Supporting Information:

Dielectric Decrement for Aqueous NaCl

Solutions: The Effect of Ionic Charge Scaling in

Non-Polarizable Water Force Fields

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1 Force Field Parameters

Table S1: Well depth parameters of Lennard-Jones potentials (γ in kJ/mol) used for various interaction pairs in the different force fields.

γ (kJ/mol)		O	H	Na$^+$	Cl$^-$
O	CC	0.6502	0.0	0.5215	0.5215
	BPCEAV	0.7749	0.0	0.7933	0.0619
	MP	0.7749	0.0	0.5036	0.2848
	MPS	0.7749	0.0	0.5036	0.2848
H	CC	0.0	0.0	0.0	0.0
	BPCEAV	0.0	0.0	0.0	0.0
	MP	0.0	0.0	0.0	0.0
	MPS	0.0	0.0	0.0	0.0
Na$^+$	CC	0.5215	0.0	0.4184	0.4184
	BPCEAV	0.7933	0.0	1.4723	1.4388
	MP	0.5036	0.0	0.3274	0.1851
	MPS	0.5036	0.0	0.3274	0.1851
Cl$^-$	CC	0.5215	0.0	0.4184	0.4184
	BPCEAV	0.0619	0.0	1.4388	0.0769
	MP	0.2848	0.0	0.1851	0.1047
	MPS	0.2848	0.0	0.1851	0.1047

Table S2: Distance parameters of Lennard-Jones potentials (σ in Å) used for various interaction pairs in the different force fields.

σ (Å)		O	H	Na$^+$	Cl$^-$
O	CC	3.169	0.0	2.8610	3.7341
	BPCEAV	3.1589	0.0	2.5133	4.2686
	MP	3.1589	1.5794	2.8279	3.8854
	MPS	3.1589	1.5794	2.8279	3.8854
H	CC	0.0	0.0	0.0	0.0
	BPCEAV	0.0	0.0	0.0	0.0
	MP	1.5794	0.0	1.2485	2.306
	MPS	1.5794	0.0	1.2485	2.306
Na$^+$	CC	2.8610	0.0	2.5830	3.7123
	BPCEAV	2.5133	0.0	2.2173	2.9051
	MP	2.8279	1.2485	2.4970	3.5545
	MPS	2.8279	1.2485	2.4970	3.5545
Cl$^-$	CC	3.7341	0.0	2.5830	4.4000
	BPCEAV	4.2686	0.0	2.9051	4.8496
	MP	3.8854	2.306	3.5545	4.6120
	MPS	3.8854	2.306	3.5545	4.6120

Table S3: Charges of hydrogen (H) and oxygen (O) atoms as well as sodium (Na$^+$) and chloride (Cl$^-$) ions used in the different force field parameters sets. The charges are given in units of elementary charge.

	q_O	q_H	q_{Na^+}	q_{Cl^+}
CC	-0.8476	0.4238	1.0	-1.0
BPCEAV	-1.1128	0.5564	0.85	-0.85
MP	-1.1128	0.5564	1.0	-1.0
MPS	-1.1128	0.5564	0.85	-0.85

2 Average Dipole Vector Alignment around Cl^- Ion

For the Cl^- ion, the average dipole vector alignment $P_{\text{Cl}^-}^{\text{r}}(r)$ could also be quantified based on the distance to the closest hydrogen atom

$$\hat{\vec{r}}_{ij}^{\,\text{Cl}^-\text{H}} = \min_{i\in\{1,2\}} \frac{\vec{r}_i^{\,\text{Cl}^-} - \vec{r}_j^{\,\text{H}_i}}{|\vec{r}_i^{\,\text{Cl}^-} - \vec{r}_j^{\,\text{H}_i}|} \tag{1}$$

instead of the distance to the oxygen atom of a surrounding water molecule. This results in

$$P_{\text{Cl}^-}^{\text{r}}(r) = \frac{1}{|\vec{p}_{\text{H}_2\text{O}}|}\,\frac{1}{N_{\text{Cl}^-}N_{\text{H}_2\text{O}}} \left\langle \sum_{i=1}^{N_{\text{Cl}^-}} \sum_{j=1}^{N_{\text{H}_2\text{O}}} \vec{p}_j \cdot \hat{\vec{r}}_{ij}^{\,\text{Cl}^-\text{O}}\, \delta(|\hat{\vec{r}}_{ij}^{\,\text{Cl}^-\text{H}}| - r) \right\rangle \tag{2}$$

instead of eq 12 that is used to obtain the bottom panels of Figures 6 and 7 in Section 3.5 of the main document. Figures S1 and S2 show the results of this substitution. Obviously, the peak positions change because the distances to the water molecules in the solvation shells are now measured differently – which is likewise reflected in the corresponding Cl^--hydrogen radial distribution functions (RDFs). The peak heights on the other hand are hardly affected. Also, as for the case in which we used the Cl^--oxygen in the main text, we observe a plateau regain at small r in which all force fields give the same polarization. This is again followed by a region in which MP-S shows a smaller polarization than MP and is very much in line with $P^{\text{r; MP rescaled}} = 0.85 \cdot P^{\text{r; MP}}$. Consequently, the conclusions from the analysis in Section 3.5 remain unchanged.
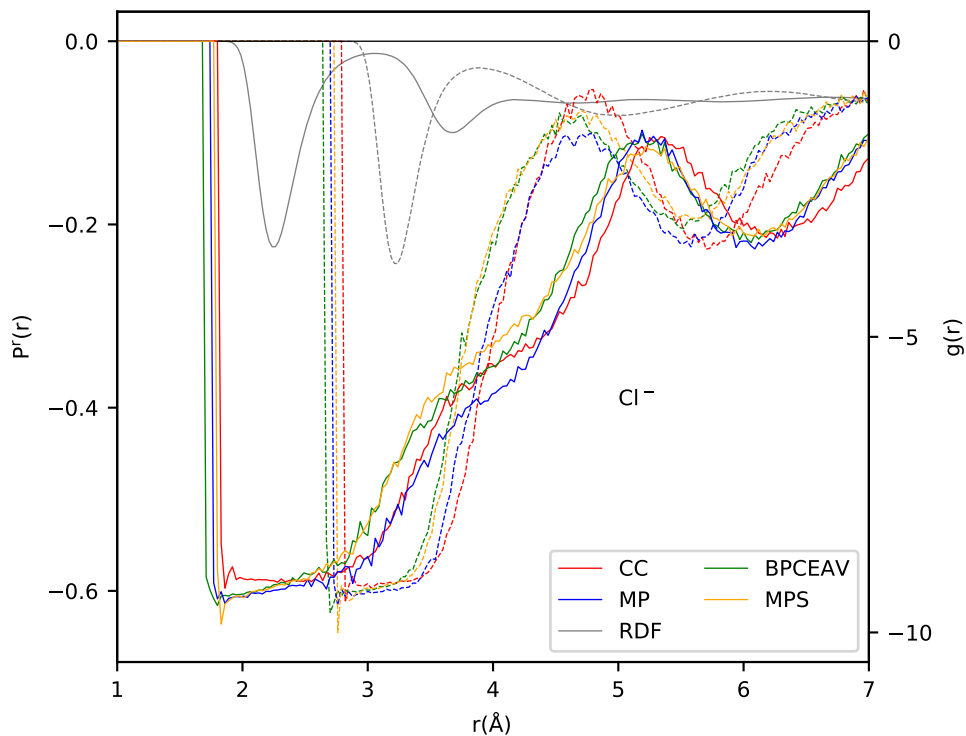
Figure S1: Average alignment of dipole vectors $P^{\mathrm{r}}(r)$ around Cl^- ions. The dashed lines are based on eq 12 in the main document and are thus identical to those of same color shown in the bottom panel of Figure 6. The corresponding solid lines have been obtained from eq 2 above. The Cl^--oxygen (Cl^--hydrogen) radial distribution functions (RDFs) for the CC force field as a representative example are shown as dashed (solid) gray lines. All results are for $0.1\,\mathrm{M}$ solutions to avoid ion-ion interactions.
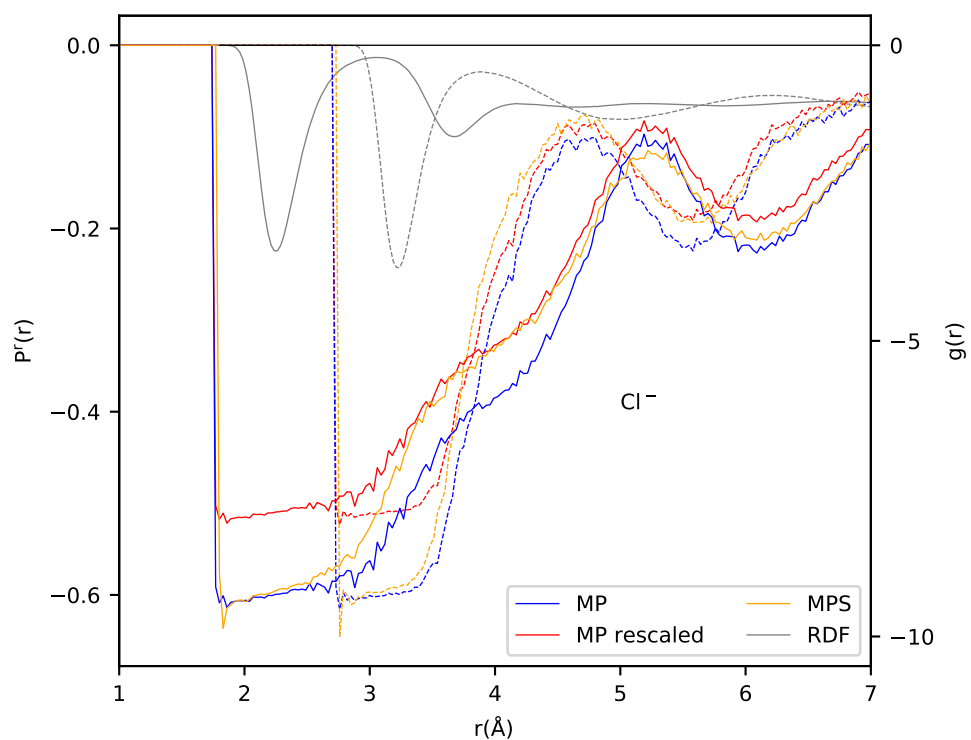
Figure S2: Same as Figure S1 for MP and MP-S, but also $P^{\mathrm{r;\ MP\ rescaled}} = 0.85 \cdot P^{\mathrm{r;\ MP}}$ for comparison – like in the bottom panel of Figure 7 in the main document.