraction) in 90% RH at 298.15 K. The black dotted lines represent the 1:1 line.

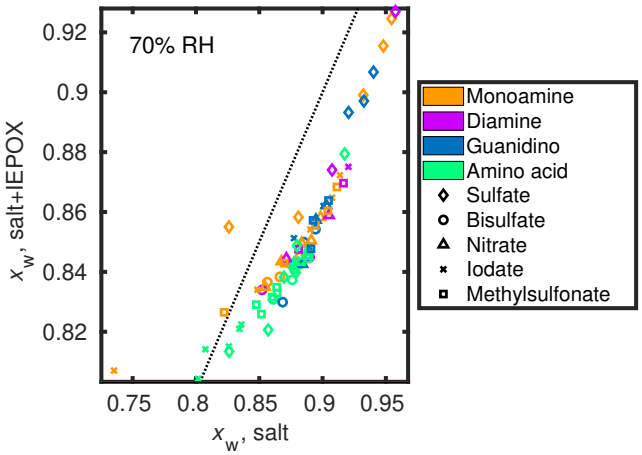

Figure S8: COSMO*therm*-derived equilibrium water contents (x_w in mole fraction) in 70% RH at 298.15 K. The black dotted lines represent the 1:1 line.

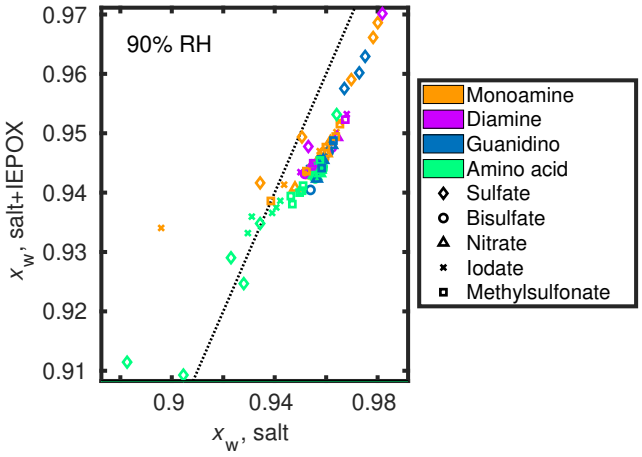

Figure S9: COSMO*therm*-derived equilibrium water contents (x_w in mole fraction) in 90% RH at 298.15 K. The black dotted lines represent the 1:1 line.

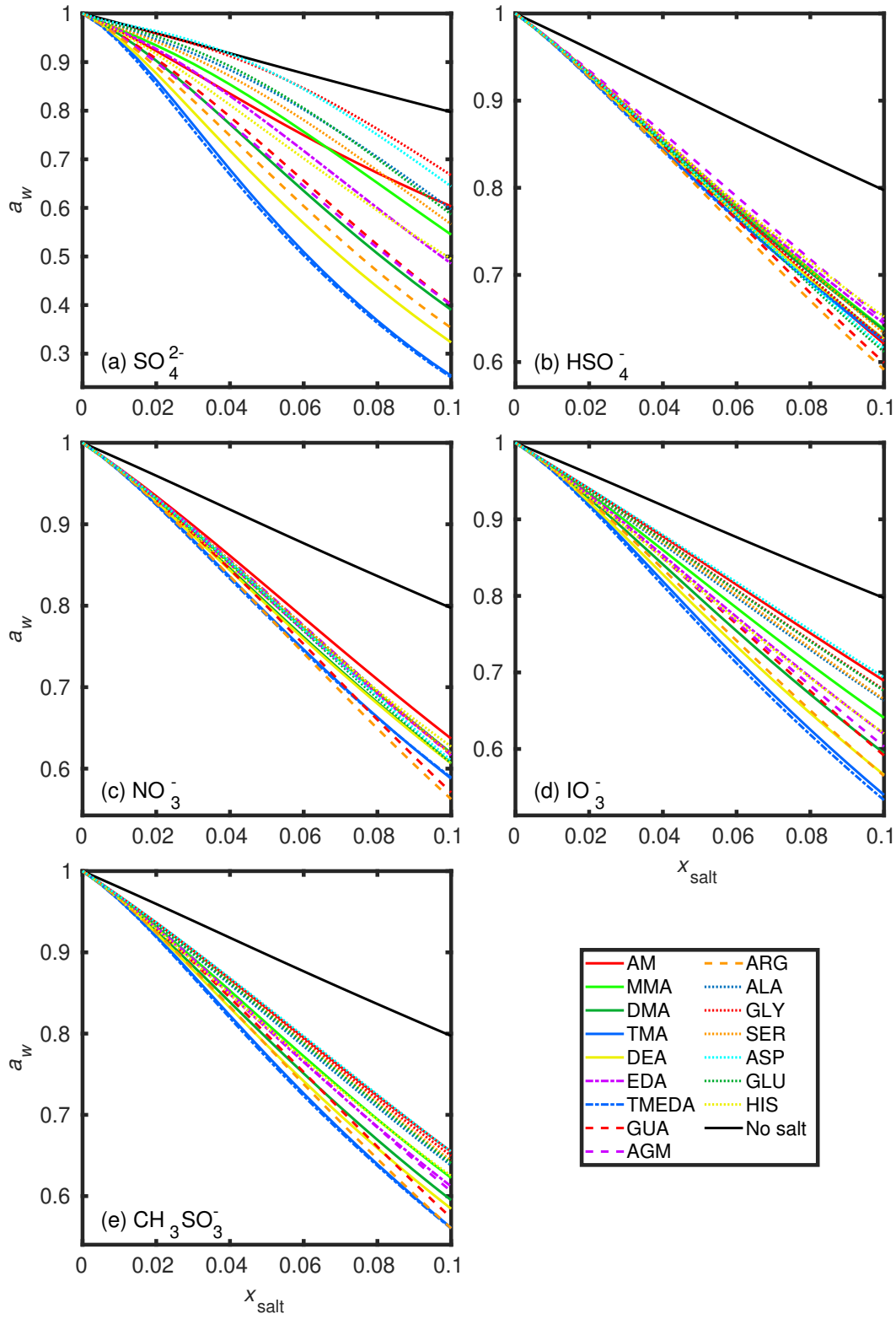

Figure S10: COSMO*therm*-derived water activities in (a) sulfate, (b) bisulfate, (c) nitrate, (d) iodate and (e) methylsulfonate in the presence of IEPOX at 298.15 K. The mole fraction of IEPOX is equal to x_{salt}, the remaining mole fraction is water. "No salt" contains only water and IEPOX with a mole fraction of $2 \times x_{\text{salt}}$. Activities were calculated in 0.01 salt mole fraction intervals and plotted as lines for clarity.

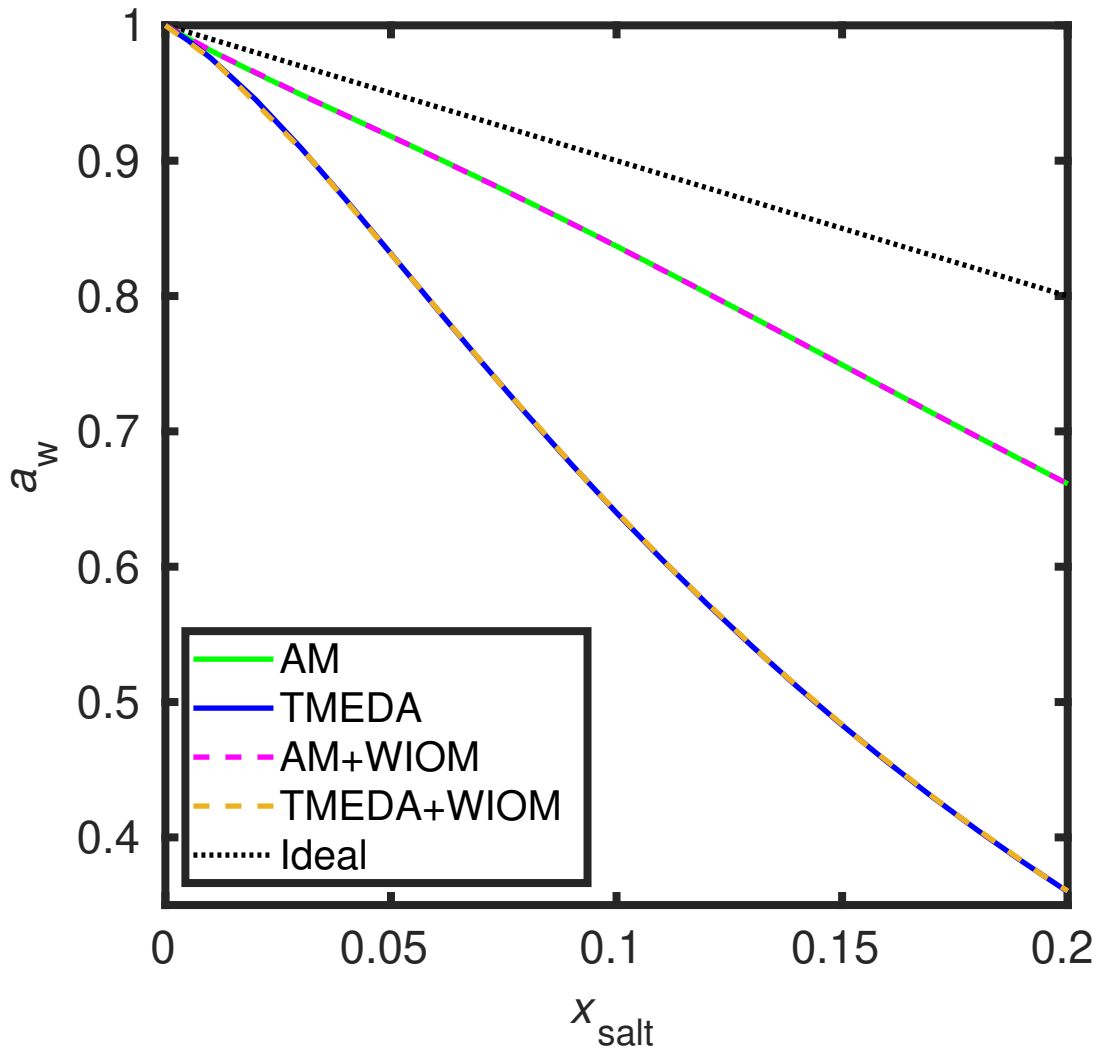

Figure S11: COSMO*therm*-predicted water activities a_w in methylsulfonate solutions with and without WIOM. The amount of WIOM at each salt mole fraction is mole fraction of WIOM at LLE computed using COSMO*therm*.

References

(S1) Clegg, S. L.; Qiu, C.; Zhang, R. The deliquescence behaviour, solubilities, and densities of aqueous solutions of five methyl-and ethyl-aminium sulphate salts. *Atmos. Environ.* **2013**, *73*, 145–158.

(S2) Sauerwein, M.; Clegg, S. L.; Chan, C. K. Water Activities and Osmotic Coefficients of

47 Aqueous Solutions of Five Alkylaminium Sulfates and Their Mixtures with H_2SO_4 at
48 25°C. *Aerosol Sci. Technol.* **2015**, *49*, 566–579.

49 (S3) Rovelli, G.; Miles, R. E. H.; Reid, J. P.; Clegg, S. L. Hygroscopic properties of aminium
50 sulfate aerosols. *Atmos. Chem. Phys.* **2017**, *17*, 4369–4385.

51 (S4) Bonner, O. D. Osmotic and activity coefficients of methyl-substituted ammonium ni-
52 trates at 298.15 K. *J. Chem. Eng. Data* **1981**, *26*, 148–149.

53 (S5) Pitzer, K. S.; Press, C. *Activity coefficients in electrolyte solutions*; CRC press Boca
54 Raton, FL, 1991; Vol. 2; p 25.