It is hard to imagine any surface on this planet more important to our survival than the surface of water. Our rivers, streams, oceans and clouds all interact with their surroundings through a molecularly thin “film” of uniquely bonded water molecules that we call the water surface. With recent technological advance in the scientific and computational tools that we now have available to probe and prod this surface, we are obtaining a deeper understanding of its molecular characteristics and what role it plays in many important chemical, biological and physical processes on this planet. In this presentation I will provide an overview of our most recent results in which we explore the structure, bonding and orientation of water, ions and adsorbed molecular species at the surface of aqueous solutions in contact with air, hydrophobic liquids and monolayer surfaces. Our studies involve a combination of nonlinear and linear spectroscopic techniques, thermodynamic measurements and molecular dynamics simulations.