A better understanding of carbon dioxide dynamics in metal–organic frameworks (MOFs) can lead to the optimal design of materials for carbon-capture applications. In their Communication on page 4410 ff., L.-C. Lin et al. utilized molecular simulation techniques to analyze CO₂ dynamics inside MOFs with open metal sites. They identified the hopping motion of CO₂ between different metal sites as the signature of the experimentally measured patterns in the ^{13}C NMR chemical shift anisotropy.