

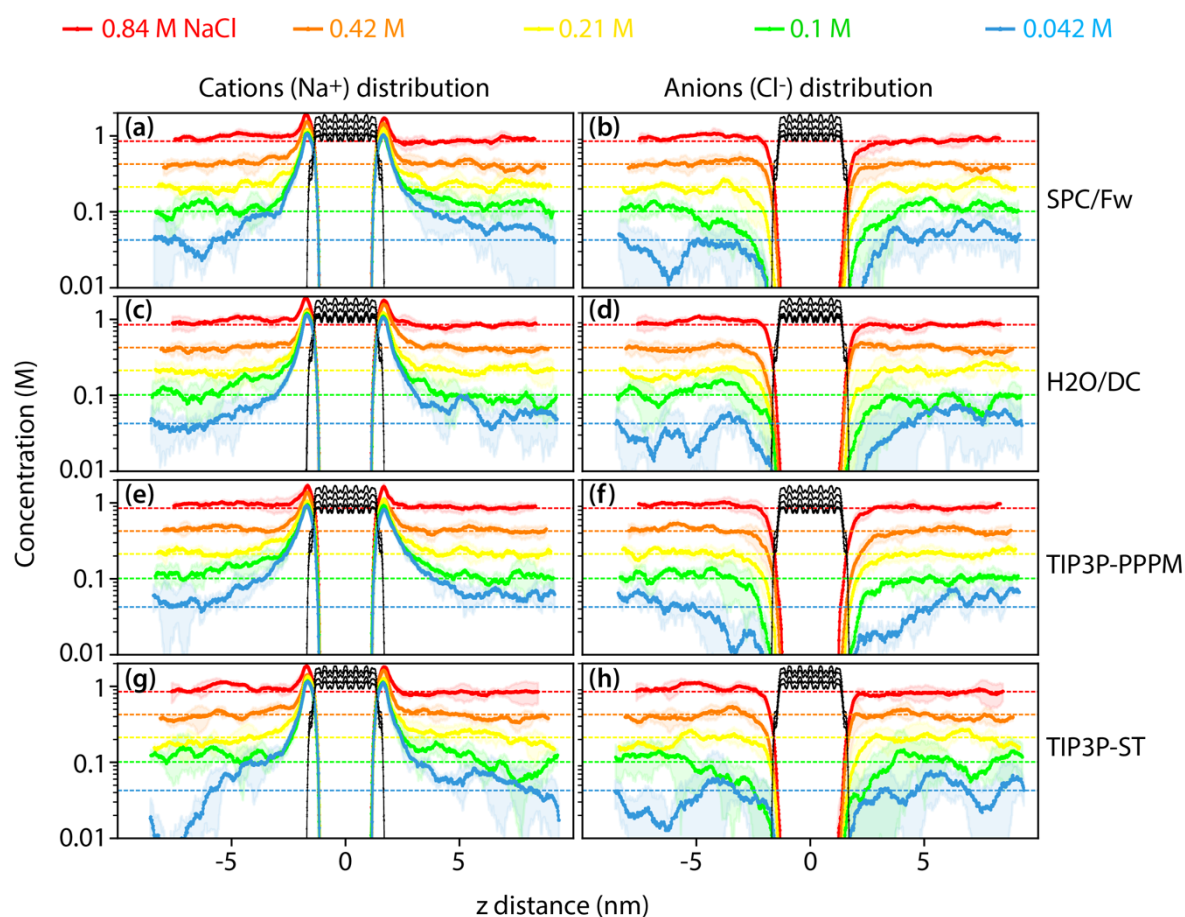
## Supplementary Information

### Comparative molecular dynamics simulations of charged solid-liquid interfaces with different water models

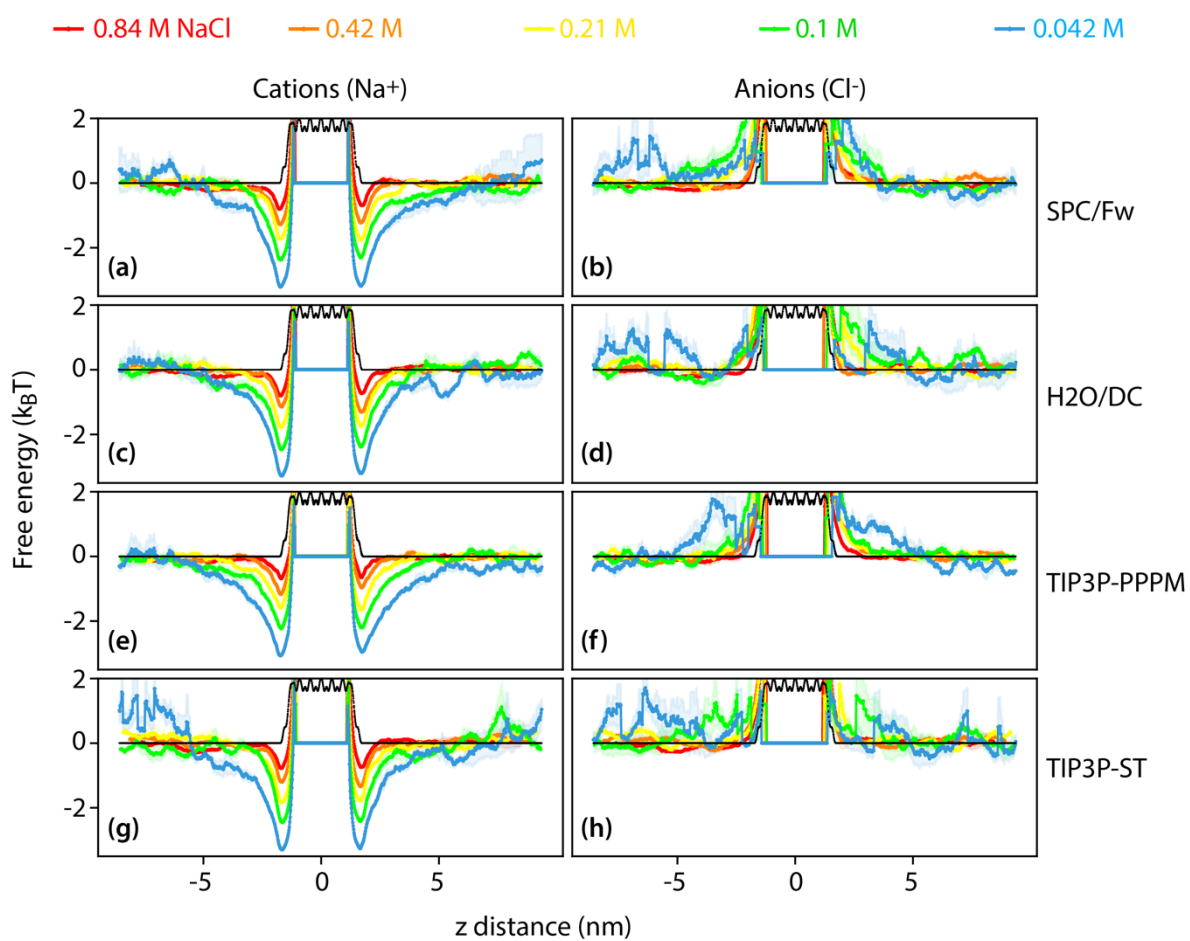
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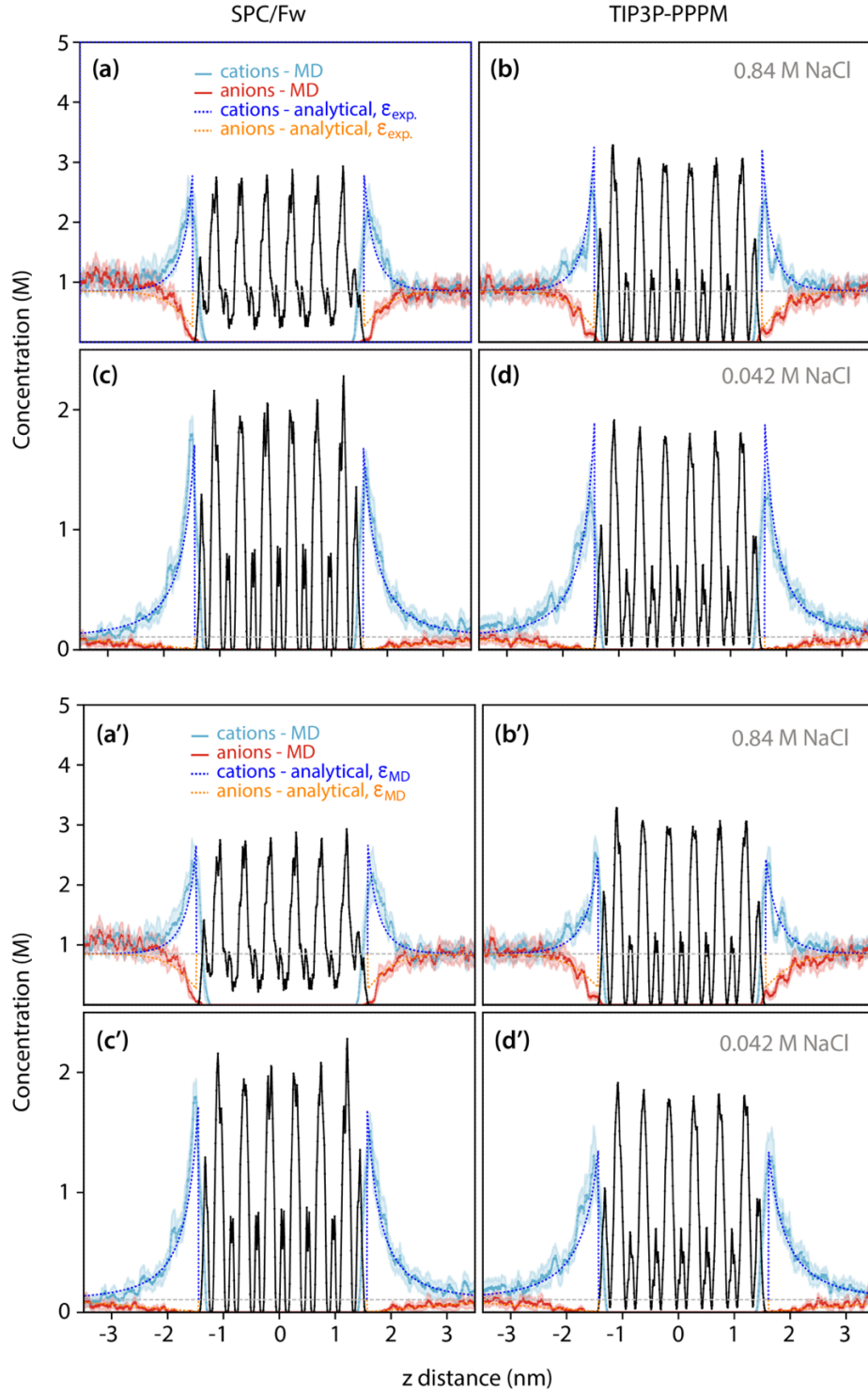
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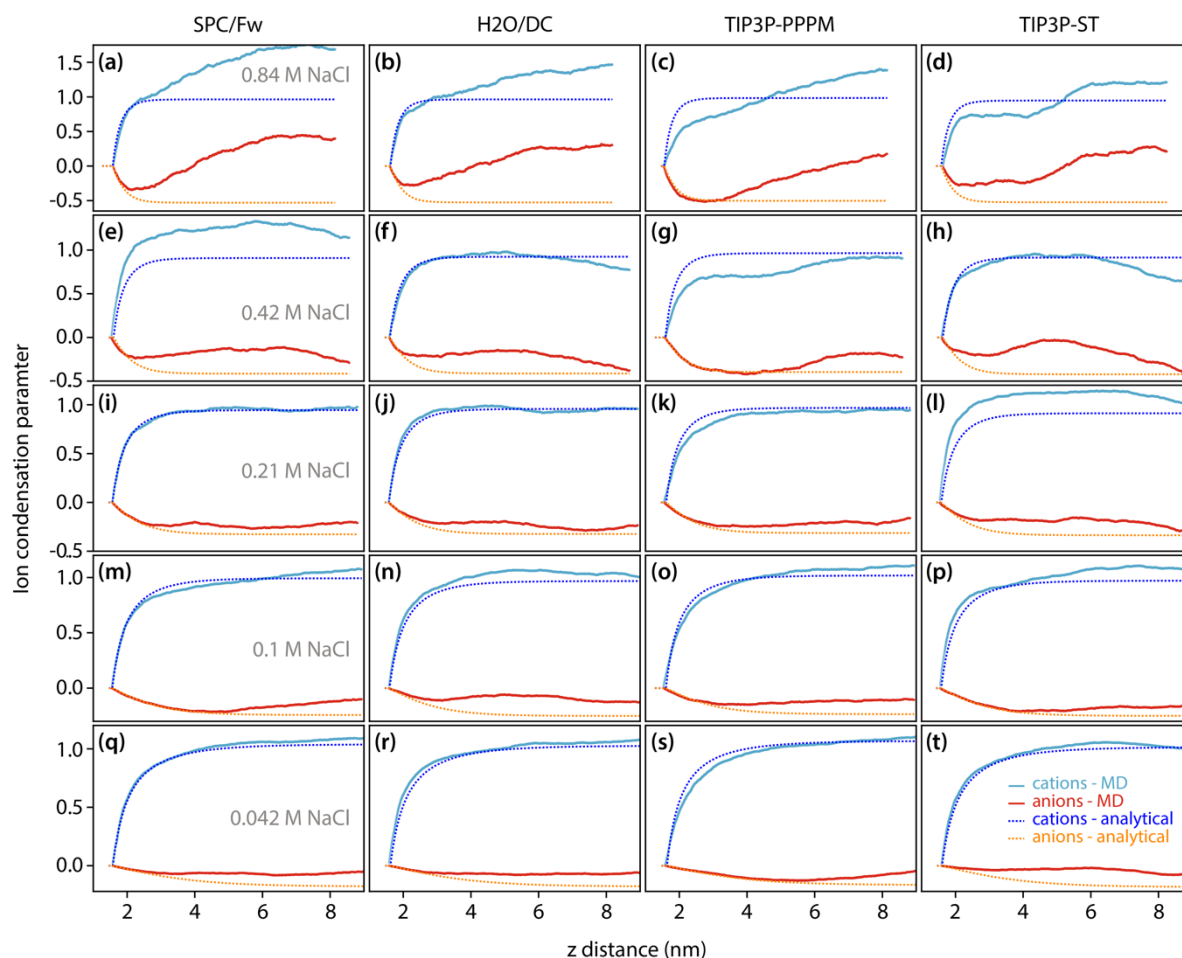
**Figure S1:** Effect of the salt concentration on the ion distribution near solid-liquid interface for the selected water models. The cation and anion distribution near a solvated silica surface is shown for SPC/Fw (a) and (b), respectively, H<sub>2</sub>O/DC (c) and (d), TIP3P-PPPM (e) and (f), and TIP3P-ST (g) and (h). The different colours represent the different bulk ionic concentrations (horizontal dashed line) with the silica density appearing in black.



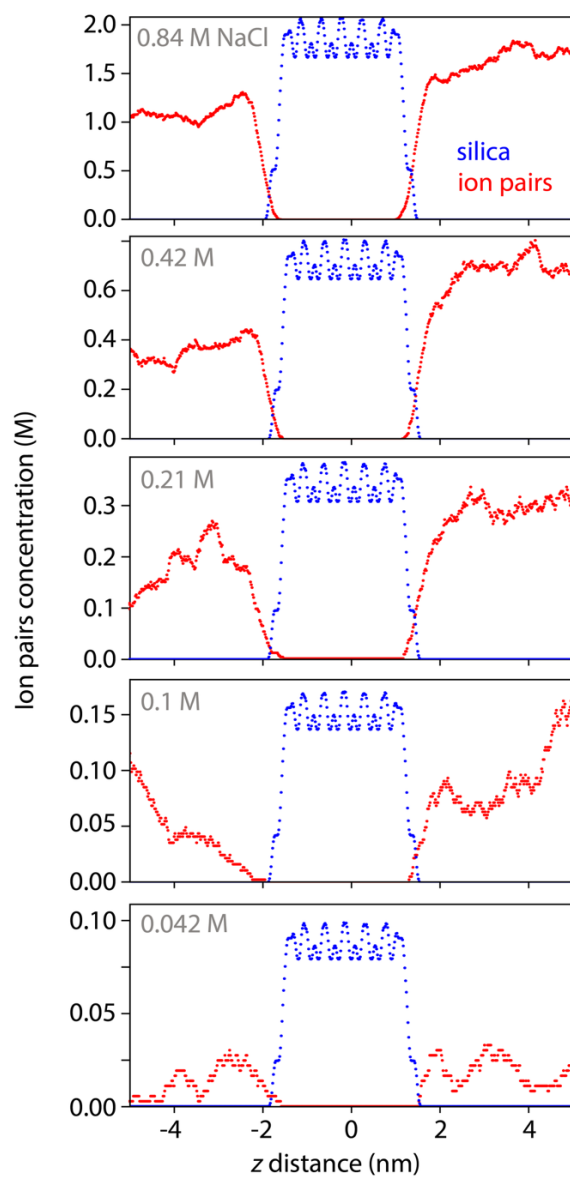
**Figure S2:** Effect of the salt concentration on the free energy of the ions near solid-liquid interface for the selected water models. The cation and anion free energy distribution near a solvated silica surface is shown for SPC/Fw (a) and (b), respectively, H<sub>2</sub>O/DC (c) and (d), TIP3P-PPPM (e) and (f), and TIP3P-ST (g) and (h). The different colours represent the different bulk ionic concentrations with the silica density appearing in black.



**Figure S3:** Ion density profiles obtained with SPC/Fw and TIP3P-PPPM at two salt concentrations. For comparison, analytical predictions from Gouy-Chapman model are given using the with experimentally measured dielectric constant for water  $\epsilon_{exp}$ . (a-d) and with the constant  $\epsilon_{MD}$  derived from MD simulations (a'-d'). The SPC/Fw water model tends to underestimates the counter-ion density near the silica surface at 0.84 M, and effect more marked when the MD calculated dielectric constant is used. However, the predictions made by TIP3P-PPPM water are in a good agreement with the analytical solution despite its poor ability to predict the dielectric constant accurately. The counter-ion and co-ion concentration profiles near the charged silica surface are given at 0.84 mM NaCl and 0.042 M NaCl for SPC/Fw with analytical predictions based on  $\epsilon_{exp}$ . (a, c) and  $\epsilon_{MD}$  (a', c') respectively. The counter-ion and co-ion concentration profiles at 0.84 M NaCl and 0.042 M NaCl for TIP3P-PPPM with analytical predictions based on  $\epsilon_{exp}$ . (b, d) and  $\epsilon_{MD}$  (b', d') respectively.



**Figure S4:** Comparison of the condensation parameter obtained at various salt concentrations for the selected water models. The results are compared with analytical prediction from the Gouy-Chapman model. Counter-ion (blue) and co-ion (red) condensation profiles are shown with a solid line for the MD simulations and a dashed line for the analytical predictions. The SPC/Fw-derived condensation parameter is given for 0.84 M NaCl (a), 0.42 M NaCl (e), 0.21 M NaCl (i), 0.1 M NaCl (m) and 0.042 M NaCl (q) respectively. The H2O/DC-derived condensation parameter is given for 0.84 M (b), 0.42 M (f), 0.21 M NaCl (j), 0.1 M (n) and 0.042 M (r) respectively. The TIP3P-PPPM-derived condensation parameter is given for 0.84 M (c), 0.42 M (g), 0.21 M NaCl (k), 0.1 M (o) and 0.042 M (s) respectively. The TIP3P-ST-derived condensation parameter is given for 0.84 M (d), 0.42 M (h), 0.21 M NaCl (l), 0.1 M (p) and 0.042 M (t) respectively.



**Figure S5:** Concentration of ion pairs as a function of  $z$  distances for different concentrations of NaCl in SPC/E water. The concentration of pairs increases non-linearly with the salt concentration. This supports the idea that effects due to ion pairs dominate at higher concentrations