The emergence of electrically conductive metal-organic frameworks (MOFs) has been one of the most paradoxical developments in the field in the last few years. Indeed, how can one transport charges through a material that is mostly “empty” space? In this sense, MOFs made from layers of organic ligands connected by (typically) square-planar metal ions have shown particularly good electrical conductivity. However, a precise mechanism for charge transport is still the subject of debate, with various experimental and computational reports describing these materials as metals, semiconductors, semimetals, or even borderline insulators. Most of the discussion on this point has been focused on the effects of in-plane metal-ligand conjugation and the efficiency of in-plane transport. This lecture will describe the latest efforts from our group to understand the intrinsic properties of these materials, especially as related to single-crystal electrical measurement studies, and will discuss in particular the unexpectedly large influence of out-of-plane transport. Time allowing, I will discuss unexpected results stemming from the behavior of these materials as 1D metals, and applications in energy storage and conversion.