

### Supporting Information

The Supporting Information is available free of charge.

**Table S1** – Urea parameters in the OPLS-DA and OPLS-DA/Urea force fields

**Table S2** – MD GAFF0.8-PPC diffusion and shear viscosity coefficients of ChCl:U:W(1:2:ζ)

**Figure S1** – Mean square displacement for the DES components

**Figure S2** – Normalized stress tensor time-correlation functions

**Figure S3** – Cumulative viscosity of GAFF0.8-PPC reline

**Figures S4-S13** – Radial distribution functions for the different components and water contents

**Figure S14** - Coordination number (Choline-urea) for the OPLS-DA and OPLS-DA/Urea force fields

**Figure S15** –Experimental and MD oxygen-oxygen radial distribution functions for pure water