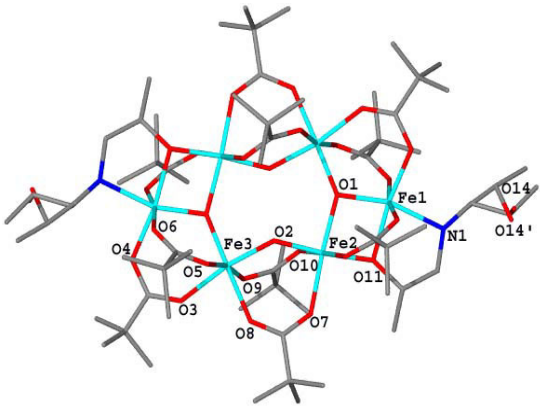


Fig. 3 Structure of **2**. Selected inter-atomic bond lengths in **1** (Å): $Fe^{(III)}$-O; 1.8745(17) – 2.0846(19), $Fe^{(III)}$-N1 = 2.174(2), (O2(H2A)···O10′ = 2.795, in **2**: $Fe^{(III)}$-O; 1.875(2)-2.056(2), $Fe^{(III)}$-N1 = 2.162(3), O2(H2A)···O10′ = 2.949. (Fe′ is related to Fe by the 1-x,1-y,1-z symmetry operator).

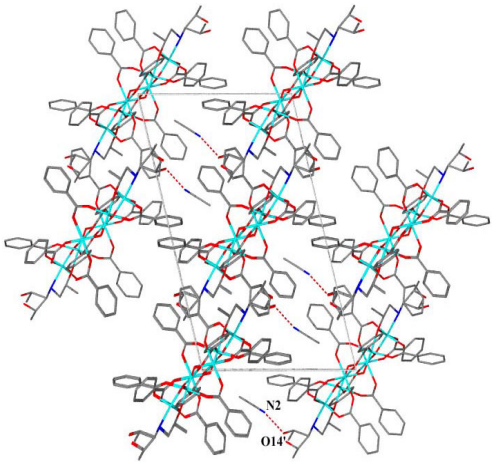


Fig. 4 Crystal packing diagram of **1** viewed along the *b* axis. Hydrogen atoms have been omitted for clarity. Red dashed line represents H-bonding between MeCN and **1** (N2···O14/O14′).

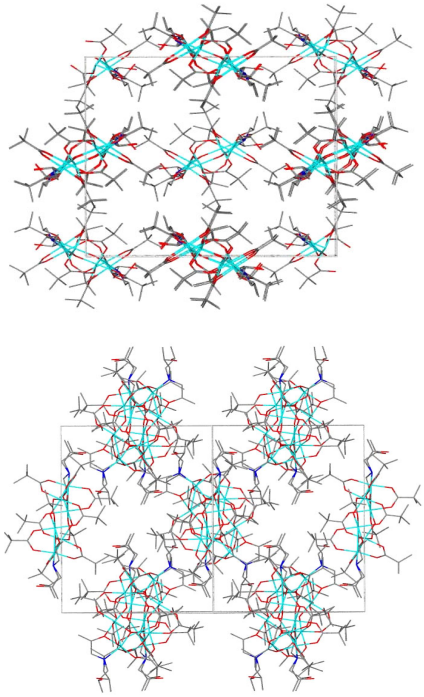


Fig. 5 Crystal packing in **2** observed along the *c* axis (top) and along the 1,0,-1 direction (below).

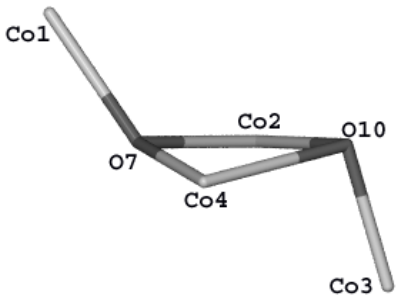


Fig. 9 Chair conformation of the common $\{M_4O_2\}$ 'butterfly' motif observed in the core of the Co_4 clusters in **3**. For selected interatomic bond lengths (Å) and angles (°) see Table 3.

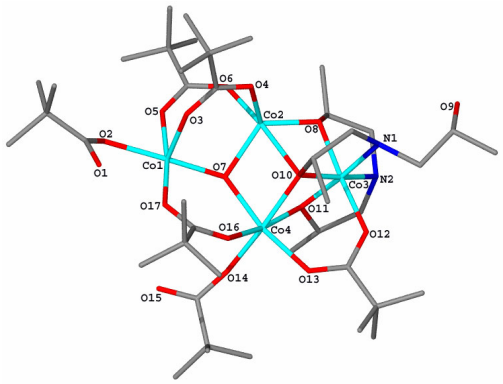


Fig. 10 The asymmetric unit of the [Co_4] clusters in **3**. Hydrogen atoms have been omitted for clarity. The sixth bonding site at Co1 has been omitted as it bridges to the next Co_4 cluster. Selected inter-atomic distances (Å): $Co^{(II)}$-O; 2.033(3) – 2.152(3), $Co^{(III)}$-O; 1.874(3) – 2.315 (3), $Co3^{(III)}$-N1 = 1.971(4), $Co3^{(III)}$-N2 = 1.961(4).

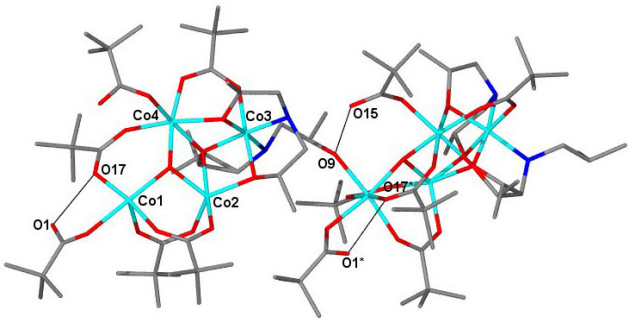
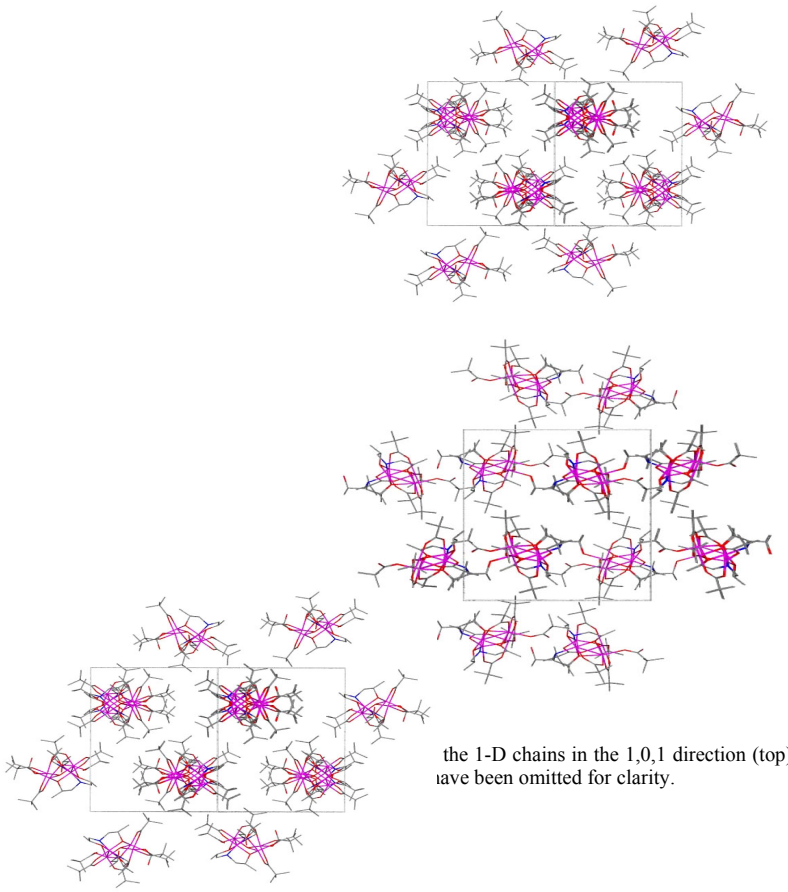


Fig. 11 Core of two [Co_4] clusters in **3**, illustrating the H-bonding interactions within and in-between the [Co_4] moieties. H-bonding distances (Å): O1(H1)···O17 = 2.585, O15(H15)···O9 = 2.592.

Electronic Supplementary Information for Dalton Transactions



the 1-D chains in the 1,0,1 direction (top). The linear rows of the [Co_4] tetramers have been omitted for clarity.