**Supplementary information**

The Supplementary Information contains the structures and summaries of the X-ray structures (see attached cif), the Slant Fourier map of the iron-aquo environment indicating the electron density attributed to the protons of the aquo ligand and a histogram of the Fe-OR\textsubscript{2} (ether) distances from the CCDC.

**Figure S11** Slant fourier map of the iron-aquo environment indicating the electron density attributable to the protons on the aquo ligand of complex \{[Fe(II)(H\textsubscript{2}O)L\textsuperscript{1b}\textsubscript{2}][Fe(III)Cl\textsubscript{4}]\}_2 \textsuperscript{16}.

![Slant Fourier map](image)

**Figure S12** A histogram of the Fe-OR\textsubscript{2} (metal-ether) distances as found in the Cambridge Crystallographic Data Centre.

![Histogram](image)

Below are Ortep depictions of the single crystal X-ray diffraction data were obtained for compounds HL\textsuperscript{1b}, HL\textsuperscript{4b}, 12-22, 27 and 32 (Fig. SI3-SI18).
**Figure SI3.** X-ray crystal structures of ligand **HL**\textsuperscript{1b}. Thermal ellipsoids drawn at 50% probability and hydrogen atoms removed for clarity.

**Figure SI4.** X-ray crystal structure of ligand **HL**\textsuperscript{1c}. Thermal ellipsoids drawn at 50% probability and hydrogen atoms removed for clarity.
**Figure SI5.** X-ray crystal structure of bis(3,5-bis(2-methoxyphenyl)-1H-pyrazol-1-yl) 4b. Thermal ellipsoids drawn at 50% probability and hydrogen atoms removed for clarity.

**Figure SI6.** X-ray crystal structure of complex [Mn(L1a)2] 12. Thermal ellipsoids drawn at 50% probability and hydrogen atoms, solvent and disorder removed for clarity.
**Figure S17.** X-ray crystal structure of complex [{MnL₁bCl₂}] 13. Thermal ellipsoids drawn at 40% probability and hydrogen atoms and solvent removed for clarity.

**Figure S18.** X-ray crystal structure of complex [Mn(L₁c)₂] 14. Thermal ellipsoids drawn at 50% probability and hydrogen atoms, solvent and disorder removed for clarity.
**Figure S19.** X-ray crystal structure of complex [Fe(L^{1a})_{2}] 15. Thermal ellipsoids drawn at 50% probability with the minor component of disorder in a phenyl ring and the hydrogen atoms removed for clarity.

**Figure S110.** X-ray crystal structure of complex [{Fe(H\textsubscript{2}O)L^{1b}}_{2}][FeCl\textsubscript{4}] 16. Thermal ellipsoids drawn at 50% probability and hydrogen atoms, solvent and counterions removed for clarity.
**Figure S11.** X-ray crystal structure of complex [Fe(L^{1b})_2] 17. Thermal ellipsoids drawn at 50% probability and hydrogen atoms removed for clarity.

**Figure S12.** X-ray crystal structure of complex [Fe(L^{1c})_2] 18. Thermal ellipsoids drawn at 50% probability and hydrogen atoms removed for clarity.
Figure S113. X-ray crystal structure of complex \([\text{Fe(III)}(L^{1b})_2][\text{Fe(III)Cl}_4]\) 19. Thermal ellipsoids drawn at 30% probability and the counterions, solvent and hydrogen atoms are omitted for clarity.

Figure S114. X-ray crystal structure of complex \([\text{Ni(L^{1a})}_2]\) 20. Thermal ellipsoids drawn at 50% probability with the minor component of disorder in a phenyl ring and the hydrogen atoms removed for clarity.
**Figure S15.** X-ray crystal structure of complex [Ni(L^1b)_2] 21. Thermal ellipsoids drawn at 30% probability with the hydrogen atoms removed for clarity.

**Figure S16.** X-ray crystal structure of complex [Ni(L^1c)_2] 22. Thermal ellipsoids drawn at 30% probability and the molecule of THF and the hydrogen atoms removed for clarity.
Figure S17. X-ray crystal structure of compound 27. Thermal ellipsoids at 50% probability and hydrogen atoms and solvent molecules have been removed for clarity.

Figure S18. X-ray crystal structure of complex 32. Thermal ellipsoids at 30% probability and the hydrogen atoms, solvent molecules and the minor component of disorder have been removed for clarity.