Moving toward a circular carbon economy requires enabling reuse of carbon-based macromolecules like those in biomass and polymers. Meeting this goal requires increased upcycling of waste products into higher value products. For biomass waste such as corn stover, pine needles, or bark, one approach is using pyrolysis to convert these feedstocks into bio-oil that can be used for liquid fuel or transformed into other hydrocarbon based products, like bio-polymers. Waste plastics can be upcycled or recycled into new products, or alternatively converted into bio-oil by pyrolysis. Macromolecules from biomass and polymers are challenging to convert into liquid fuel or chemical feedstocks, due to large and sometimes heterogeneous networks of polymeric bonds. We work with the Idaho National Laboratory’s Biomass Feedstock National User Facility and Department of Energy’s Feedstock Conversion Interface Consortium to develop a molecular understanding of biomass variability that occurs naturally and as a result of storage or preprocessing techniques applied to feedstocks, to understand chemical reactions occurring during pyrolysis and other biomass or polymer conversion techniques. Using two dimensional gas chromatography mass spectrometry coupled to analytical pyrolysis provides insight into the molecular changes that occur during the pyrolysis event, and these insights can extend to bench or pilot scale pyrolysis conversions. Understanding pyrolysis of biomass and polymers at a molecular level contributes to development of new tools to predict and manipulate the optimum conditions and identify the best storage, preprocessing, and conversion techniques to maximize output of specific high-value chemical species in the conversion to fuels, or to characterize feedstocks for blending before pyrolysis. Identification and understanding of the molecular level changes resulting from novel preprocessing techniques that can be used to control and manipulate the chemical output of bench or pilot scale pyrolysis is another focus of investigation, including the use of gamma irradiation, acid, or enzyme pre-treatments. We also focus on the impacts of storage conditions and techniques, which can affect moisture levels, degradation, and impacts from naturally occurring microbes in the environment.