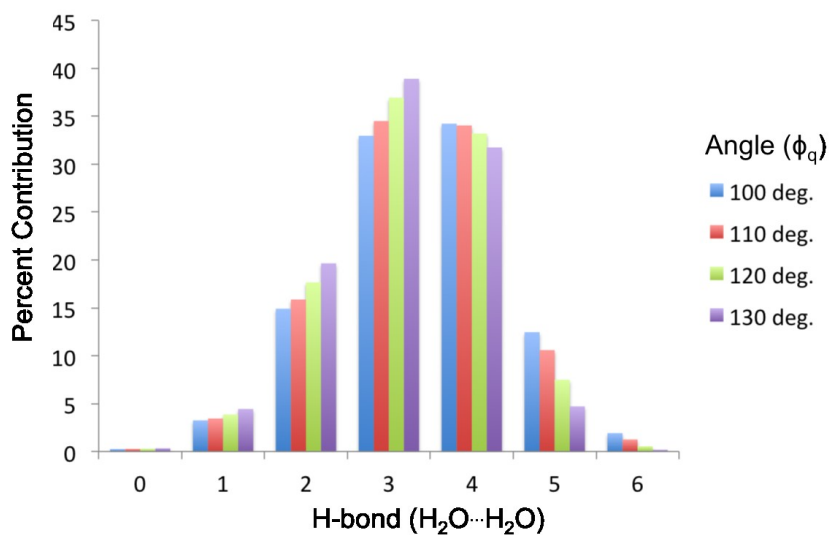# Supplemental Information

Figure S1. Distribution of hydrogen bonds, formed by waters with the quartz surface and other waters, as a function of angle criterion ϕ_q. Bulk waters within a perpendicular distance of 5 Å from the quartz surface are used to show the dependence of hydrogen bond distributions upon ϕ_q. H_2O-silanol distance is set to <3 Å.

Figure S2. Percent of water-water H-bonds as a function of durations over the layers.
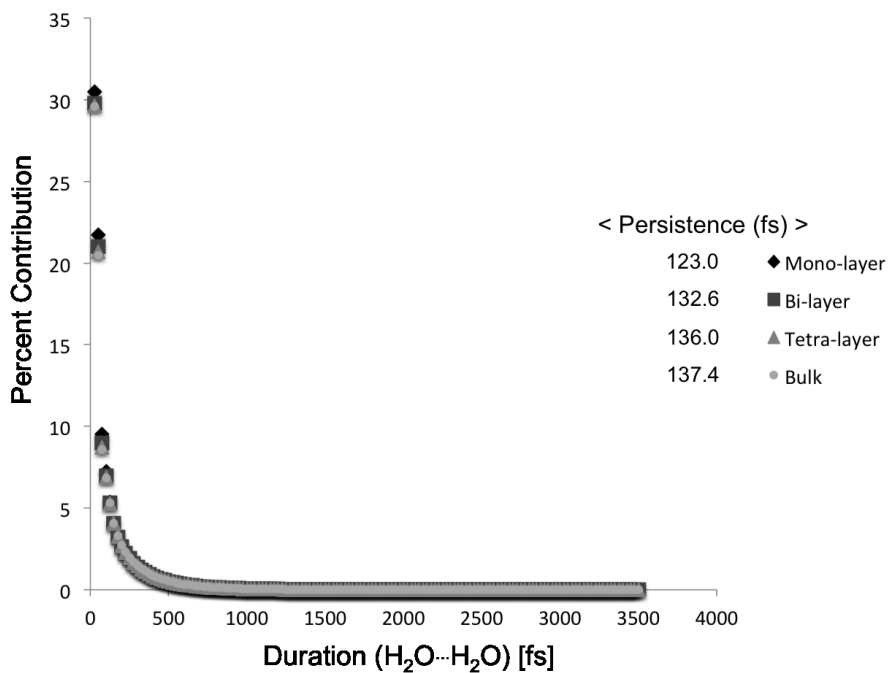
Figure S3. Percent of silanol-water hydrogen bonds as a function of durations over the hydration layers.
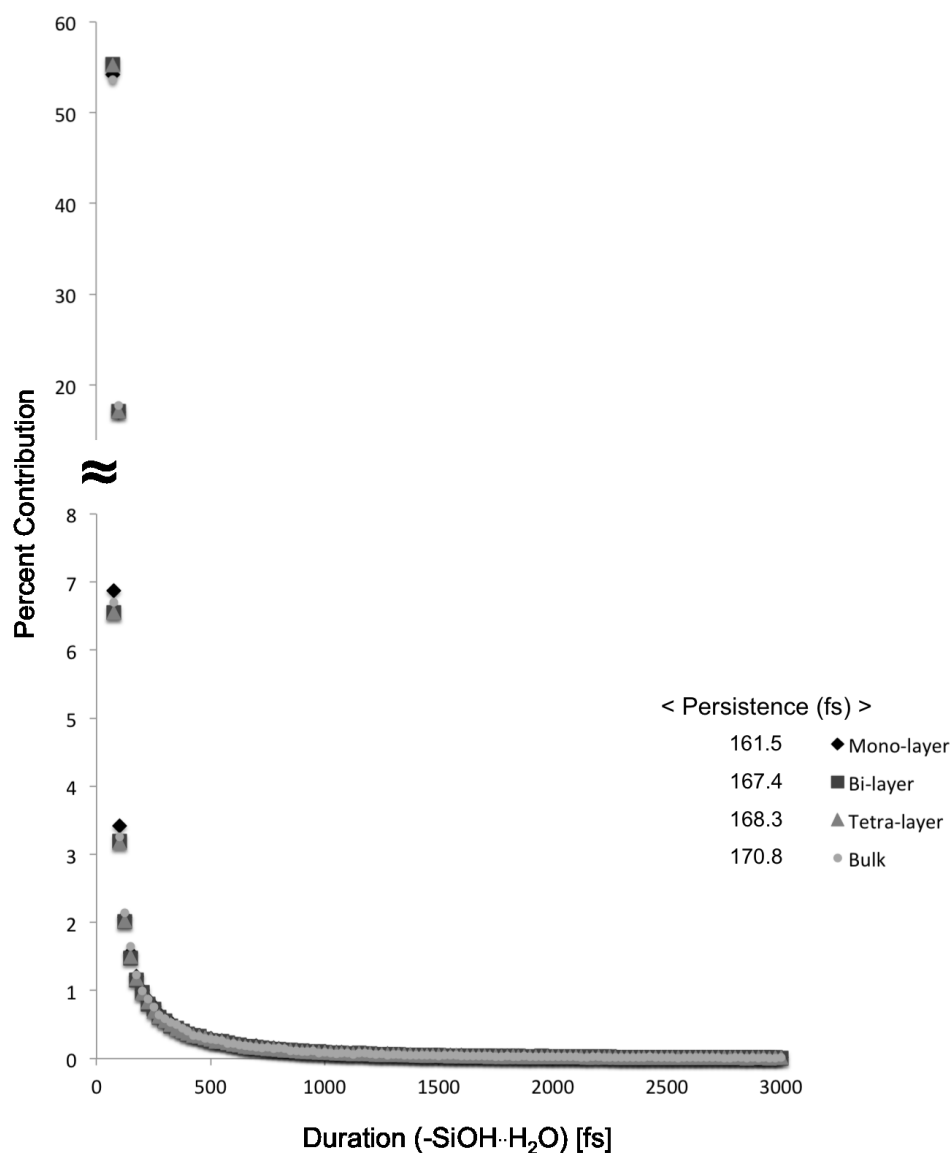
Table S1. Average water dipole angles (α and β) over the hydration levels.

Water/Quartz interface	<Dipole angle α> H-bond donor	<Dipole angle β> H-bond acceptor
Mono-layer	124.8	56.0
Bi-layer	125.0	50.9
Tetra-layer	125.0	50.5
Bulk	125.3	50.3