Abstract: Organic carbon molecules in the atmosphere are emitted from anthropogenic and natural sources, and play a key role in atmospheric chemistry, air quality and climate. Recent studies have discovered organic molecular chromophores that absorb in the near UV (300-400 nm) and visible regions that are present in aerosols, termed “brown carbon” (BrC) aerosols. These light-absorbing BrC molecules possessing a diversity of functional groups determine the optical properties of aerosols, even at the nanomolar concentration. Following solar absorption, a multitude of photo-initiated processes takes place on multiple potential energy surfaces arising from the chromophore’s structure, chemical functionality, and the immediate solvent environment. Therefore, a synergy of spectroscopy and chemical dynamics methods is required to obtain a molecular-level view of such complex chemical systems. To address these opportunities, we will illustrate our efforts to investigate the photochemistry and photophysics of BrC chromophores and describe the impacts of solvent intermolecular interactions using single-conformation spectroscopy and dynamics techniques. Thus, the photophysical, photochemical and structural details of the target conformational isomers and molecular complexes enable multifaceted comparisons to theoretical predictions.