A methane nano-trap based on an alkyl metal–organic framework (MOF) and featuring oppositely adjacent open metal sites and dense alkyl groups is reported by S. Ma, Z. Niu, and co-workers in their Communication (DOI: 10.1002/anie.201904507). The methane nano-trap endows MOF with the record-high methane uptake and excellent CH4/N2 separation performance, thereby providing a new perspective for capturing methane from coal-mine methane to recover fuel and reduce greenhouse gas emissions.