While single-site catalysis dominates research in organometallic chemistry and homogeneous catalysis, bimetallic and multimetallic cooperation is prevalent in many biological and heterogeneous catalytic systems. Our group seeks to understand and exploit bimetallic cooperation in homogeneous catalysis using a two-pronged approach. First, we conduct synthetic model studies that aid in understanding the role of multimetallic catalysis in biological systems. In this presentation, synthetic efforts towards modeling the [Cu4S] catalytic site of nitrous oxide reductase and the [CuSMo] catalytic site of aerobic carbon monoxide dehydrogenase will be discussed. Second, we leverage our understanding of multimetallic catalysts in nature to design bimetallic systems that catalyze reactions of importance to chemical synthesis. In this presentation, the development of catalytic transformations involving binuclear bond activation and formation steps, as well as orthogonal mononuclear catalysis in tandem and mutually dependent scenarios, from a family of (NHC)Cu-[MCO] complexes will be discussed (NHC = N-heterocyclic carbene, MCO = metal carbonylate).