A great challenge in material science is to tune the properties of organic crystals using the strategy of substitutional solid-solutions (SSS), a method commonly used in inorganic chemistry. The main reasons for this challenge are due to 1) the inability to predict the crystal packing of low-symmetry organic molecules, 2) their trend to phase-separate upon crystallization, 3) and their loss of expected properties due to aggregation. In this seminar we explore the use of multivariate MOFs—those that are made from mixtures of isostructural organic links—as platforms for making SSS that behave like liquid solutions. To this end, we developed a series of molecular and materials design rules that enabled us to prepare a variety of MTV MOFs that feature solution properties of organic molecules with an unprecedented level of synthetic control: we can predict their high-symmetry crystal structure, avoid phase separation, and retain the molecular properties of the multivariate links. We demonstrate the generality of our approach by preparing water and acid stable MOFs that exhibit solution-type properties rarely observed in crystalline solids, like dilute fluorescence, molecular aggregation and diffusion-limited redox electron transfer. Furthermore, the properties observed enabled us to prepare metal-organic materials that exhibit multicolor and white light emission, J-dimer emission and redox conductivity with high precision for applications in lighting, displays, batteries, and photocatalysis.