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Jarosite, a mineral with a kagomé lattice, displays magnetic frustration yet orders magnetically below 65 K. As magnetic frustration can engender exotic physical properties, understanding the complex magnetism of jarosite comprises a multi-decade interdisciplinary challenge. Unraveling the nature of the disparate magnetic coupling interactions that engender magnetic order in jarosite remains as an open question. Specifically, there is no observed trend in the interlayer spacing with magnetic order. Similarly, the relationship between metal–ligand bond distance and magnetic order remains uninvestigated. Here, we use applied pressure as a vector to smoothly vary jarosite's structure without manipulating the chemical composition, enabling a chemically invariant structure–function study. Using single-crystal and powder X-ray diffraction, we show that high applied pressures alter both the interlayer spacing and the metal–ligand bond distances. By harnessing a suite of magnetic techniques under pressure, including SQUID-based magnetometry, time-resolved synchrotron Mössbauer spectroscopy, and X-ray magnetic circular dichroism, we construct the magnetic phase diagram for jarosite up to 40 GPa. Notably, we demonstrate that the magnetic ordering temperature increases dramatically to 240 K at the highest pressures. Additionally, we conduct X-ray emission spectroscopy, Mössbauer spectroscopy, and UV-visible absorption spectroscopy experiments to comprehensively map the magnetic and electronic structures of jarosite at high pressure. We use these maps to construct chemically-pure magnetostructural correlations which fully explain the nature and role of the disparate magnetic coupling interactions in jarosite.