Electronic Supplementary Material (ESI) for CrystEngComm

Supplementary Materials

Solid State Coordination Chemistry of Oxomolybdenum-Organodiphosphonate Materials: Consequences of Introducing Xylyldiphosphonate Components.

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Figures S1-S14. ORTEP views of the building units of the compounds of this study, showing the partial atom-labeling schemes and 50% thermal ellipsoids.

Figures S15-S28. Packing views of the compounds of this study.

Figures S29-S42. Infrared spectra for the compounds of this study.

Figures S43-S56. TGA plots for the compounds of this study.

Figures S57-60. Temperature dependent magnetic susceptibility plots for compounds **5**, **14**, **10** and **12**, respectively.

Tables S1-S109. Tables of crystal data, atomic positional parameters and isotropic temperature factors, bond lengths and angles, anisotropic temperature factors, and hydrogen atom positions for the compounds of this study.

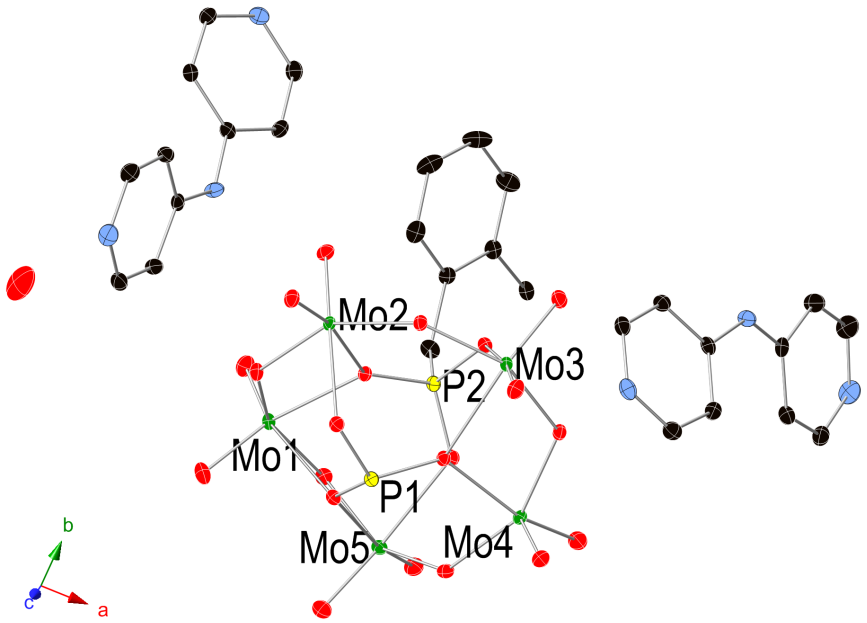


Figure S1. ORTEP view of compound **1**·H_2O, showing the atom-labeling scheme and 50% thermal ellipsoids.

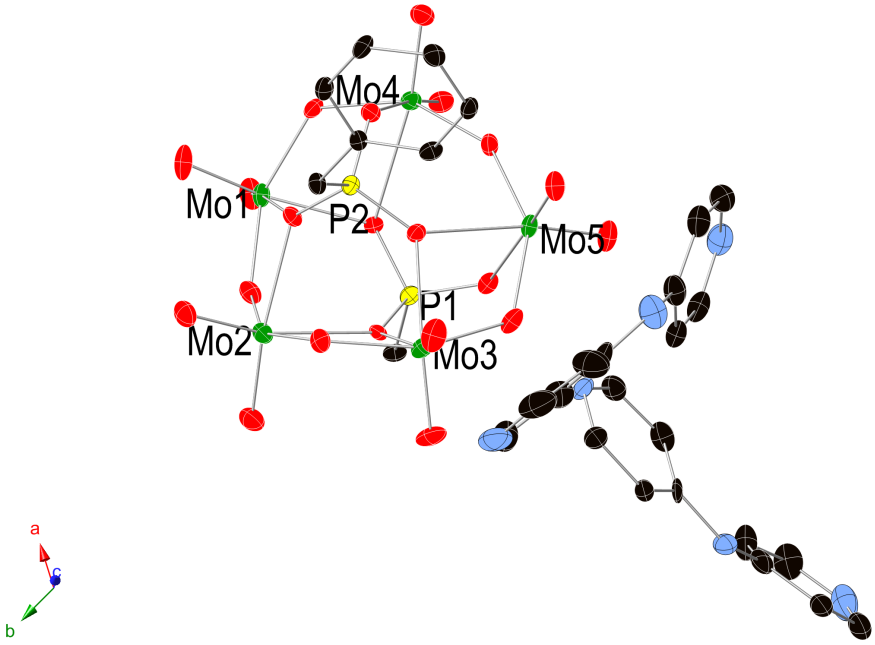


Figure S2. ORTEP view of compound **2**, showing the atom-labeling scheme and 50% thermal ellipsoids.

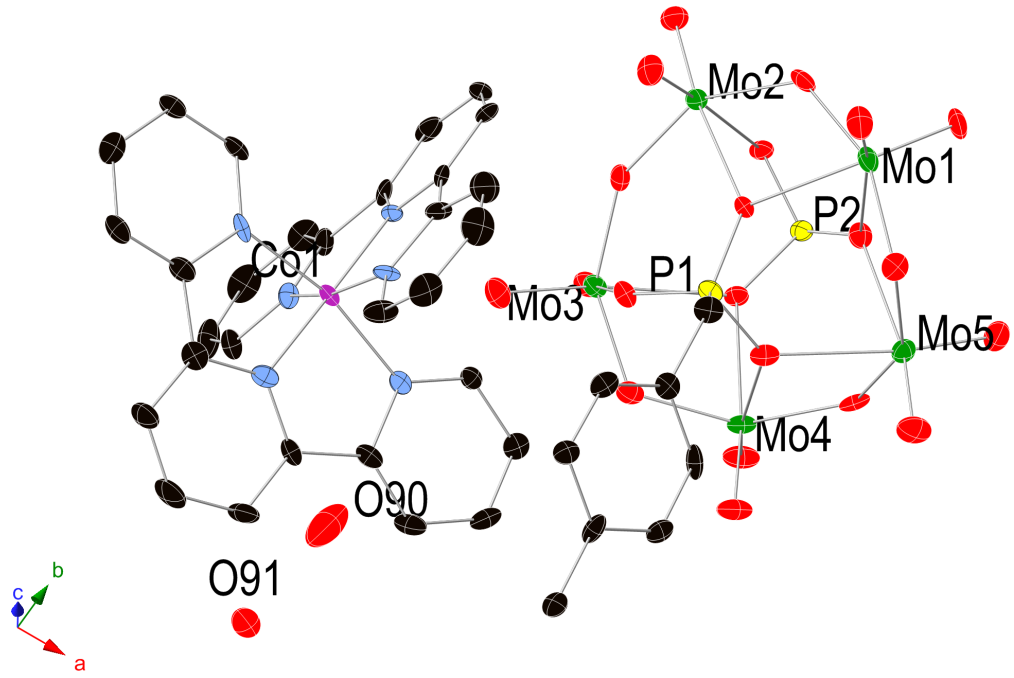


Figure S3. ORTEP view of compound **3**·H_2O, showing the atom-labeling scheme and 50% thermal ellipsoids.

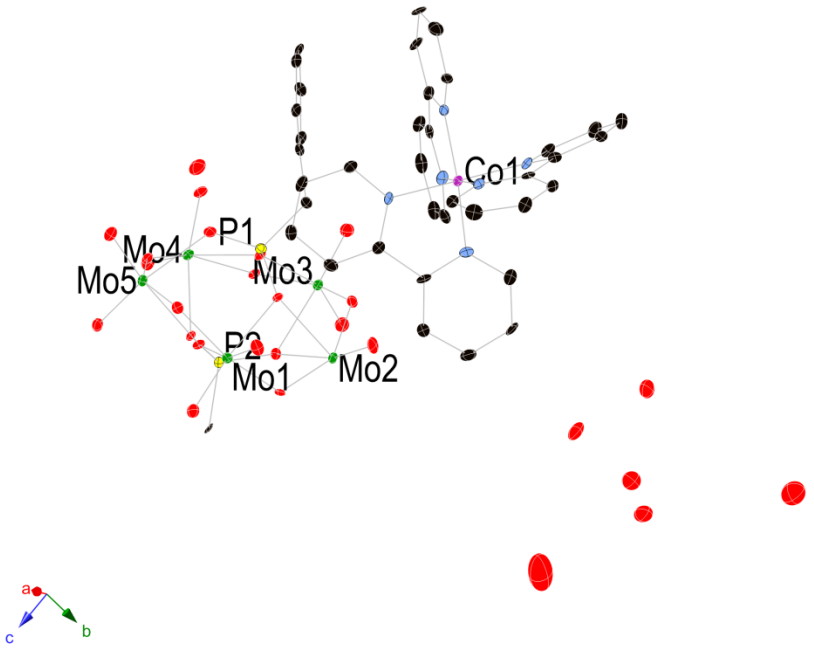


Figure S4. ORTEP view of compound **4**·$6H_2O$, showing the atom-labeling scheme and 50% thermal ellipsoids.

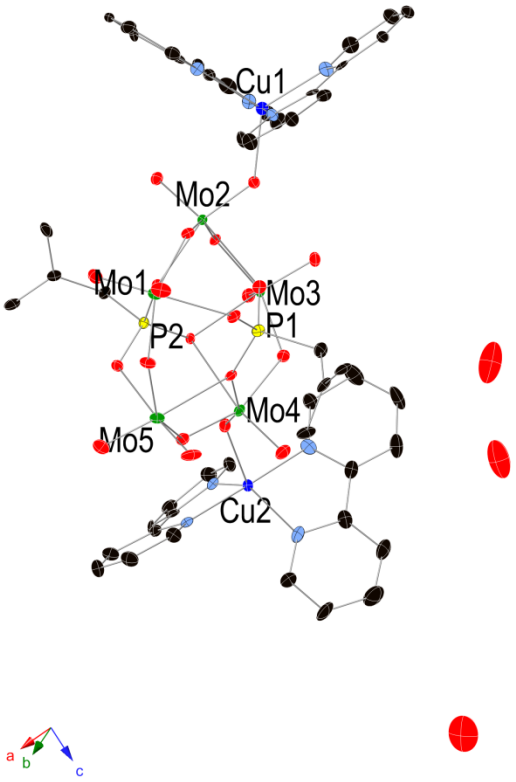


Figure S5. ORTEP view of compound **5**·6H_2O, showing the atom-labeling scheme and 50% thermal ellipsoids.

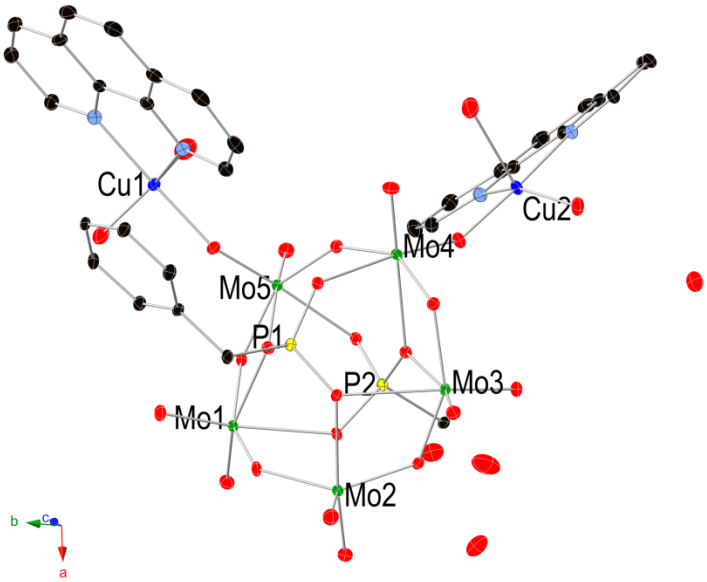


Figure S6. ORTEP view of compound **6**·4H_2O, showing the atom-labeling scheme and 50% thermal ellipsoids.

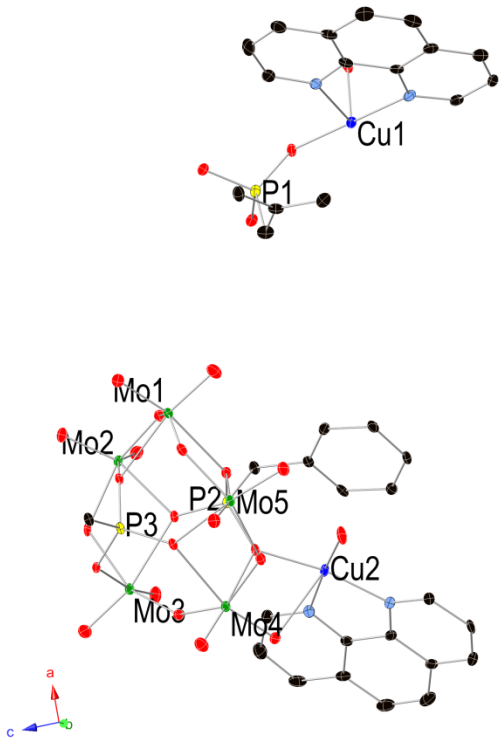


Figure S7. ORTEP view of compound **7**, showing the atom-labeling scheme and 50% thermal ellipsoids.

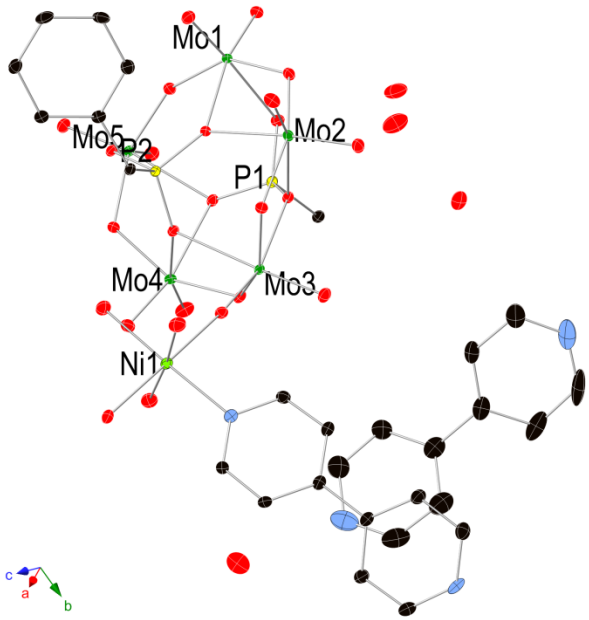


Figure S8. ORTEP view of compound **8**·4H_2O, showing the atom-labeling scheme and 50% thermal ellipsoids.

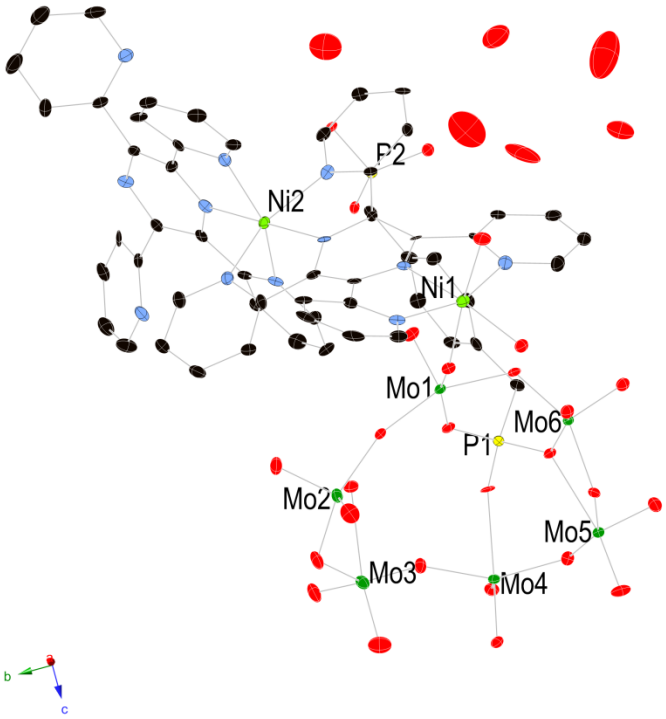


Figure S9. ORTEP view of compound **9**·6H_2O, showing the atom-labeling scheme and 50% thermal ellipsoids.

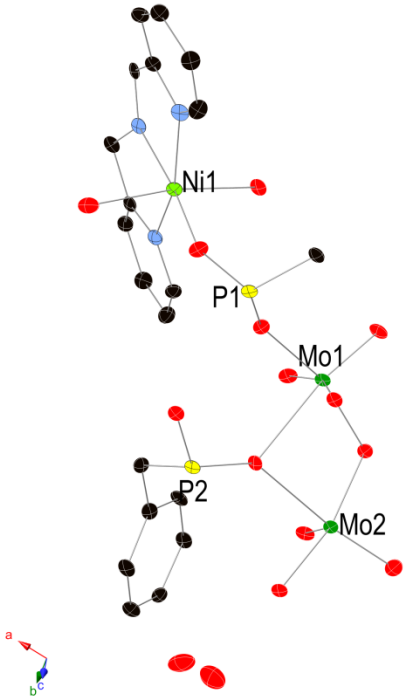


Figure S10. ORTEP view of compound **10**·4H_2O, showing the atom-labeling scheme and 50% thermal ellipsoids.

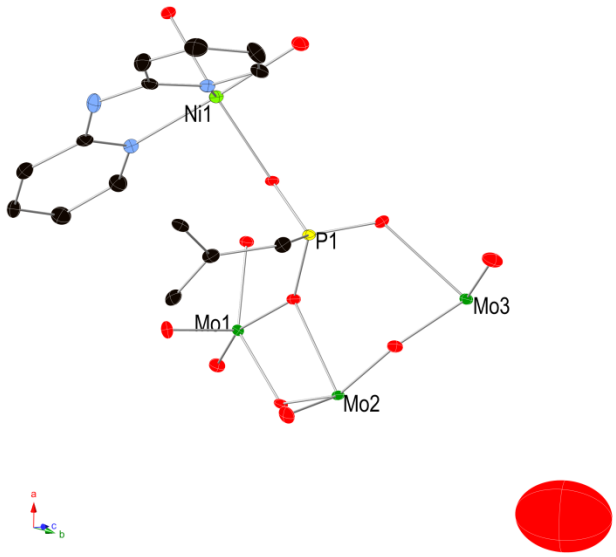


Figure S11. ORTEP view of compound **11**·2H_2O, showing the atom-labeling scheme and 50% thermal ellipsoids.

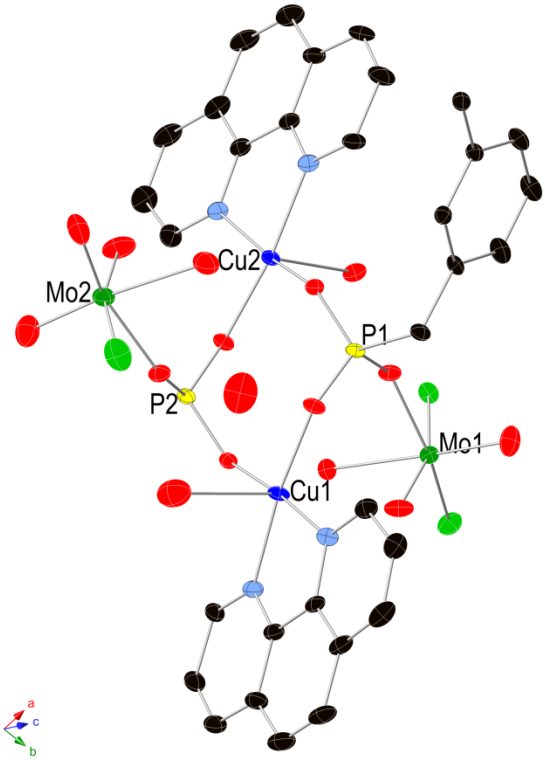


Figure S12. ORTEP view of compound **12**·H_2O, showing the atom-labeling scheme and 50% thermal ellipsoids.

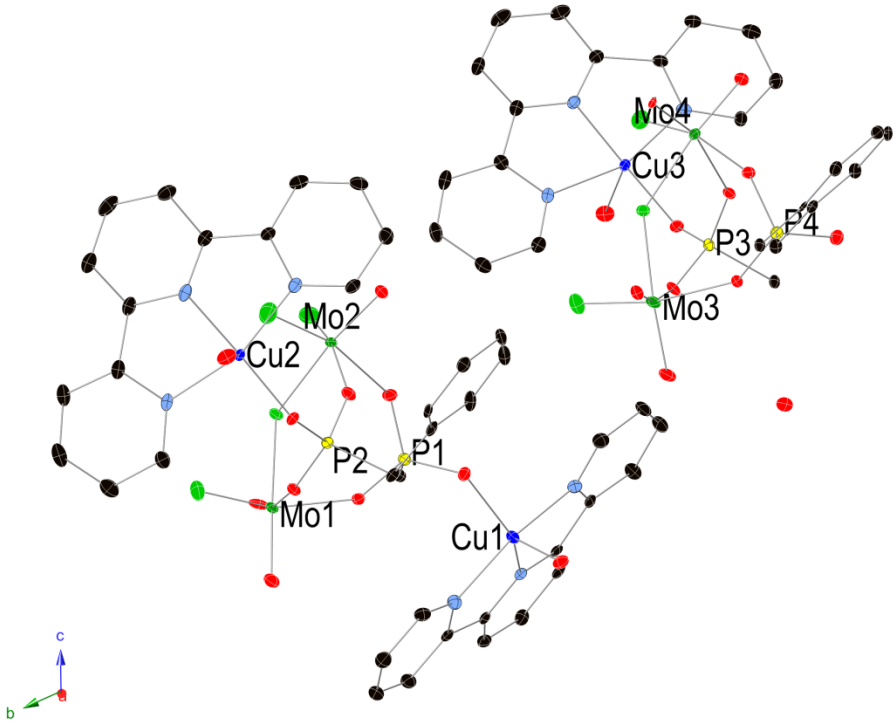


Figure S13. ORTEP view of compound **13**·H_2O, showing the atom-labeling scheme and 50% thermal ellipsoids.

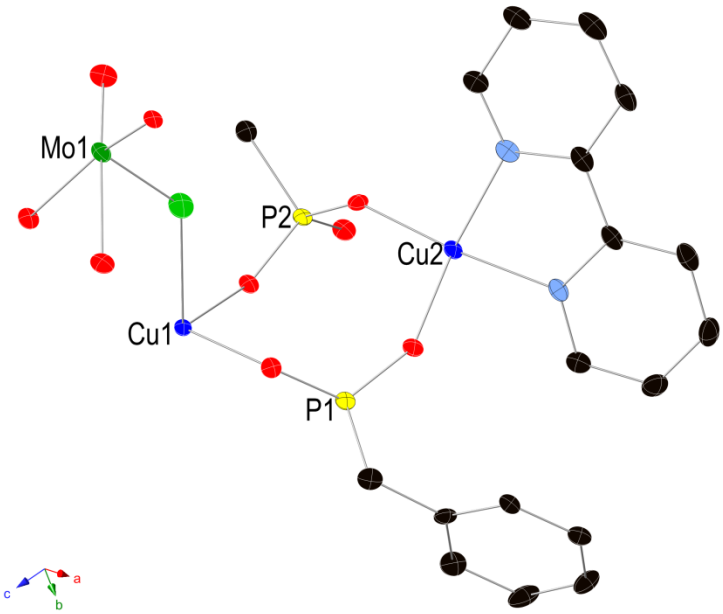
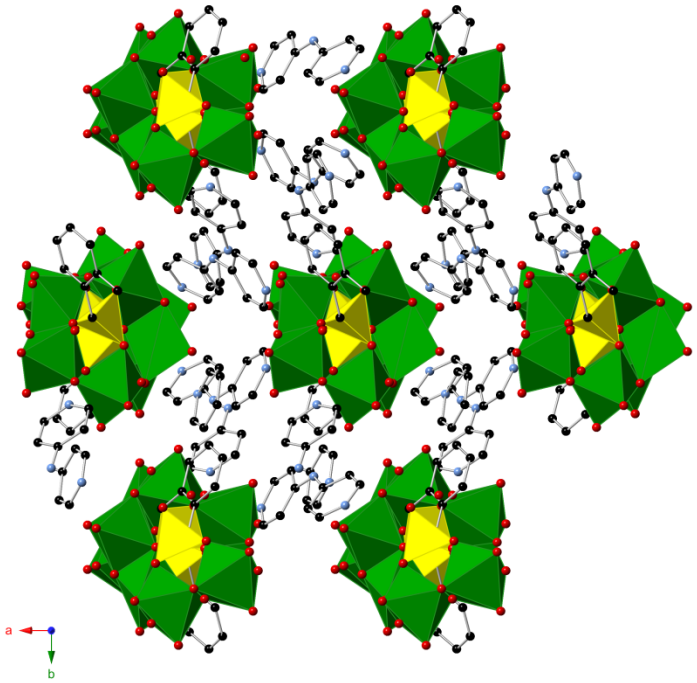
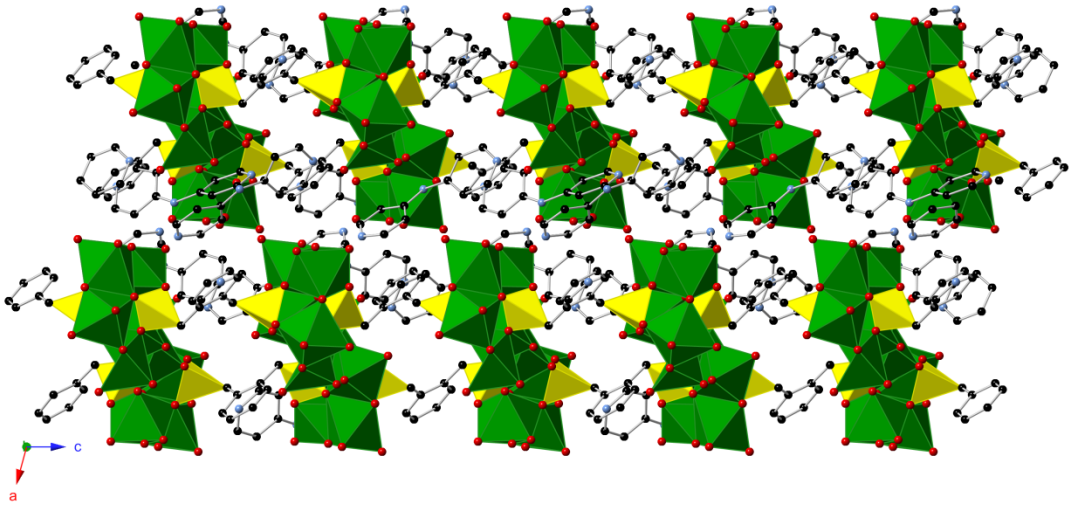


Figure S14. ORTEP view of compound **14**, showing the atom-labeling scheme and 50% thermal ellipsoids.



(**a**)



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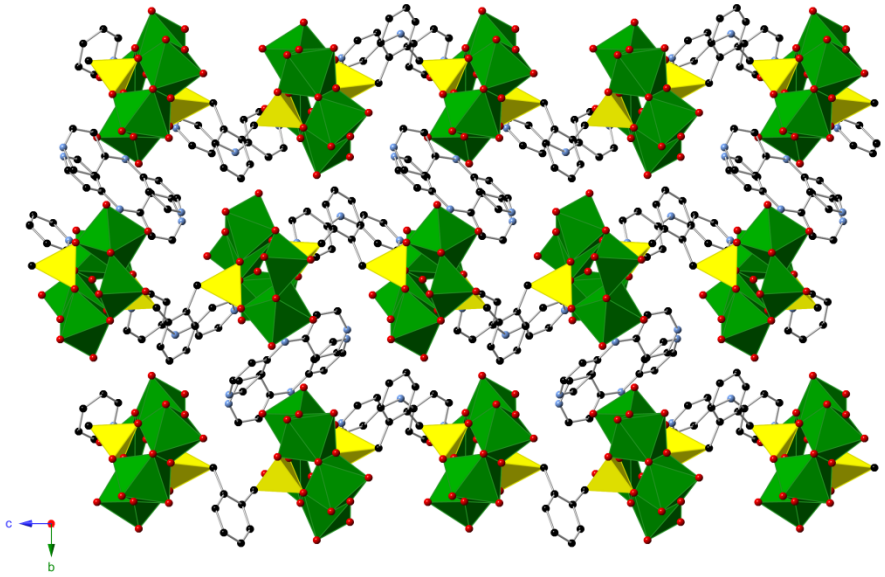
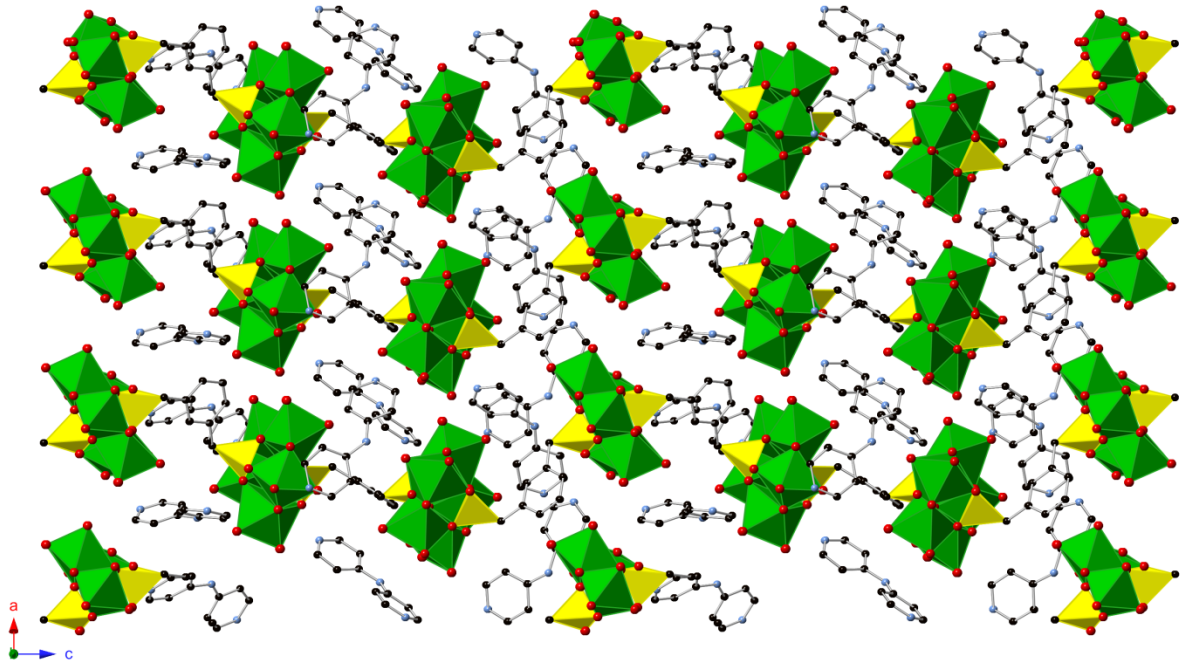
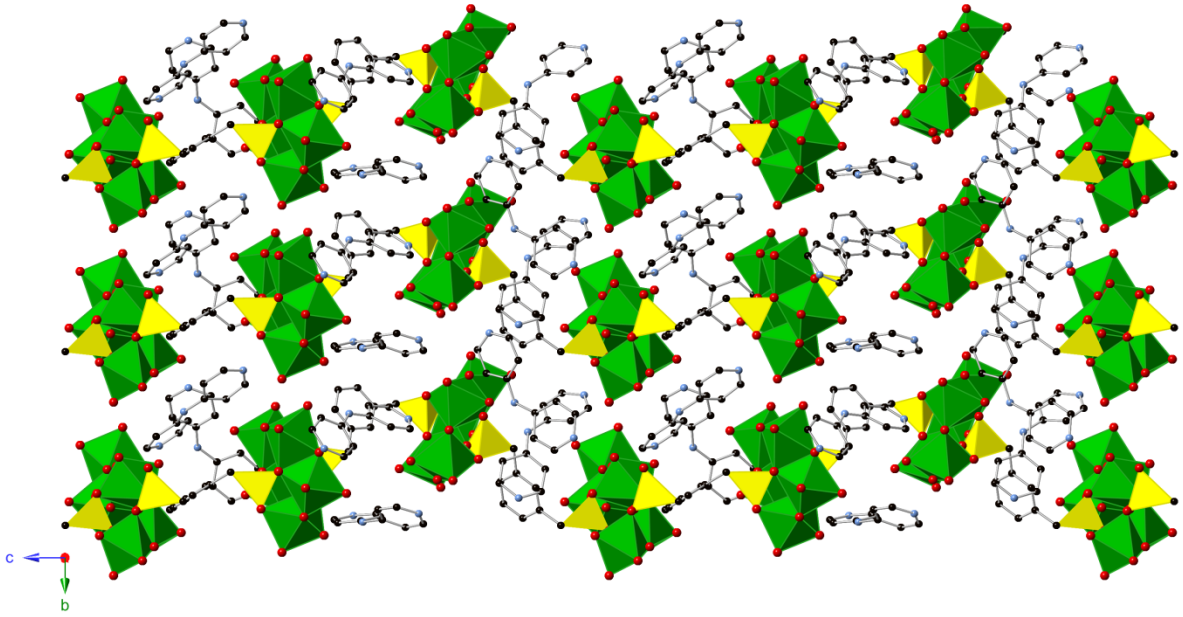


(**c**)

Figure S15. Packing view of **1**·H_2O in the (**a**) *ab* plane (**b**) *ac* plane and (**c**) *bc* plane. Water molecules of crystallization removed for clarity.

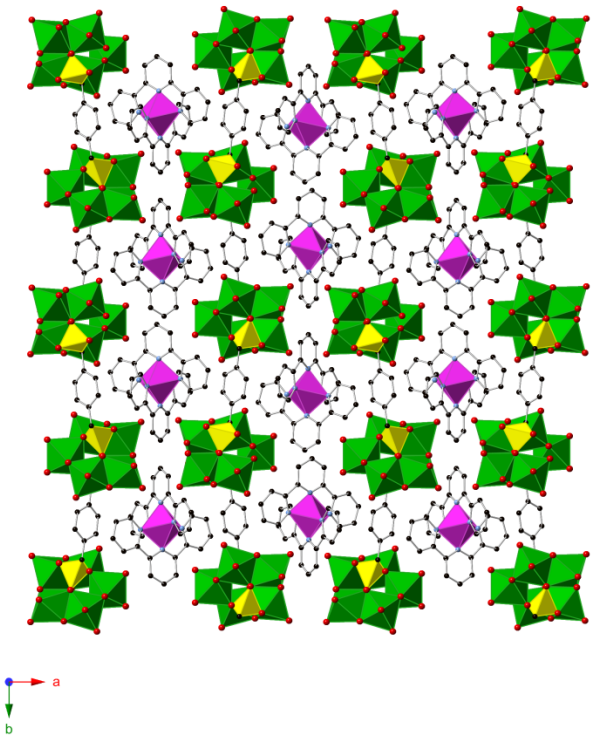


(**a**)

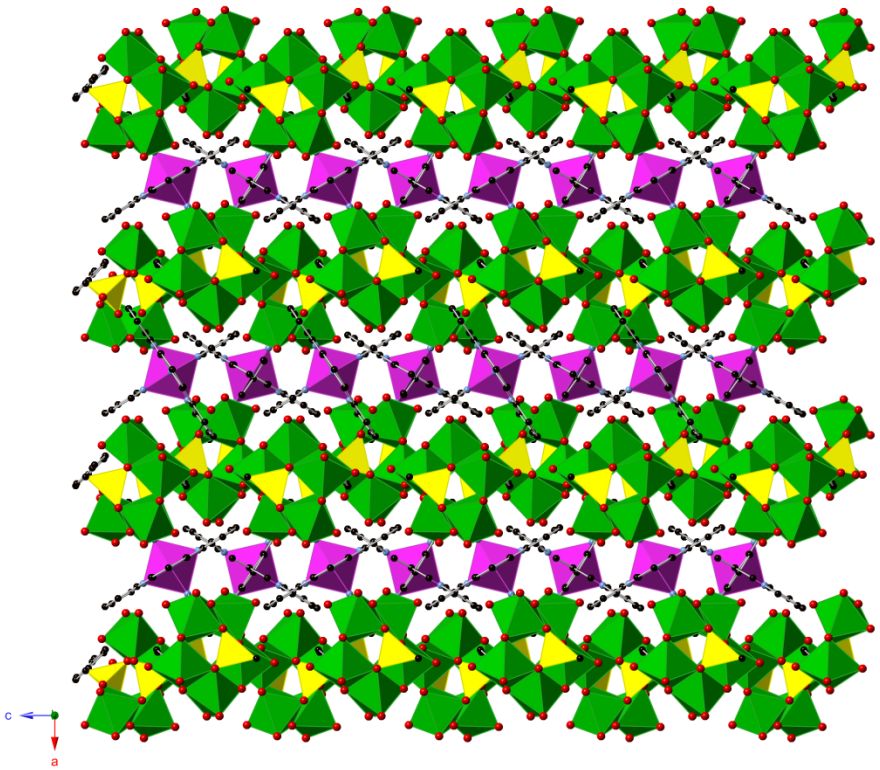


(**b**)

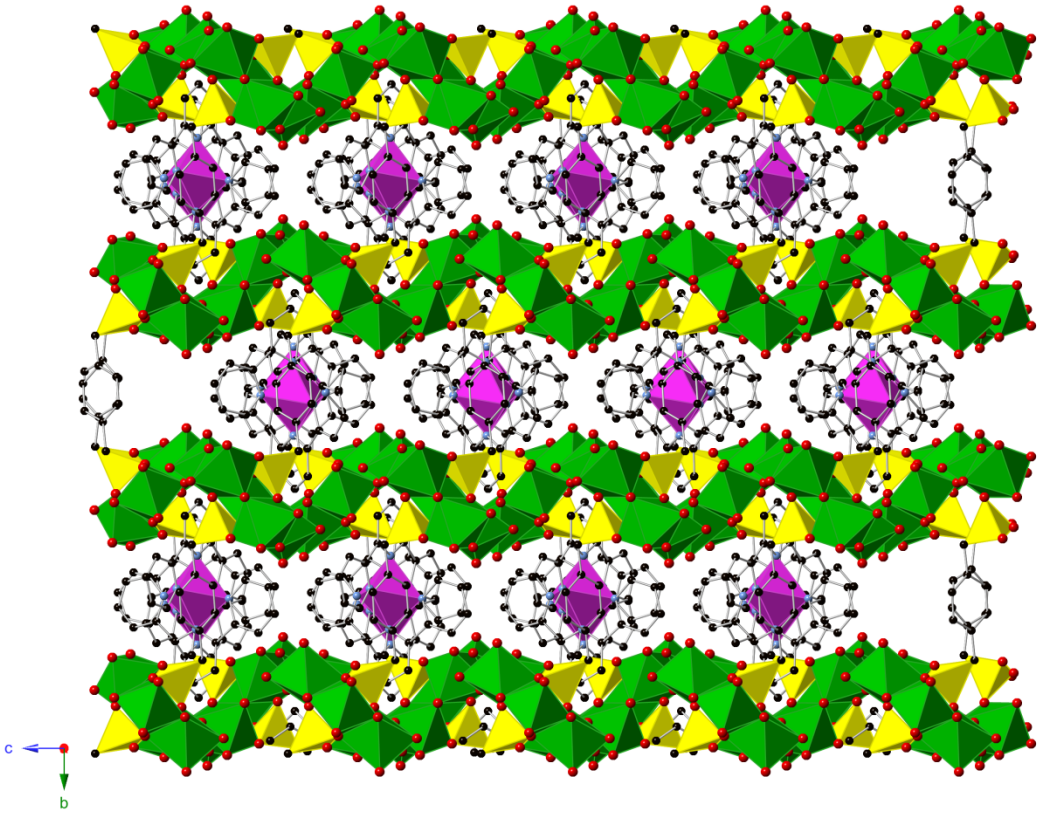
Figure S16. Packing view of **2** in the (**a**) *ac* plane and (**b**) *bc* plane.



(**a**)

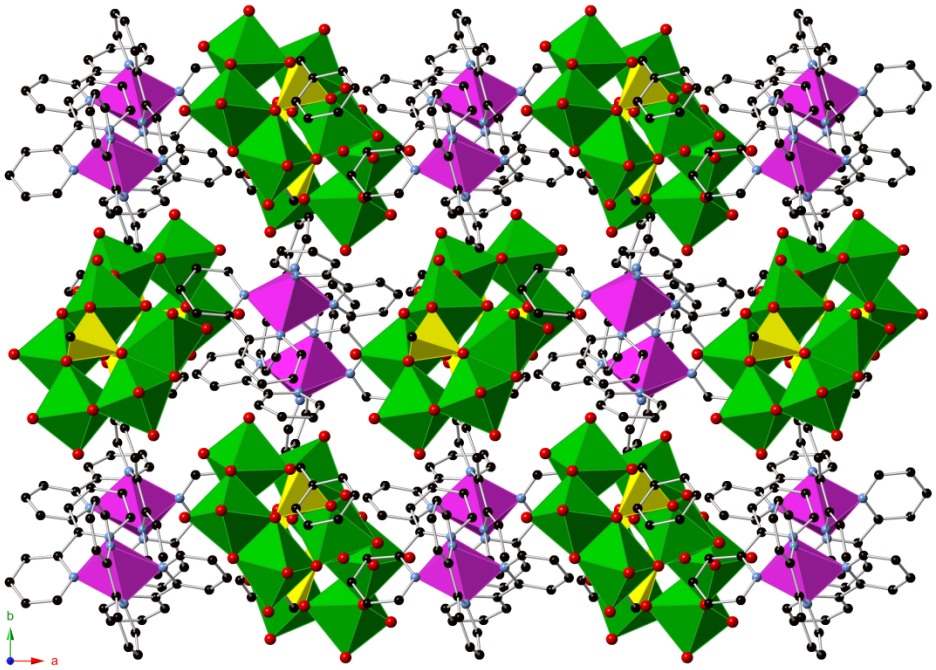


(**b**)



(**c**)

Figure S17. Packing view of **3**·H_2O in the (**a**) *ab* plane (**b**) *ac* plane and (**c**) *bc* plane. Water molecules of crystallization removed for clarity.



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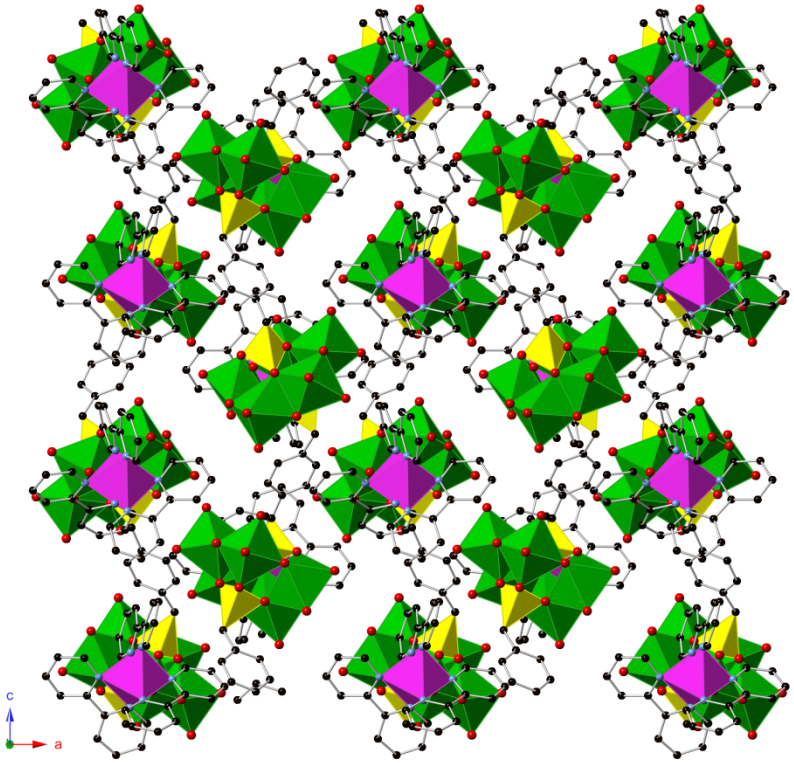
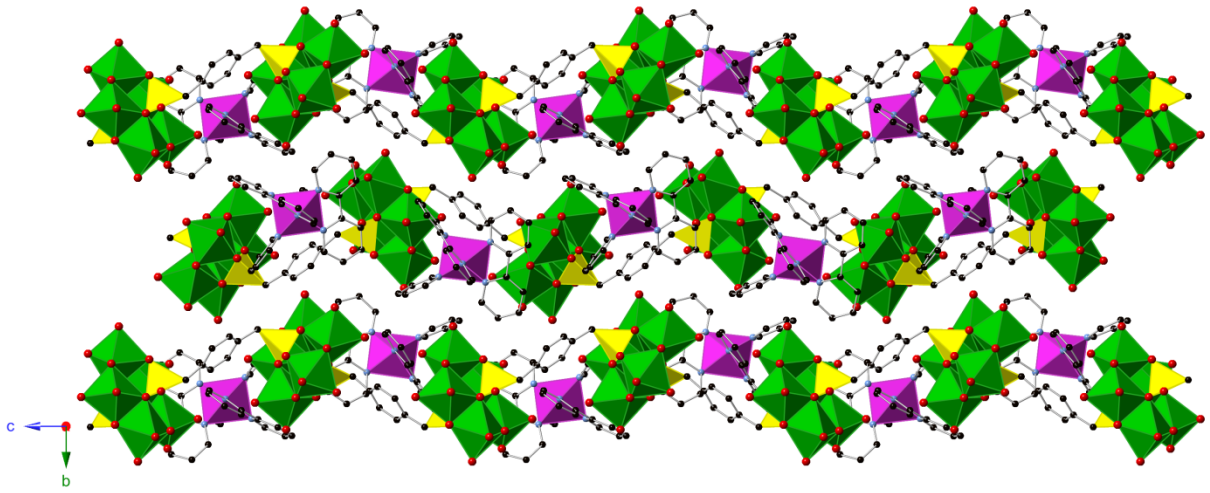
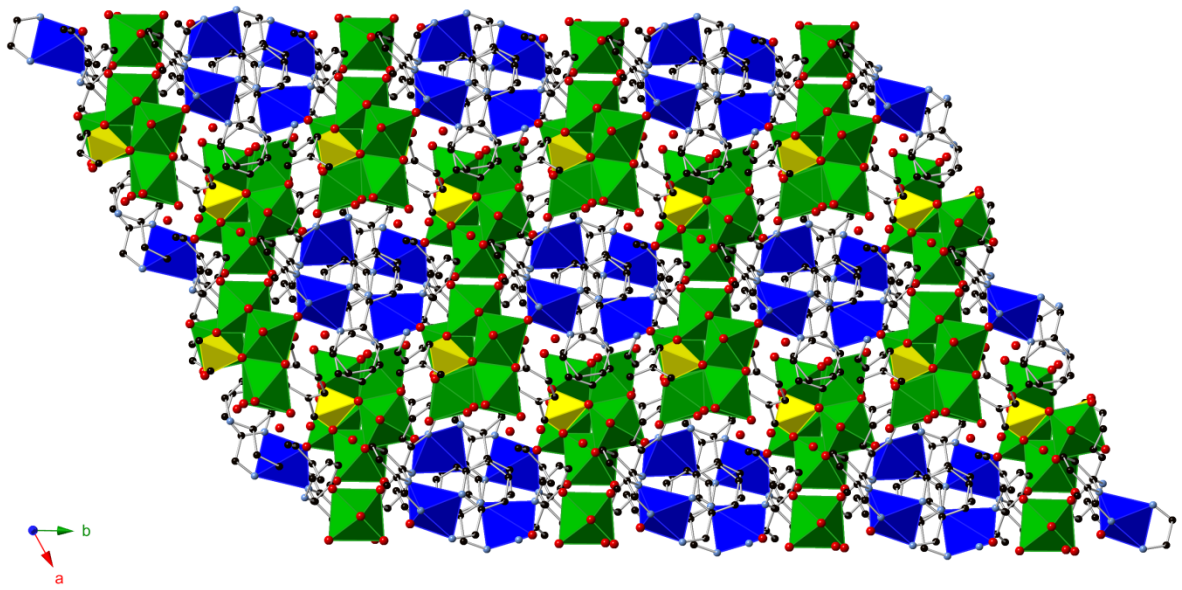


(**b**)

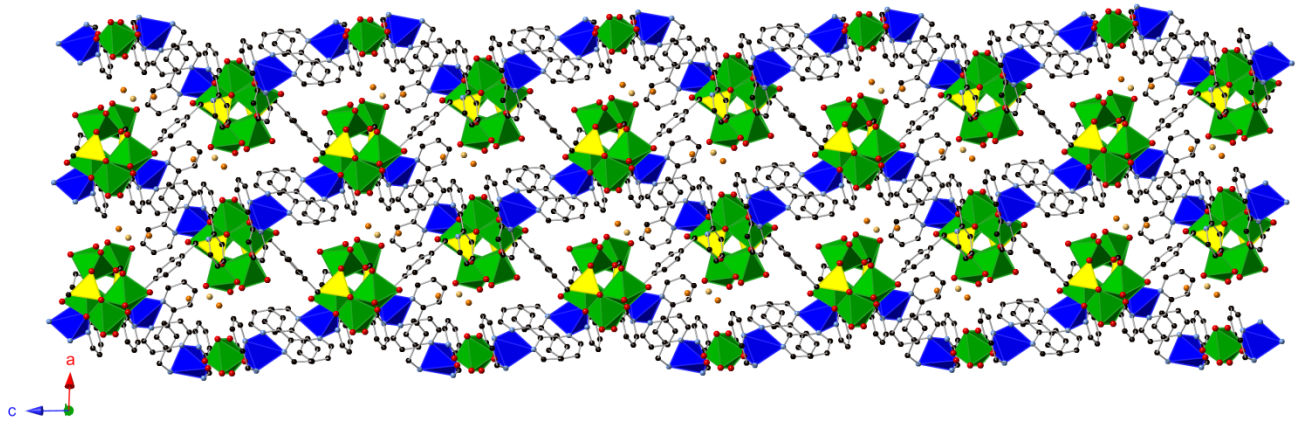


(**c**)

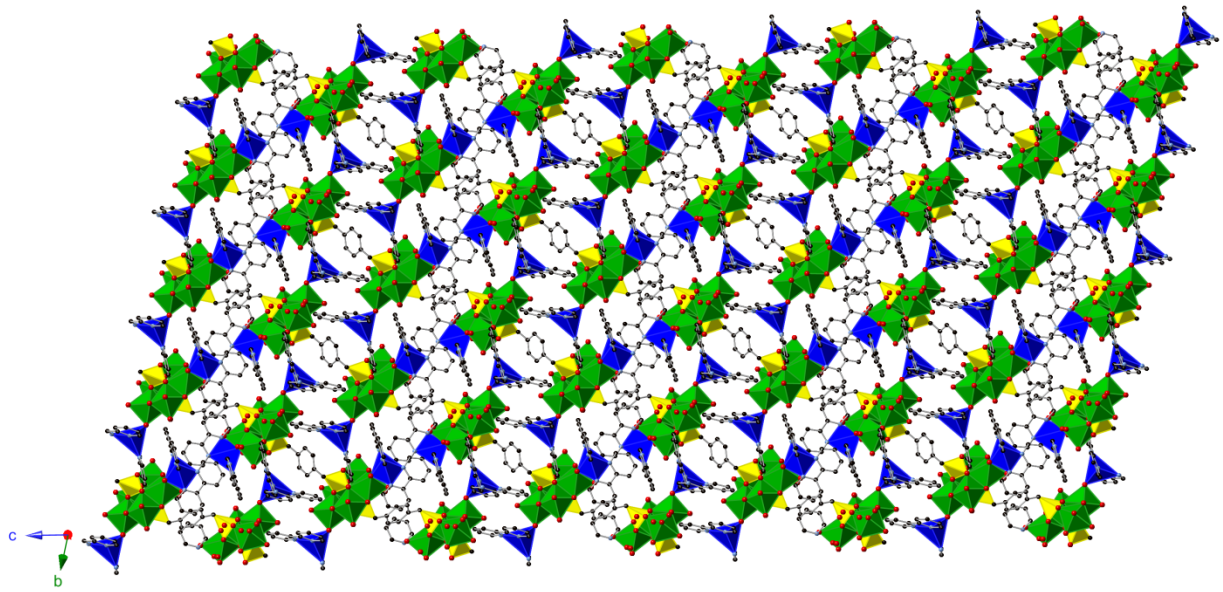
Figure S18. Packing view of **4**·6H_2O in the (**a**) *ab* plane (**b**) *ac* plane and (**c**) *bc* plane. Water molecules of crystallization removed for clarity.



(**a**)

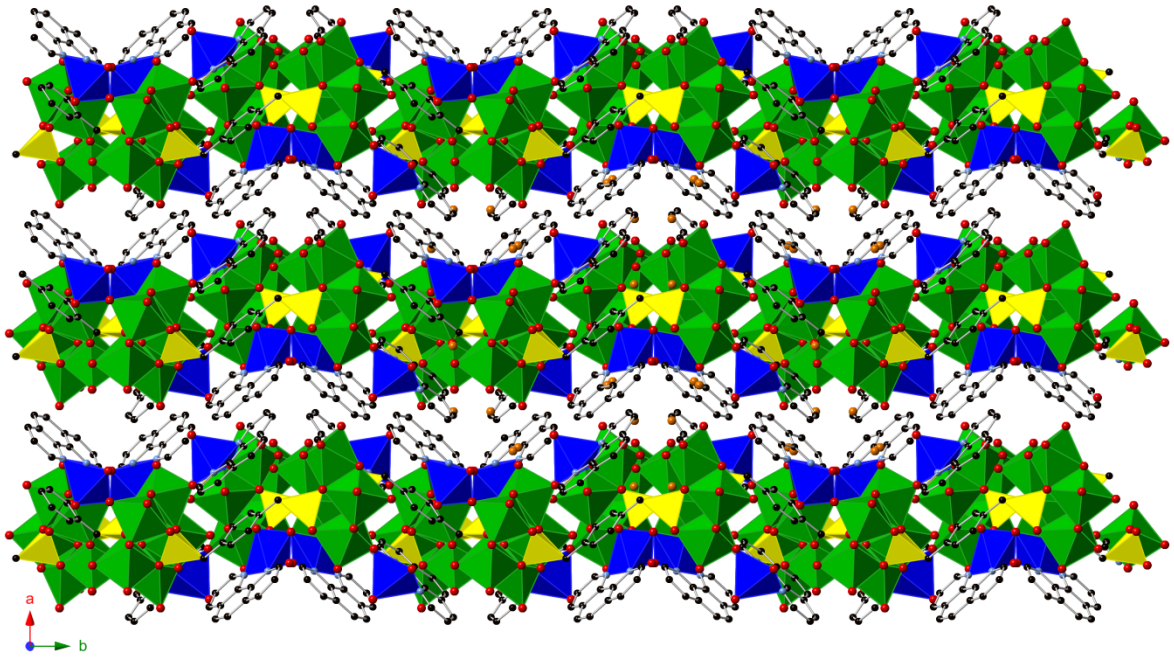


(**b**)

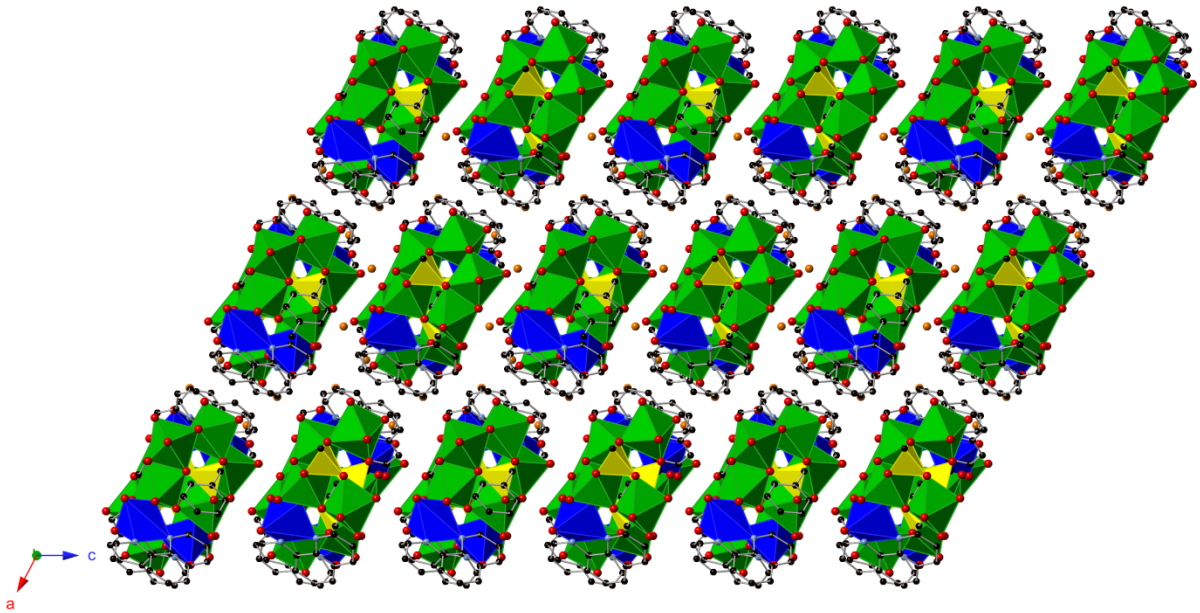


(**c**)

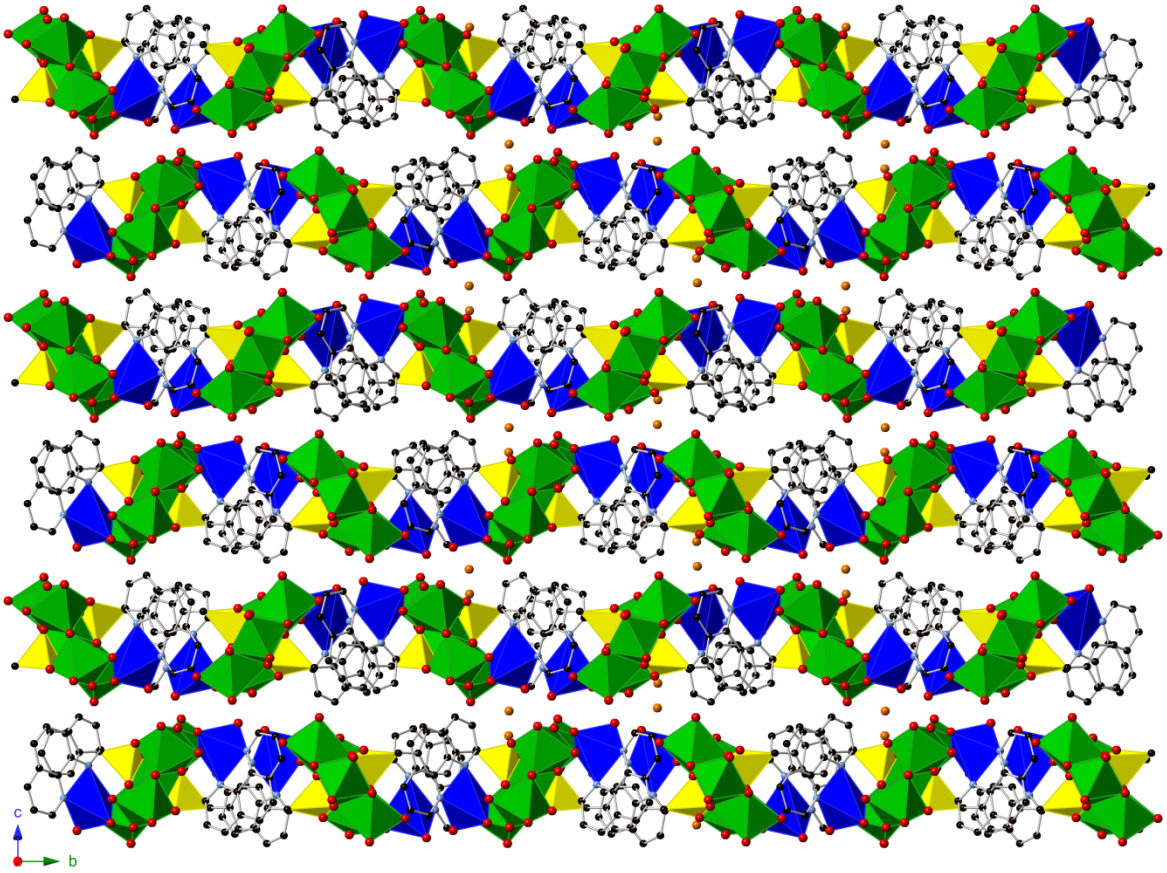
Figure S19 Packing view of **5**·6H_2O in the (**a**) *ab* plane (**b**) *ac* plane (orange spheres: water molecules of crystallization) and (**c**) *bc* plane. Water molecules of crystallization are removed for clarity unless otherwise noted.



(**a**)

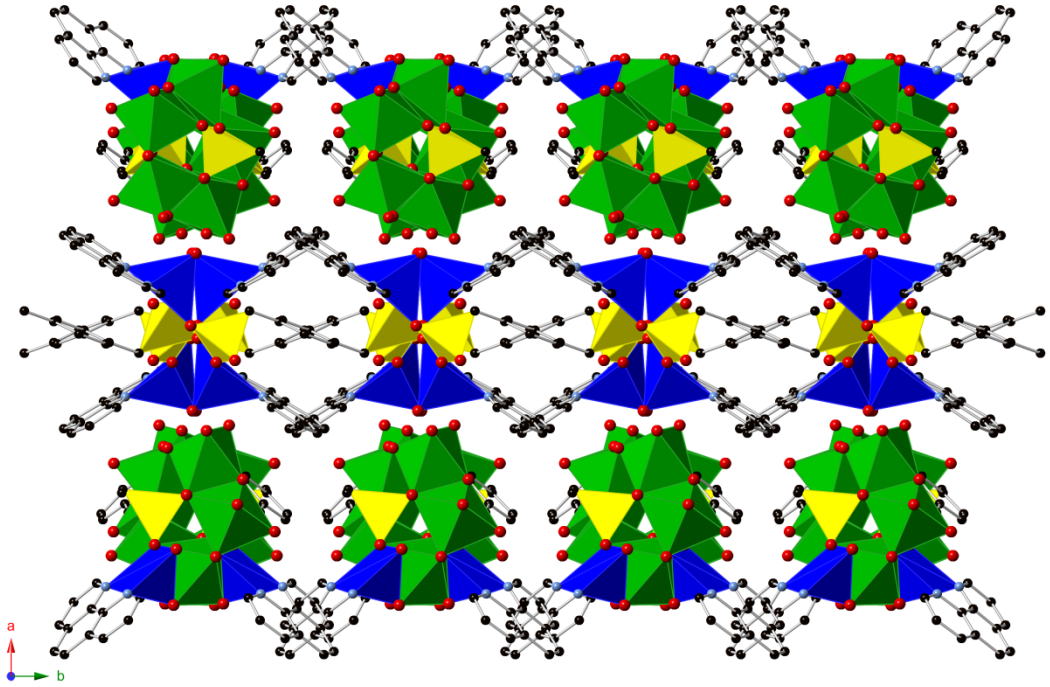


(**b**)

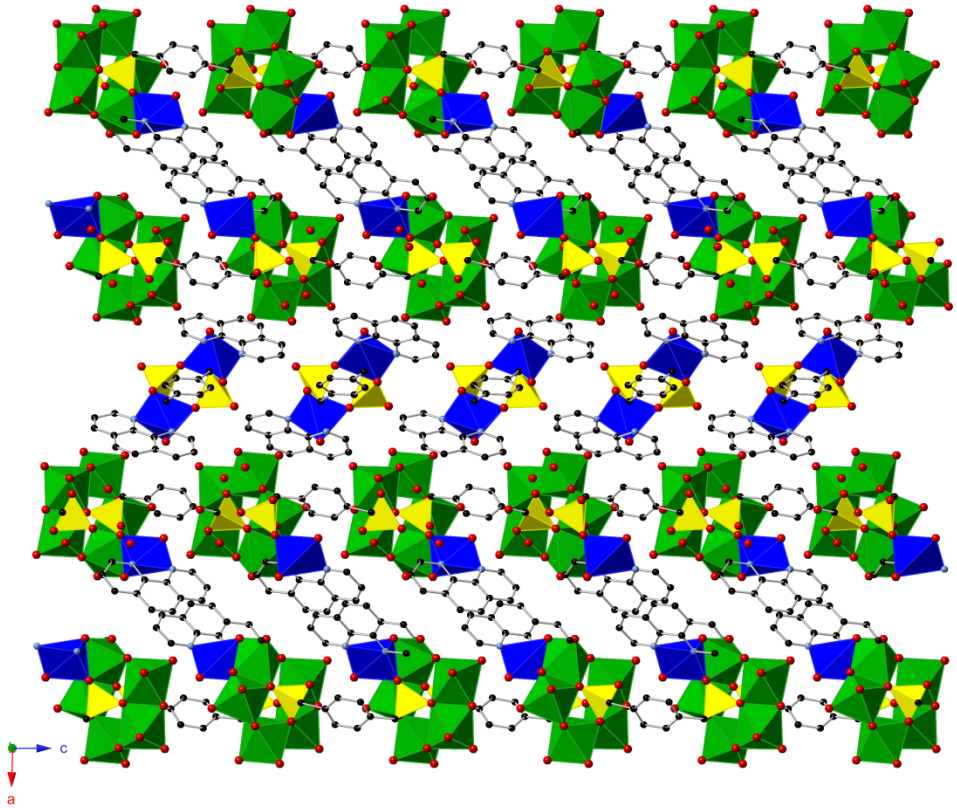


(**c**)

Figure S20. Packing view of **6**·4H_2O Packing view of **5**·6H_2O in the (**a**) *ab* plane (**b**) *ac* plane and (**c**) *bc* plane. Water molecules of crystallization are removed for clarity.



(**a**)



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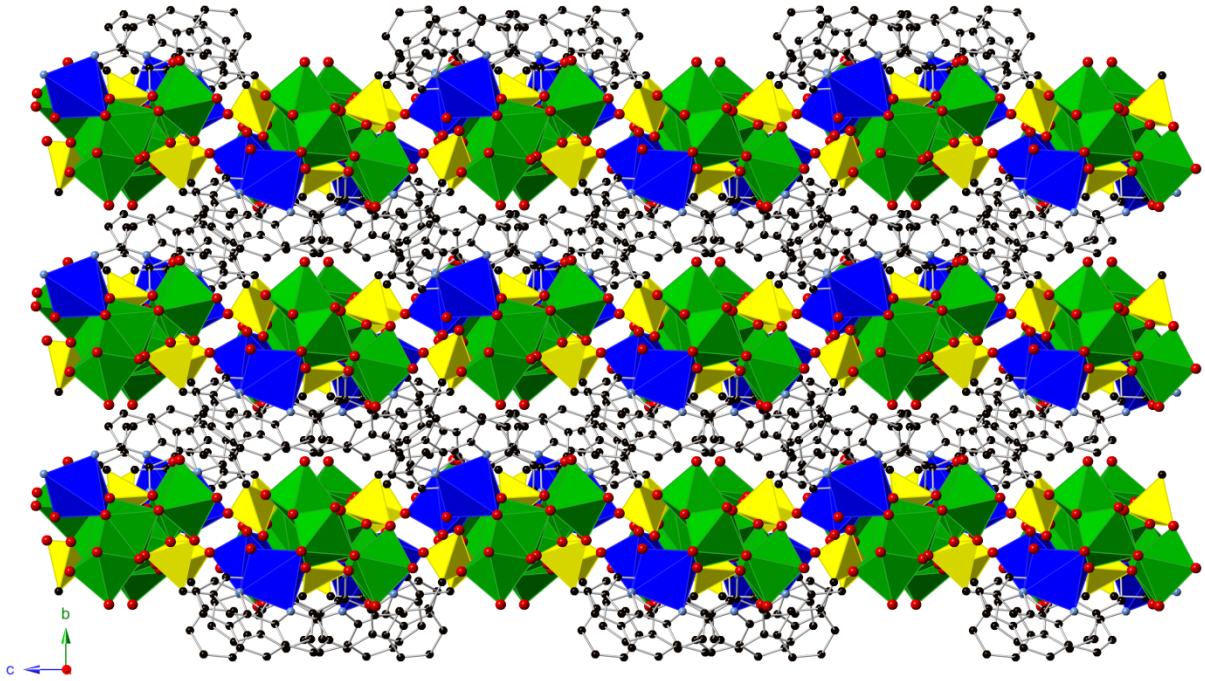
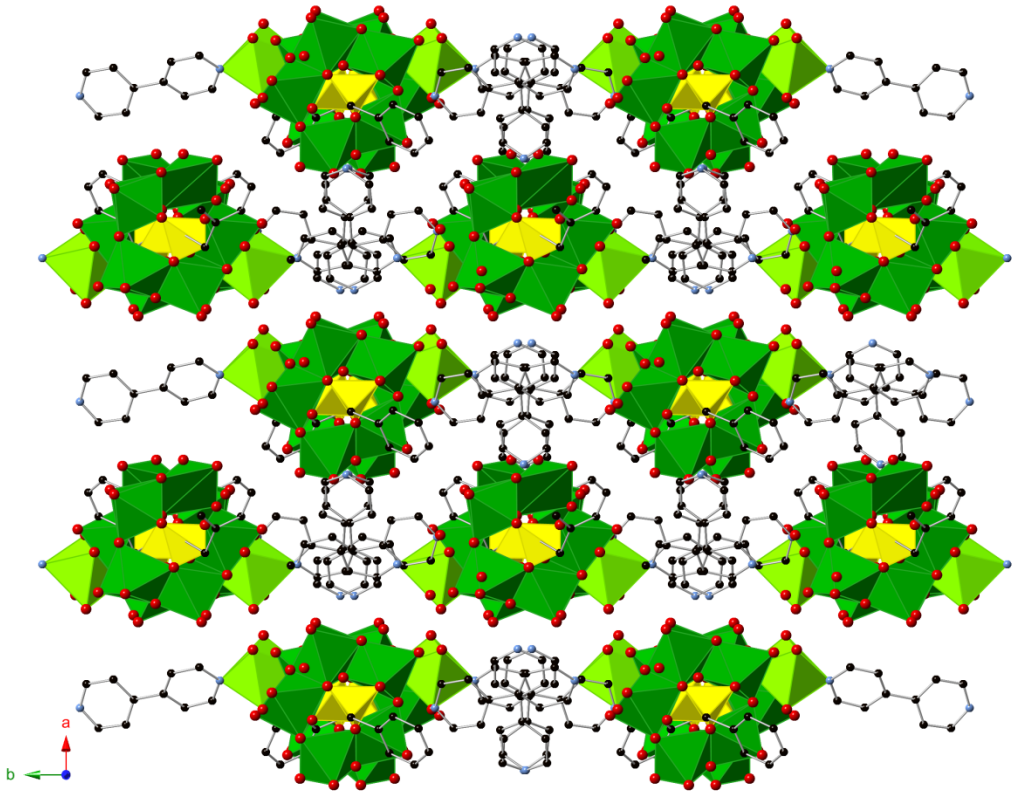
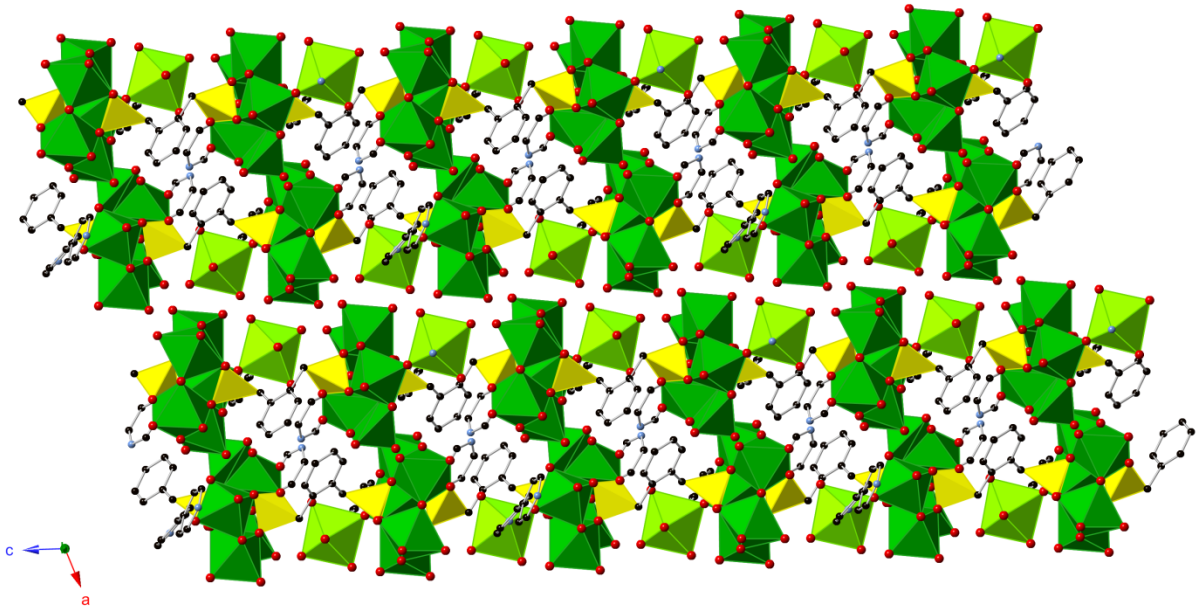


(**c**)

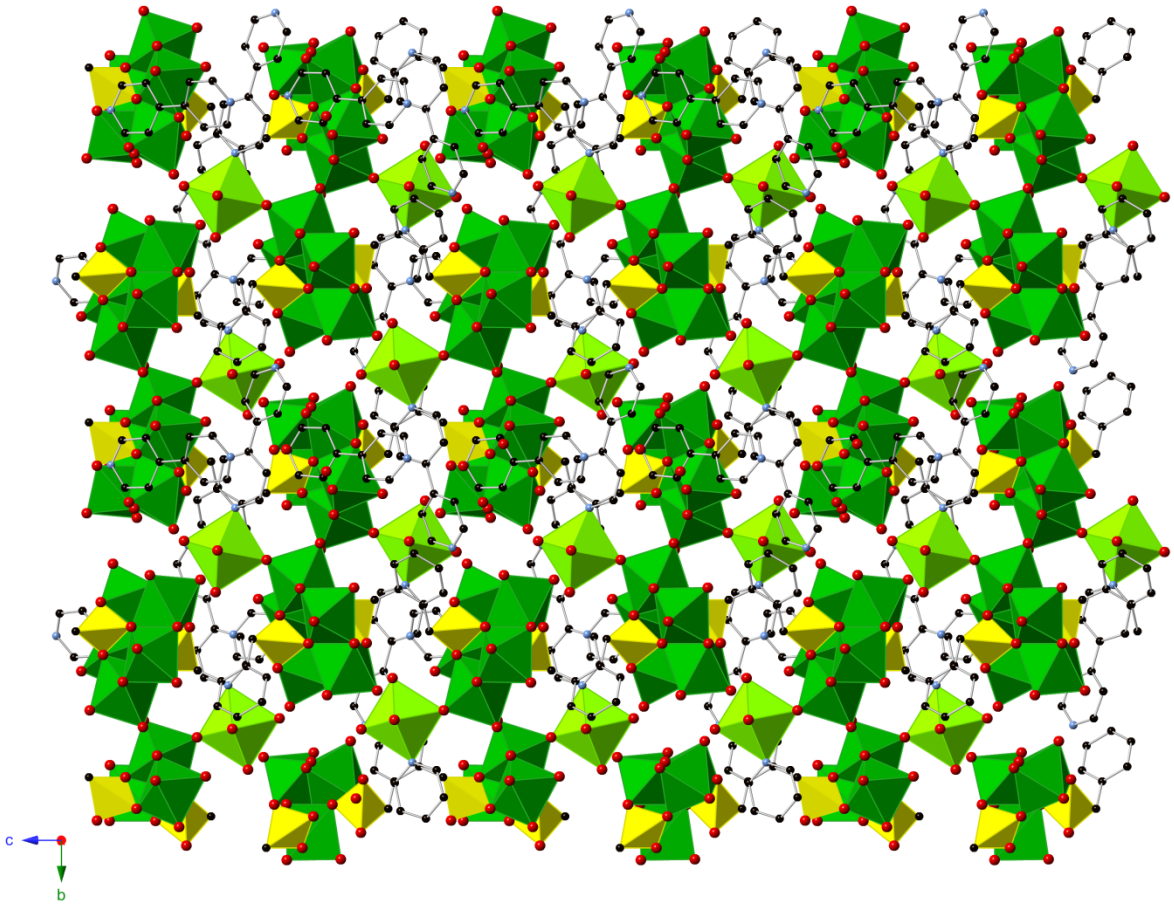
Figure S21. Packing view of **7** in the (**a**) *ab* plane (**b**) *ac* plane and (**c**) *bc* plane.



(**a**)

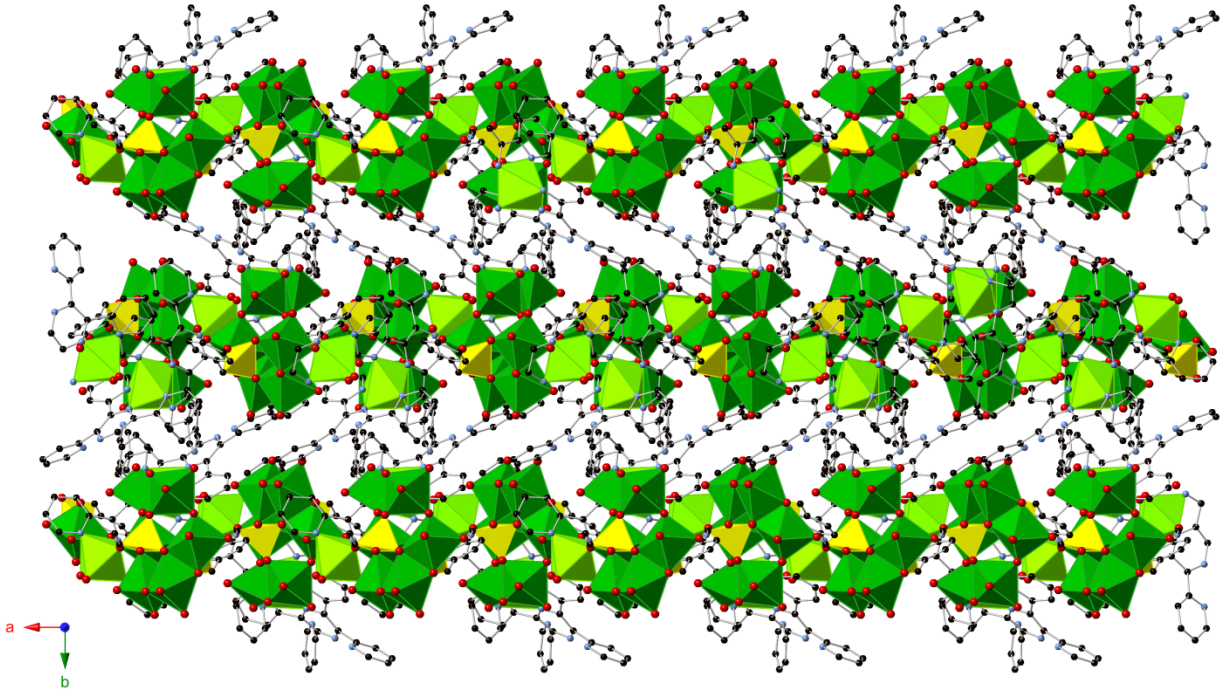


(**b**)

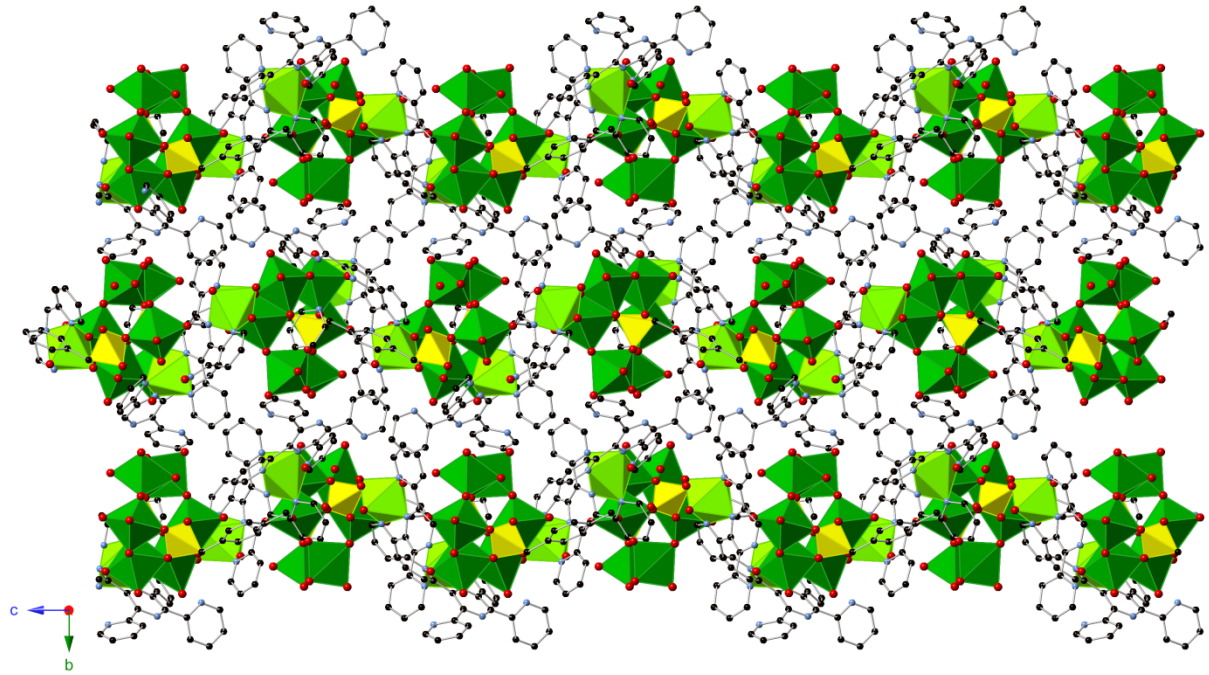


(**c**)

Figure S22. Packing view of **8**·4H_2O in the (**a**) *ab* plane (**b**) *ac* plane and (**c**) *bc* plane. Water molecules of crystallization are removed for clarity.

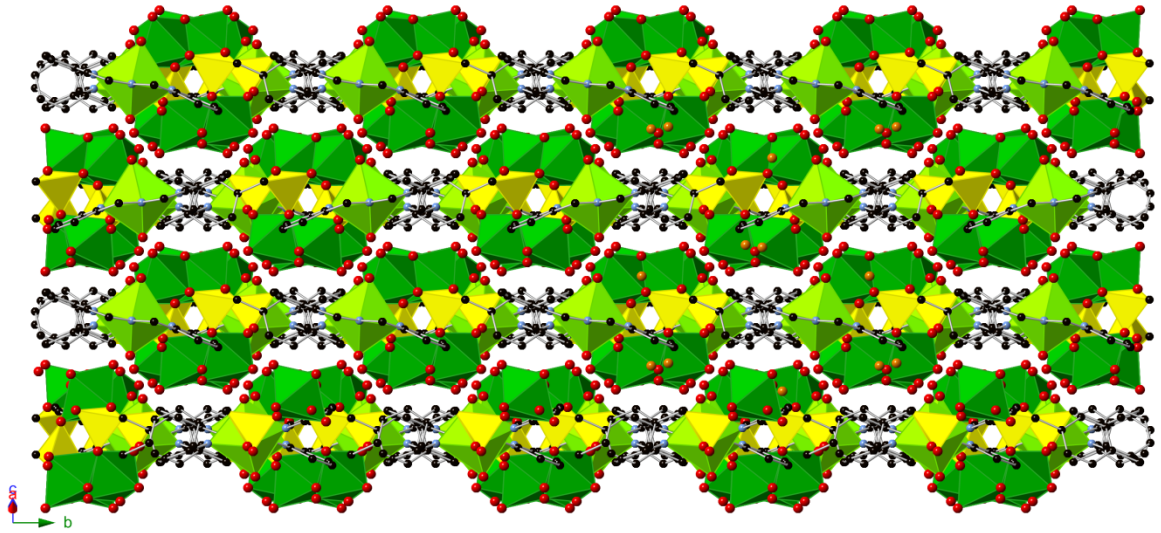


(**a**)

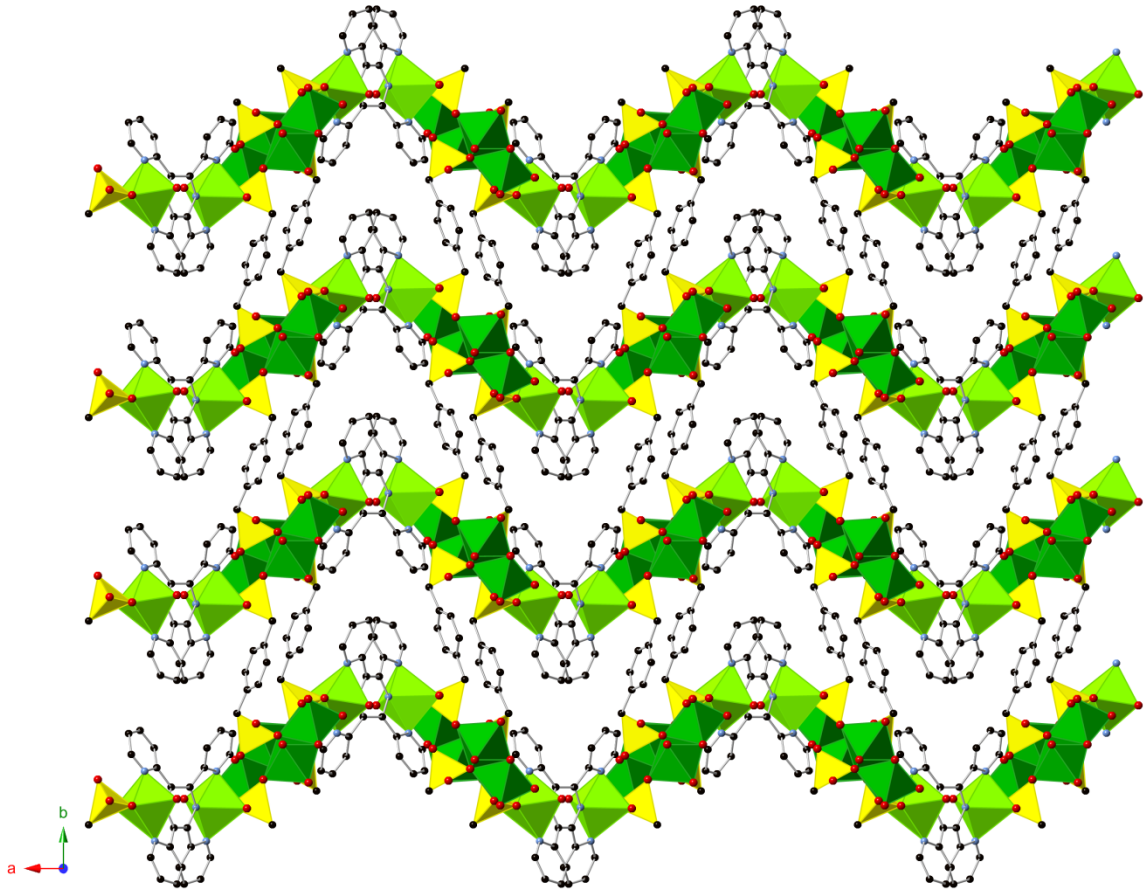


(**b**)

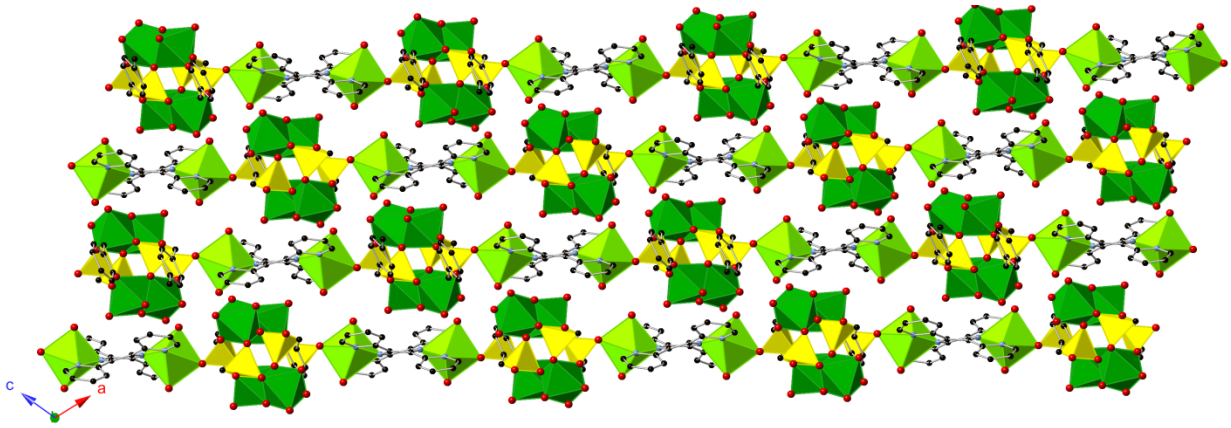
Figure S23. Packing view of **9**·6H_2O in the (**a**) *ab* plane and (**b**) *bc* plane. Water molecules of crystallization are removed for clarity.



(**a**)

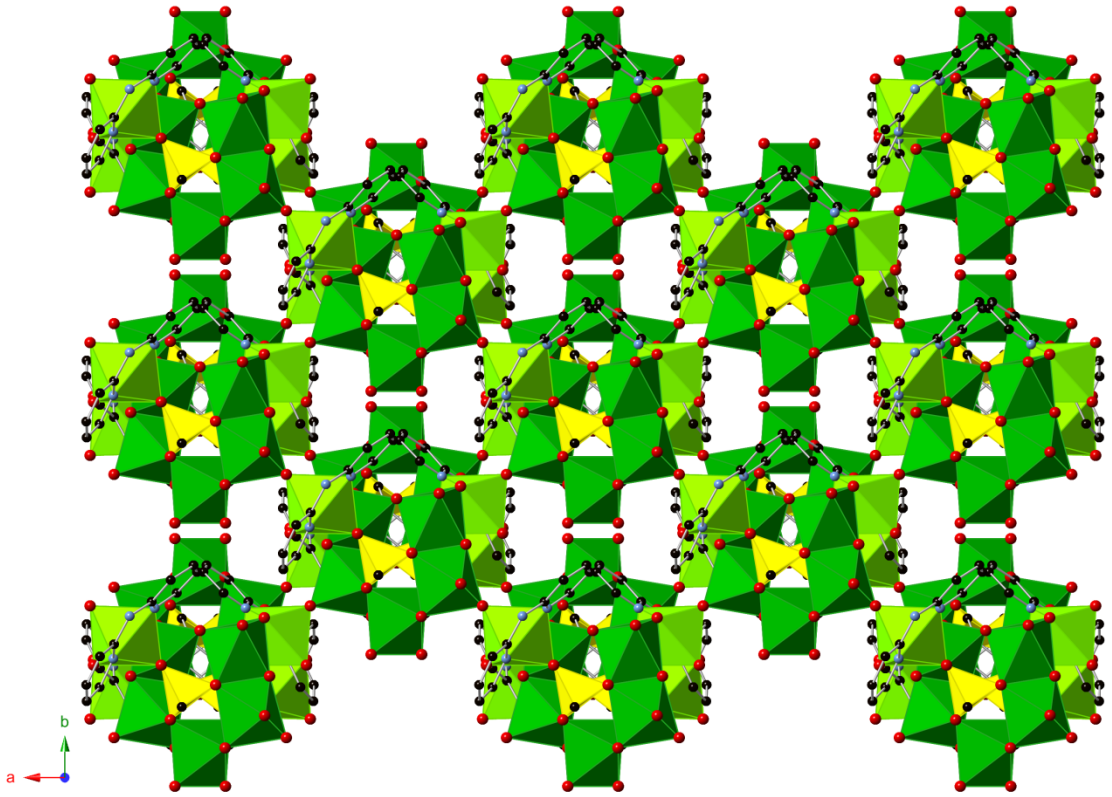


(**b**)

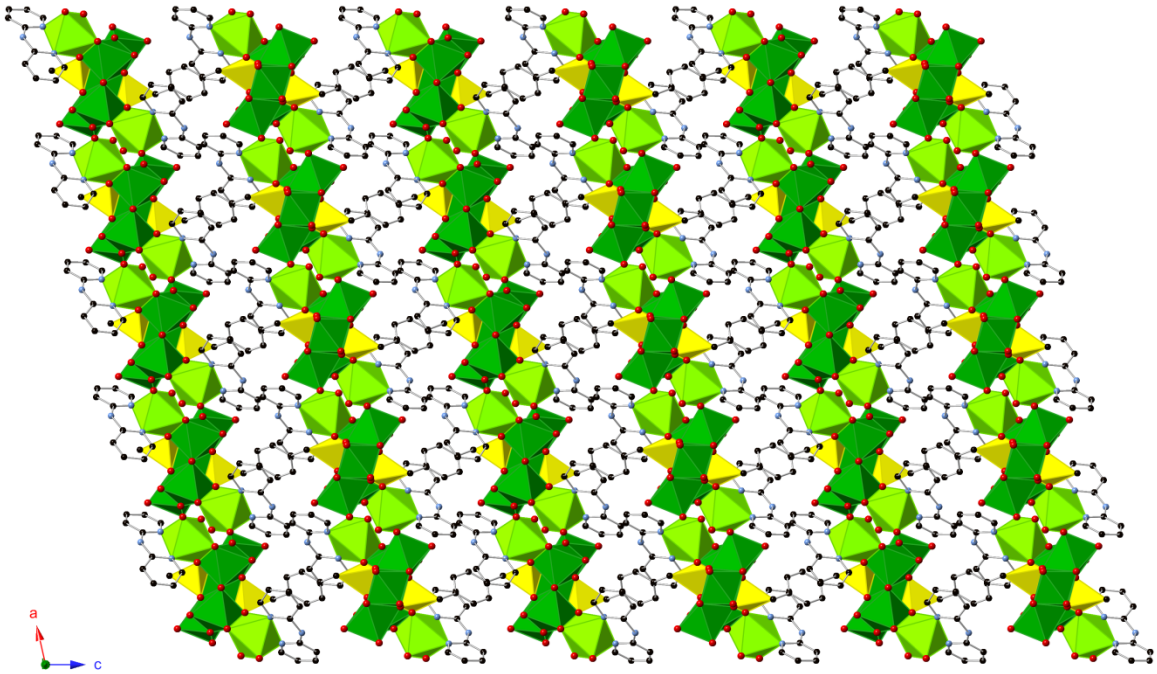


(**c**)

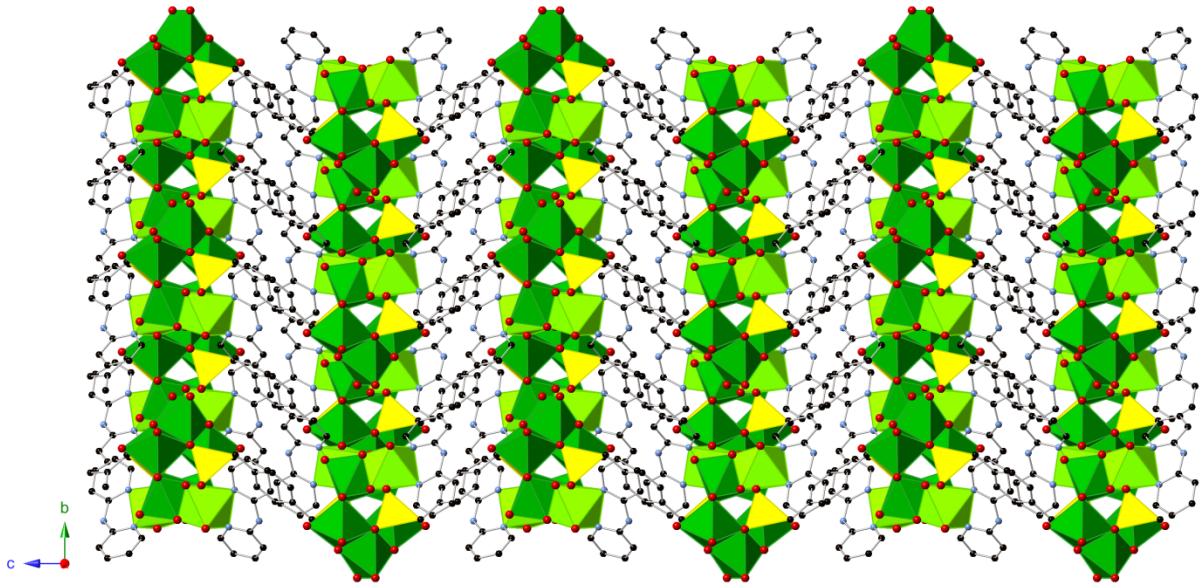
Figure S24. Packing view of **10**·4H_2O in the (**a**) *101* plane (orange spheres: water molecules of crystallization) (**b**) *ab* plane and (**c**) *ac* plane. Water molecules of crystallization are removed for clarity unless otherwise noted.



(**a**)

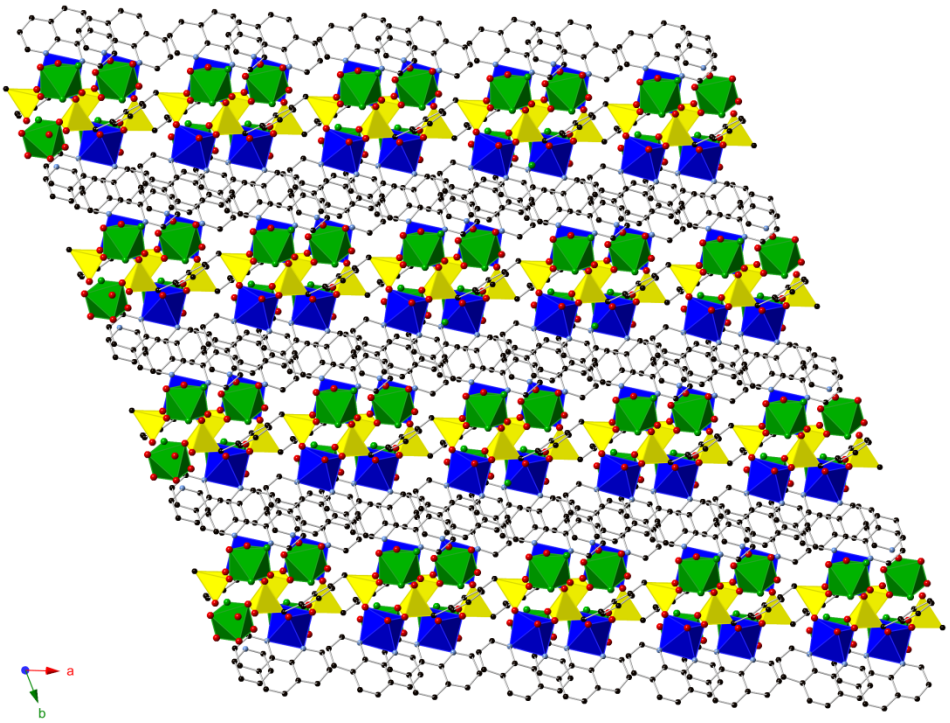


(**b**)

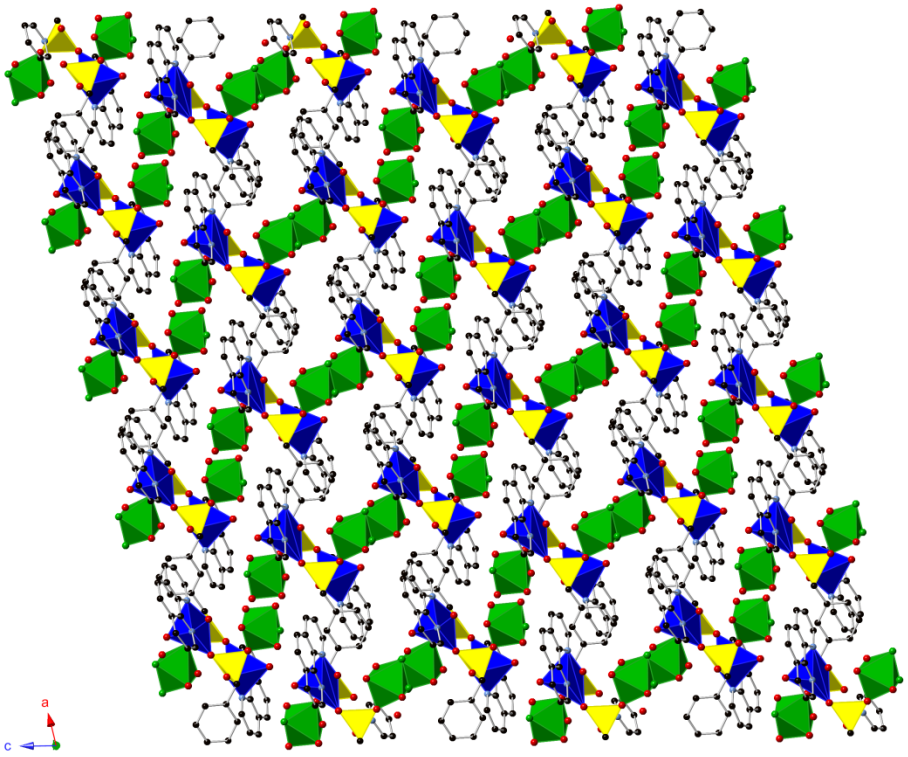


(**c**)

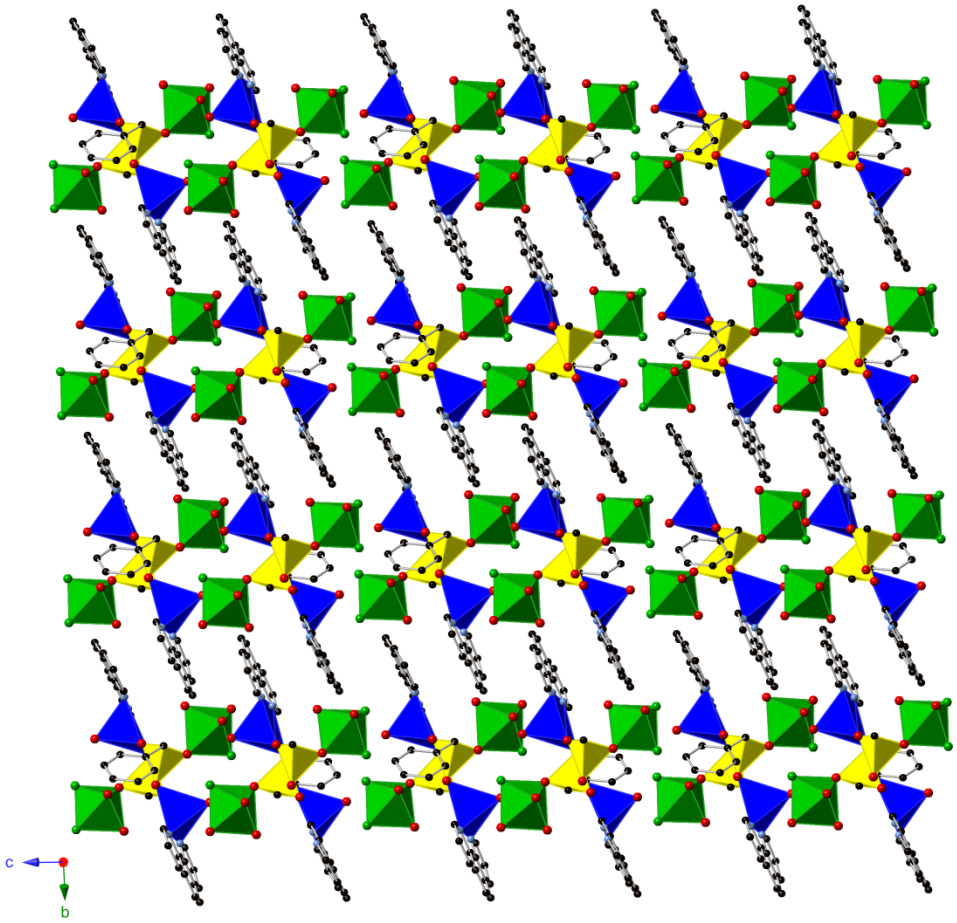
Figure S25. Packing view of **11**·2H_2O in the (**a**) *ab* plane (**b**) *ac* plane and (**c**) *bc* plane. Water molecules of crystallization are removed for clarity.



(**a**)

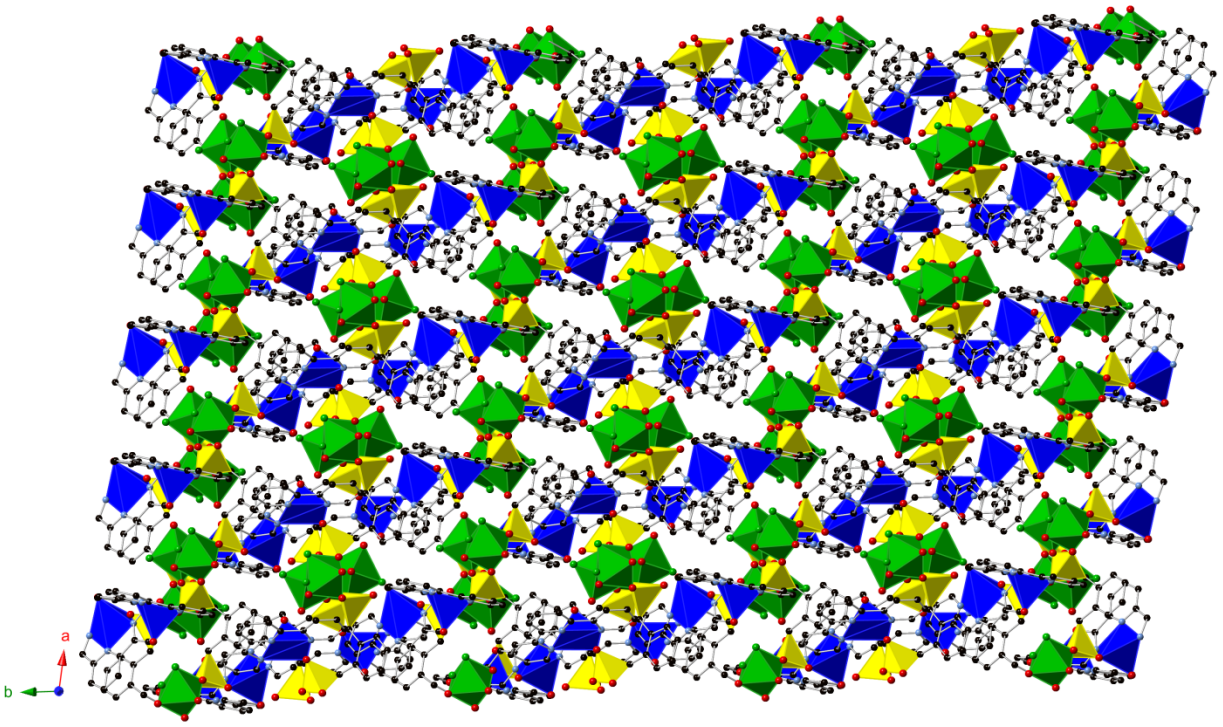


(**b**)

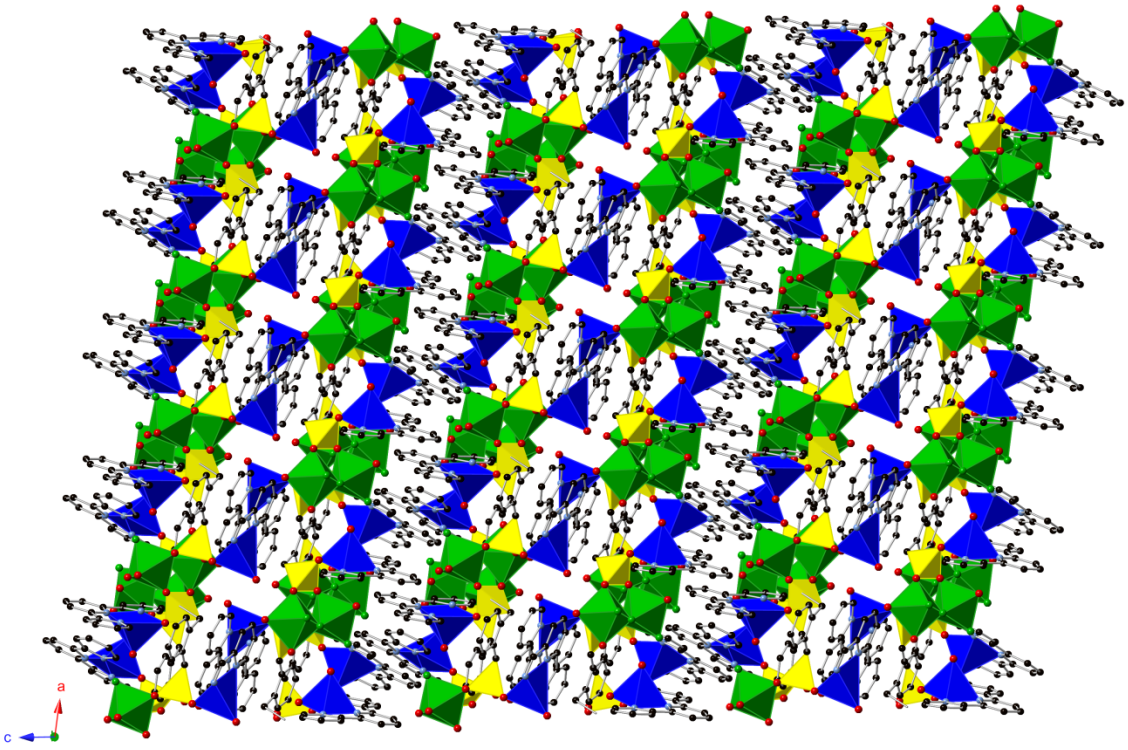


(**c**)

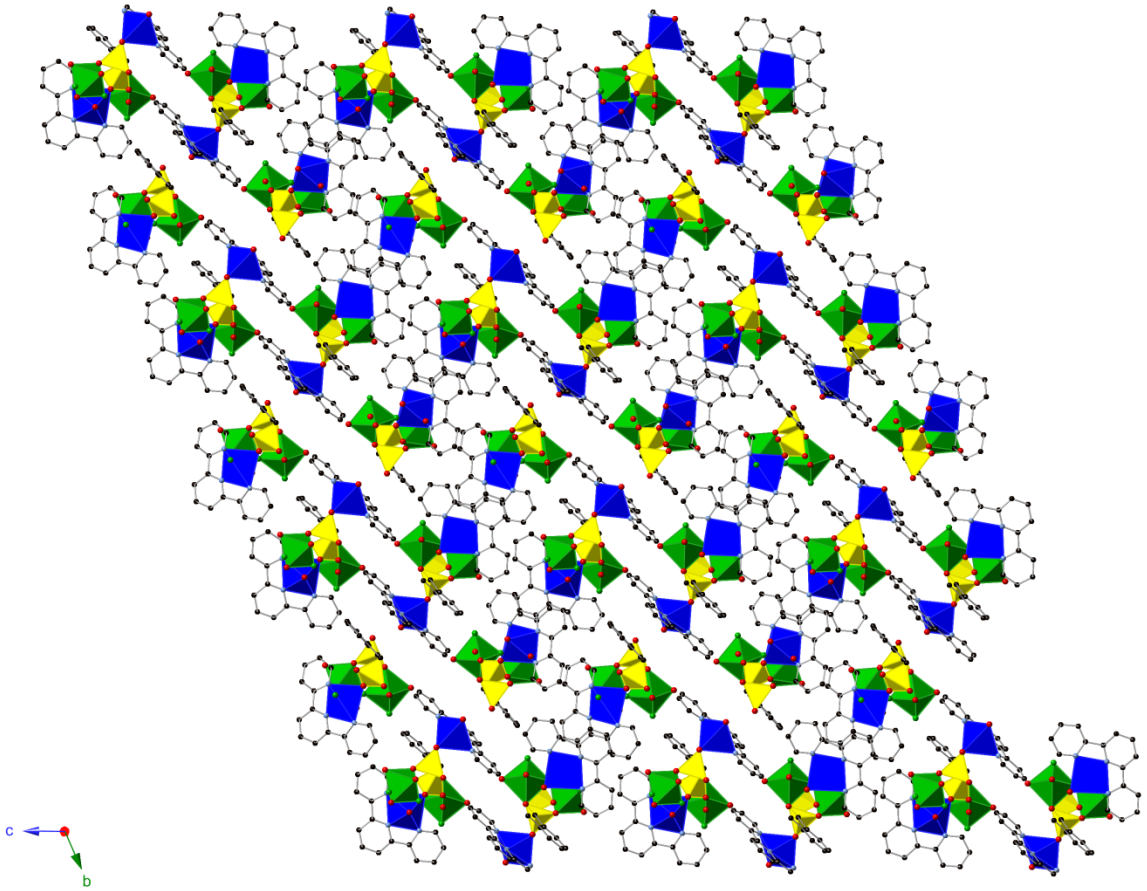
Figure S26. Packing view of **12**·H_2O in the (**a**) *ab* plane (**b**) *ac* plane and (**c**) *bc* plane. Water molecules of crystallization are removed for clarity.



(**a**)

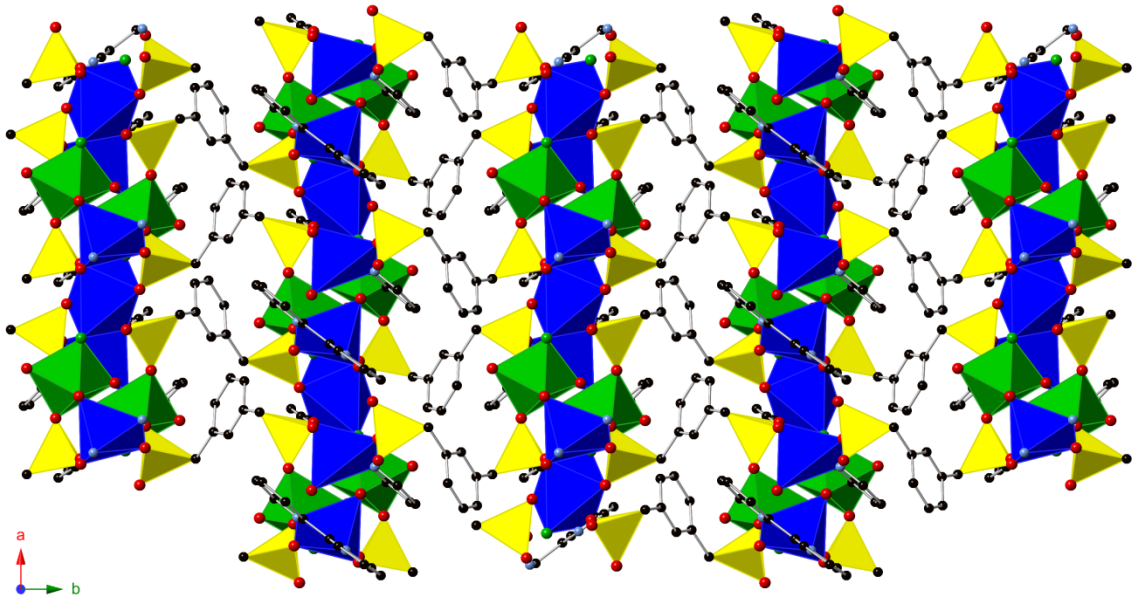


(**b**)

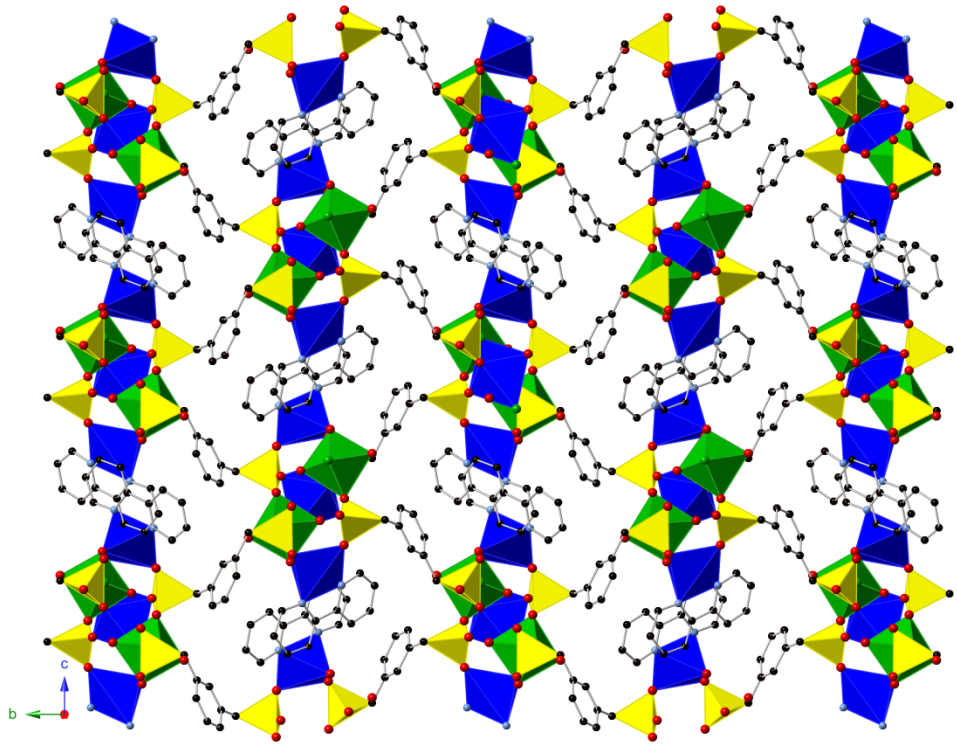


(**c**)

Figure S27. Packing view of **13**·H_2O in the (**a**) *ab* plane (**b**) *ac* plane and (**c**) *bc* plane. Water molecules of crystallization are removed for clarity.



(**a**)



(**b**)

Figure S28. Packing view of **14** in the (**a**) *ab* plane and (**b**) *bc* plane.

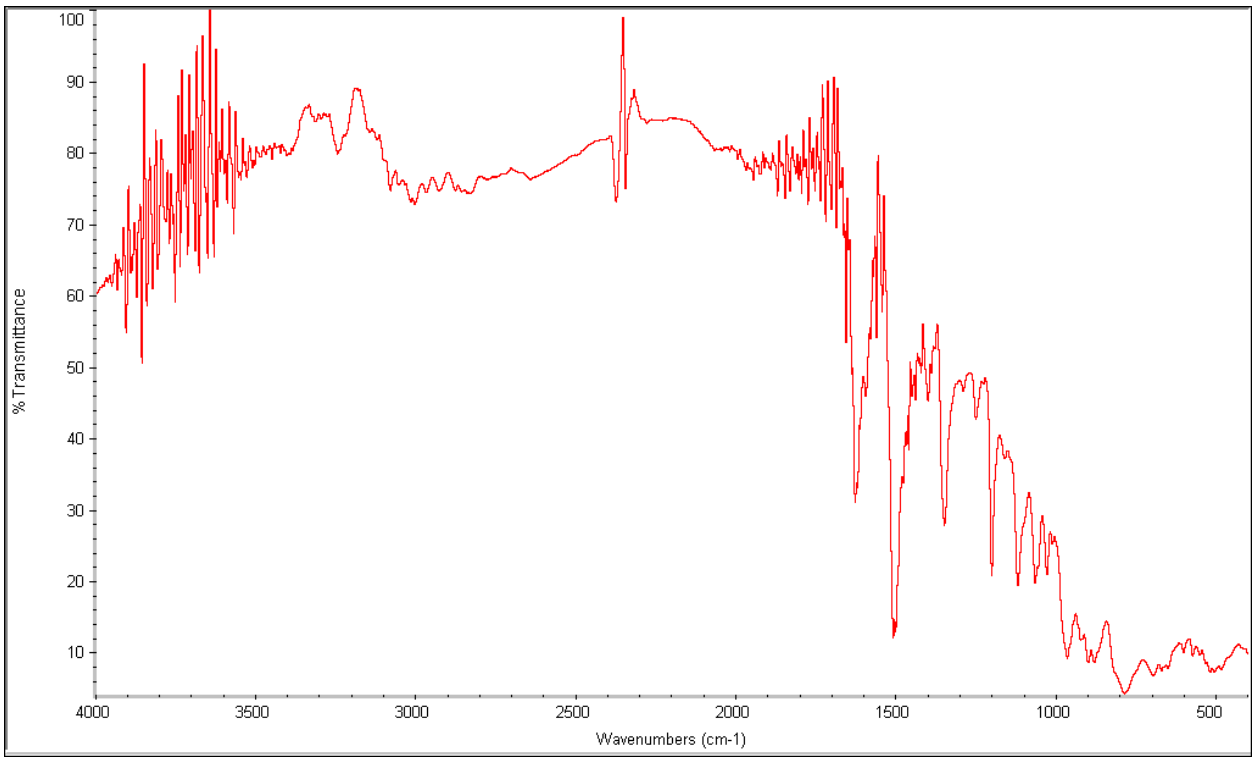


Figure S29. Infrared spectra for **1**·H_2O.

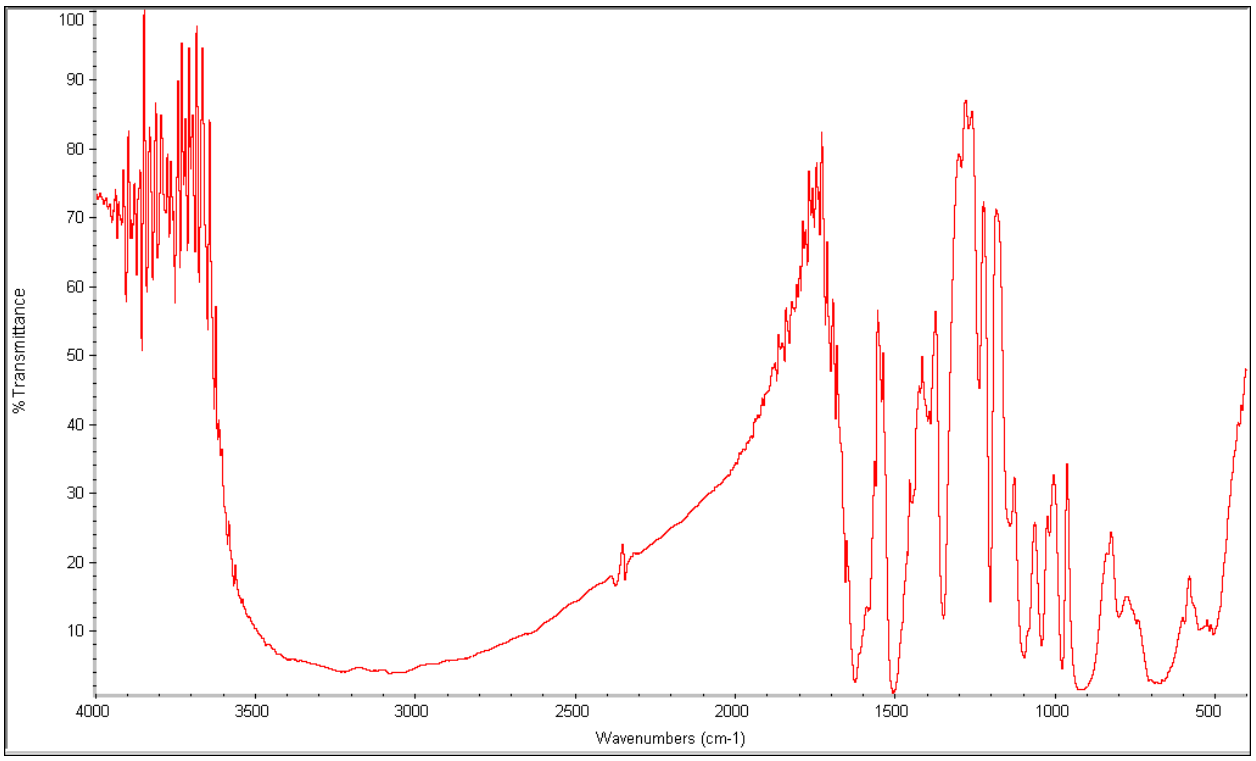


Figure S30. Infrared spectra for **2**.

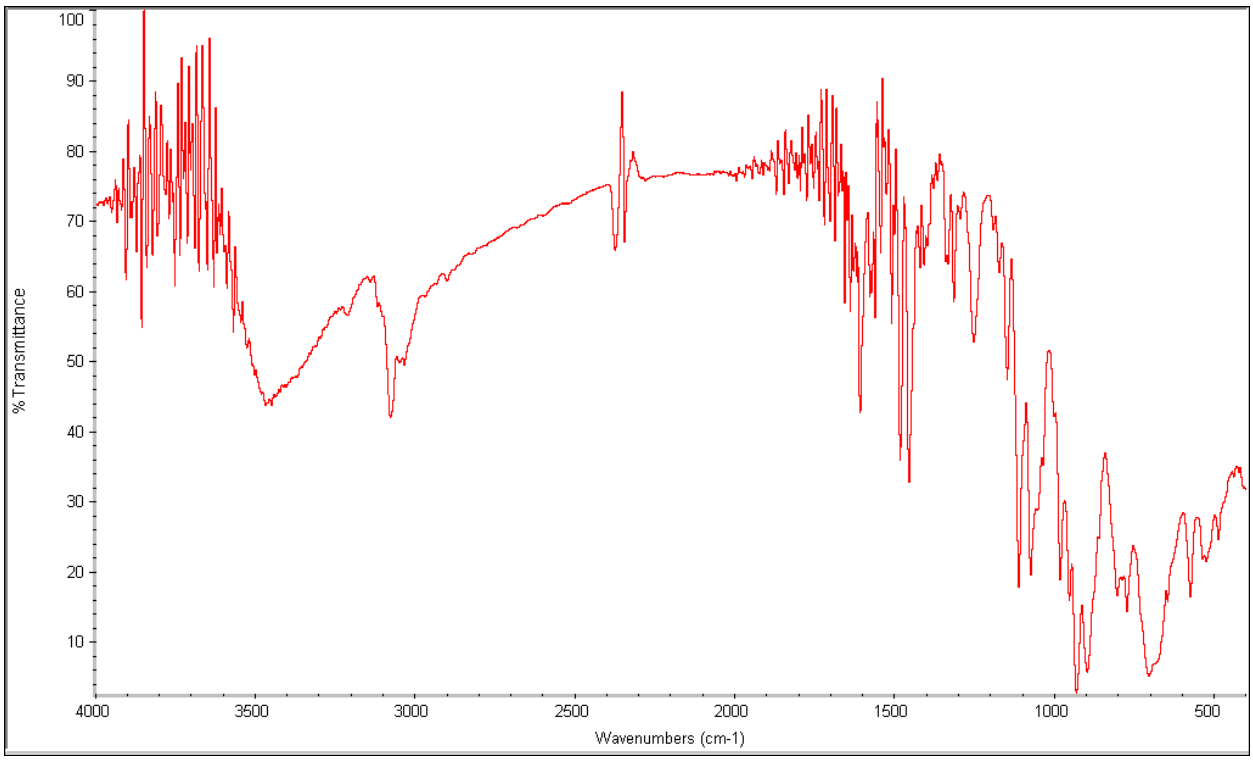


Figure S31. Infrared spectra for **3**·H_2O.

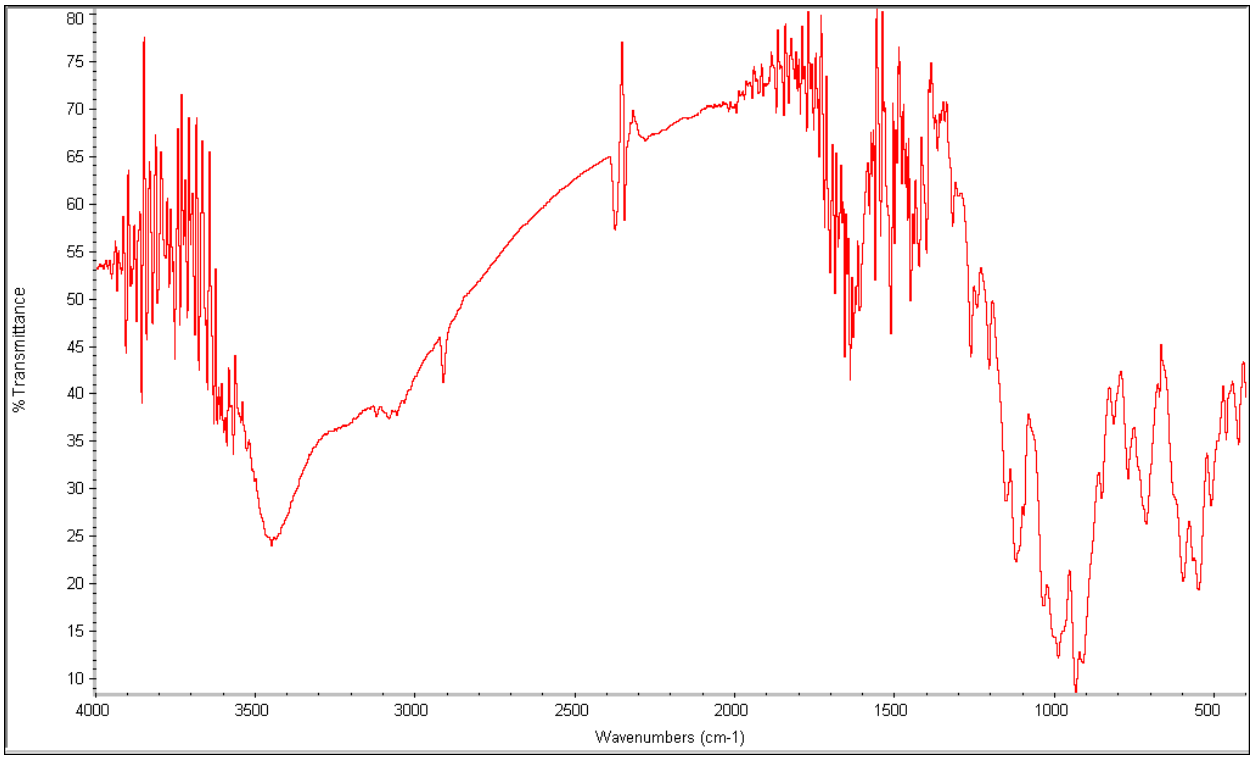


Figure S32. Infrared spectra for **4**·$6H_2O$.

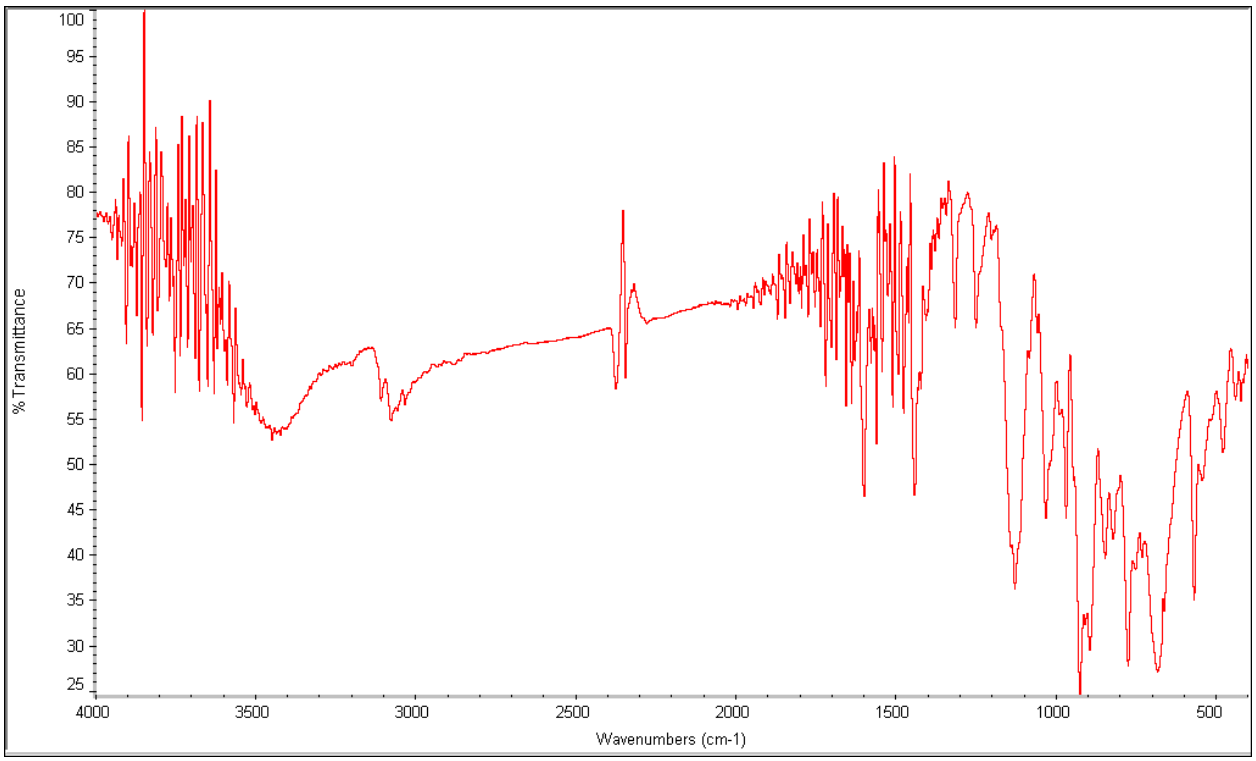


Figure S33. Infrared spectra for **5**·6H_2O.

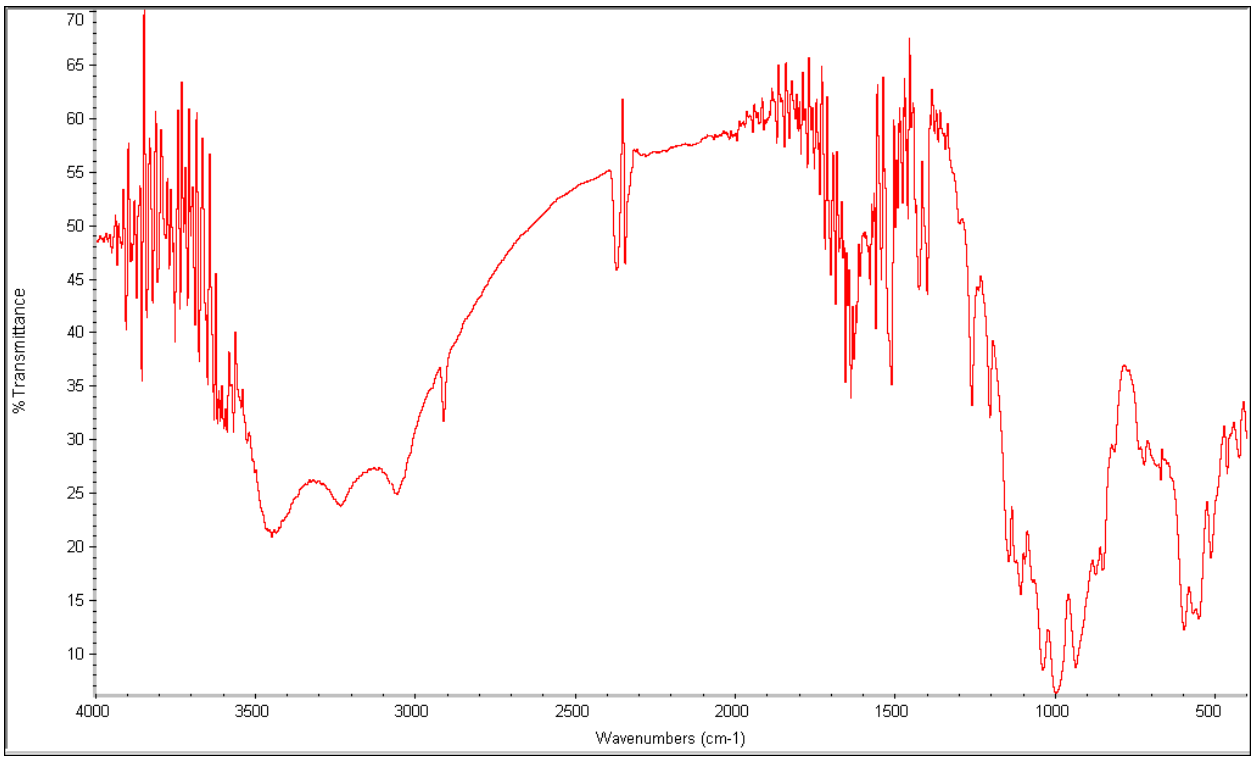


Figure S34. Infrared spectra for **6**·4H_2O.

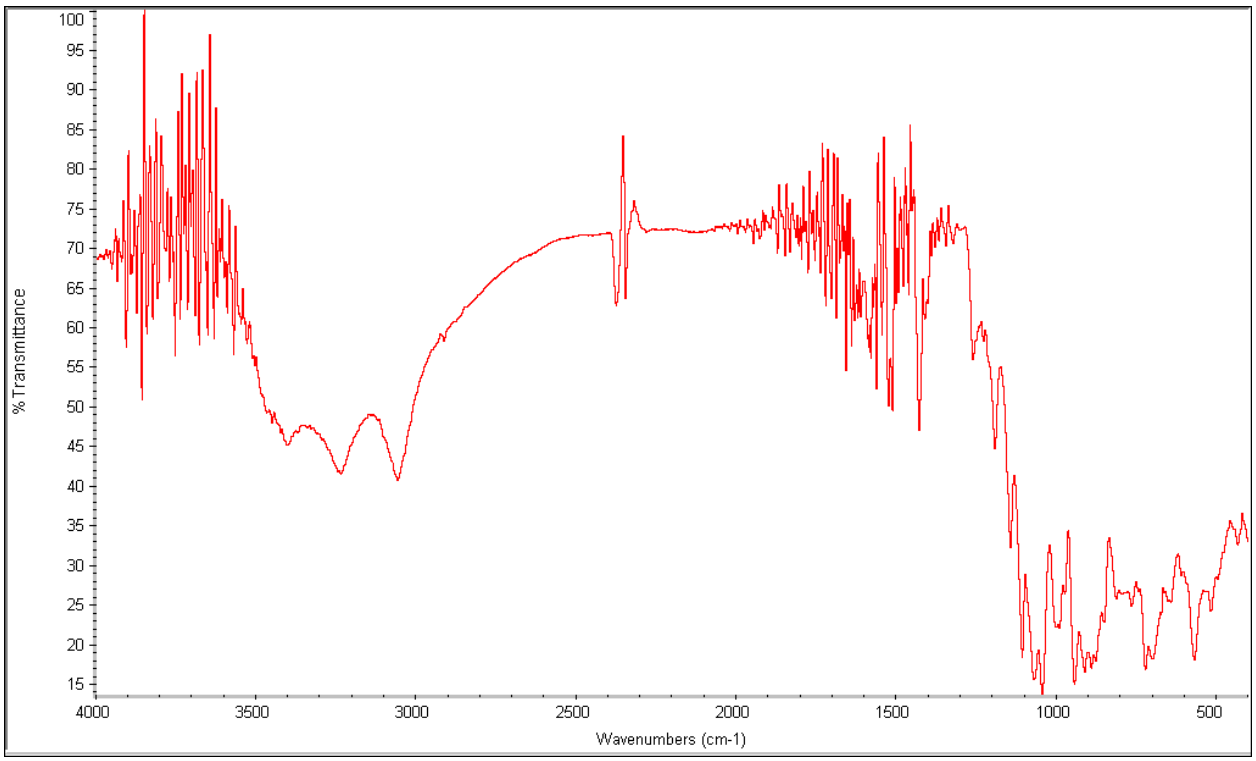


Figure S35. Infrared spectra for **7**.

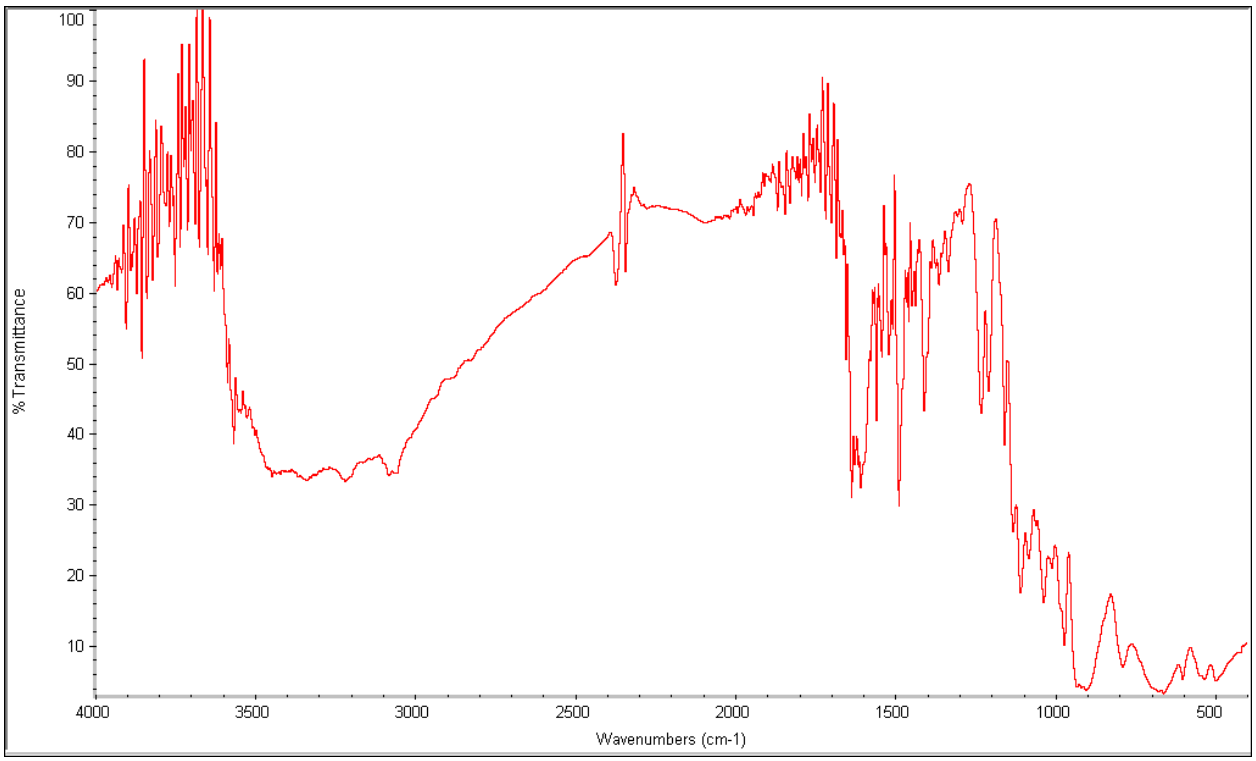


Figure S36. Infrared spectra for **8**·4H_2O.

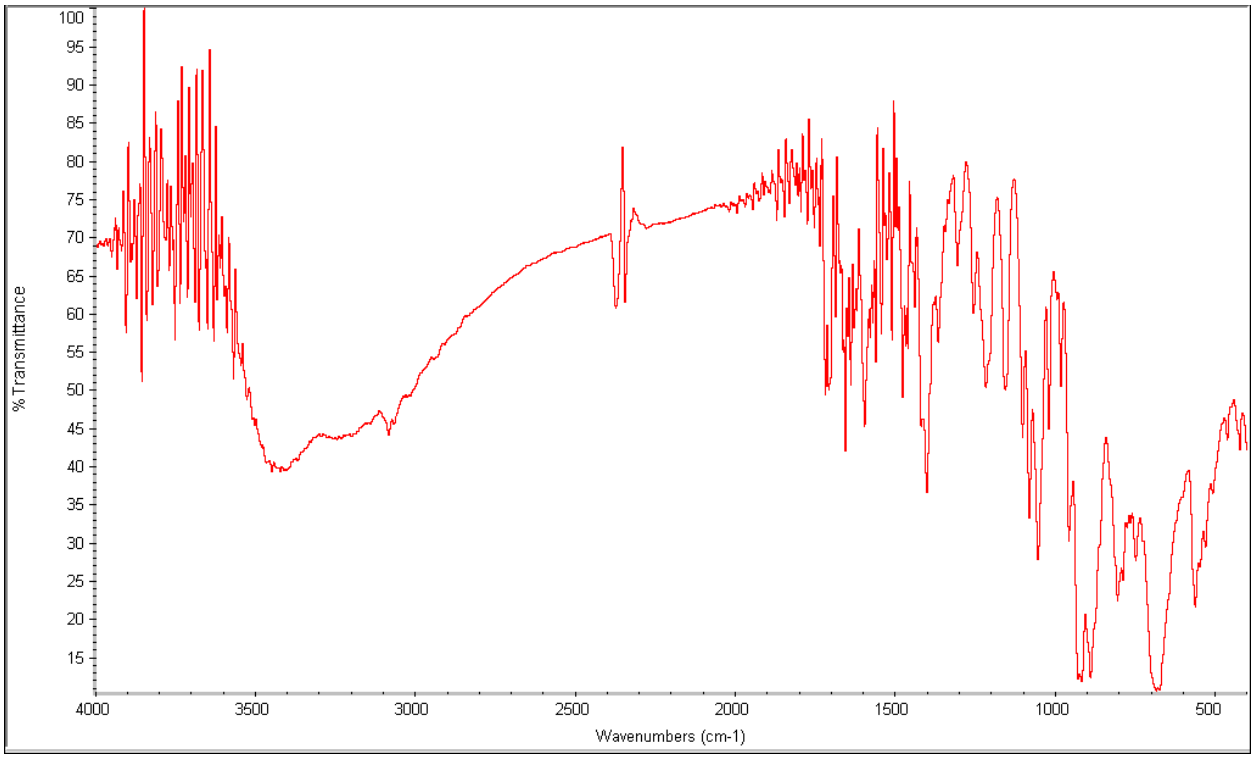


Figure S37. Infrared spectra for **9**·6H_2O.

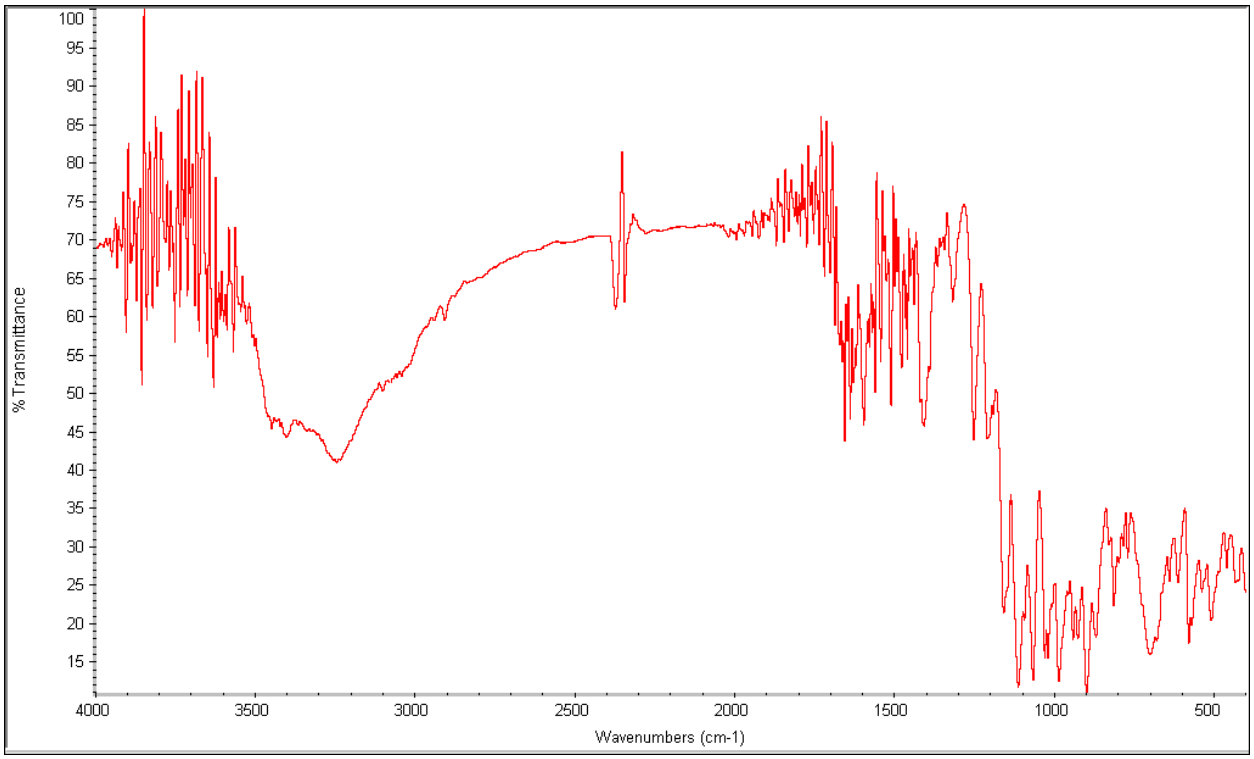


Figure S38. Infrared spectra for **10**·4H_2O.

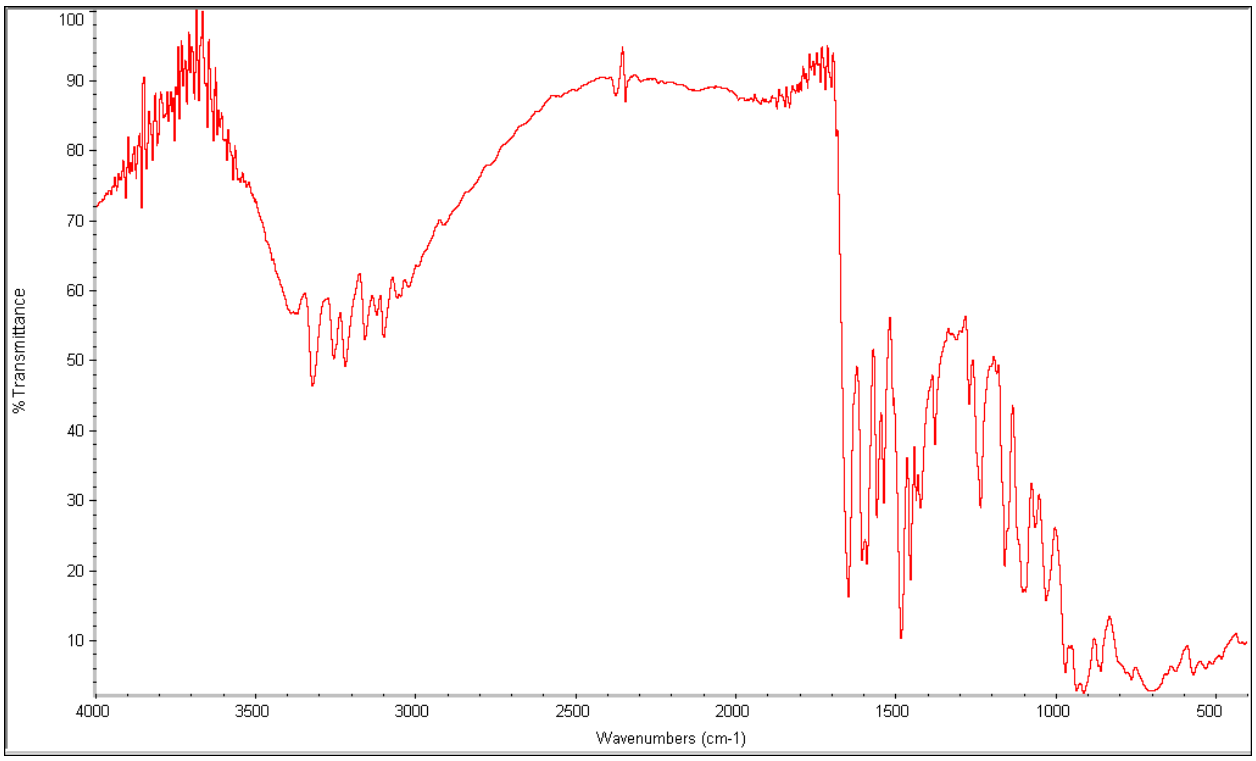


Figure S39. Infrared spectra for **11**·2H_2O.

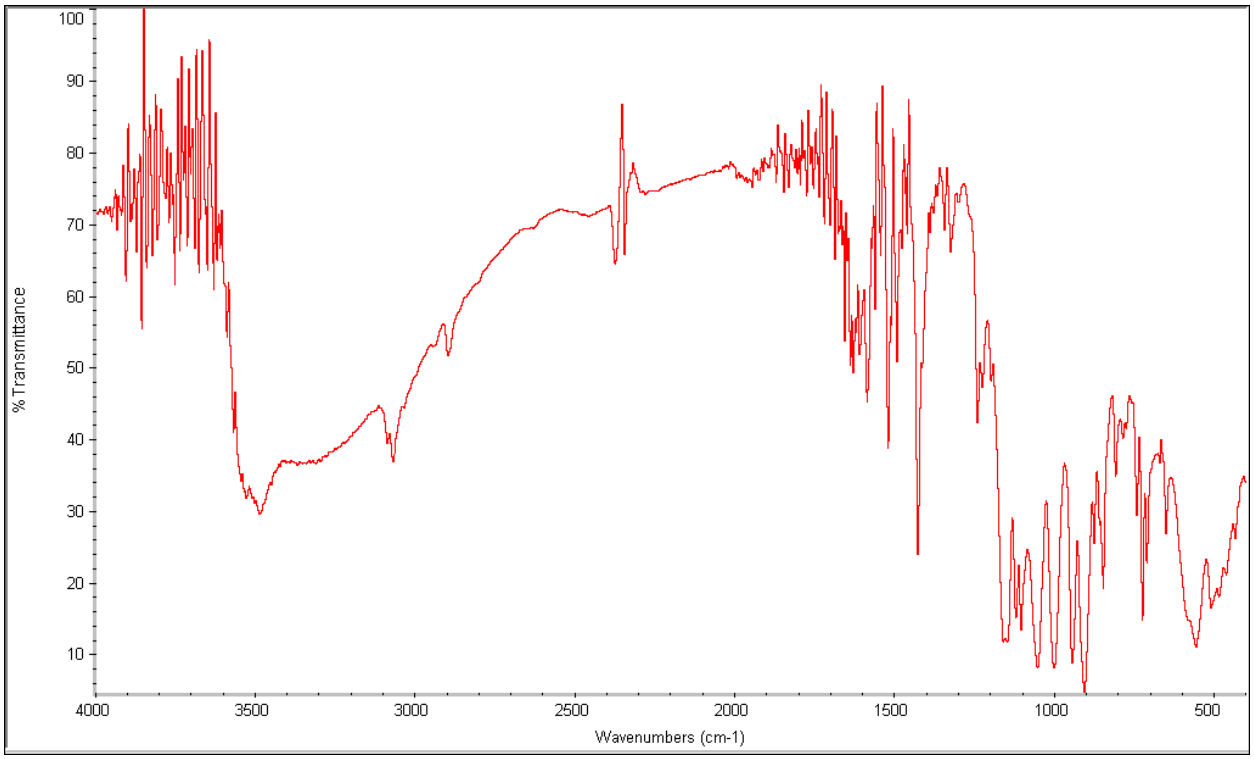


Figure S40. Infrared spectra for **12**·H_2O.

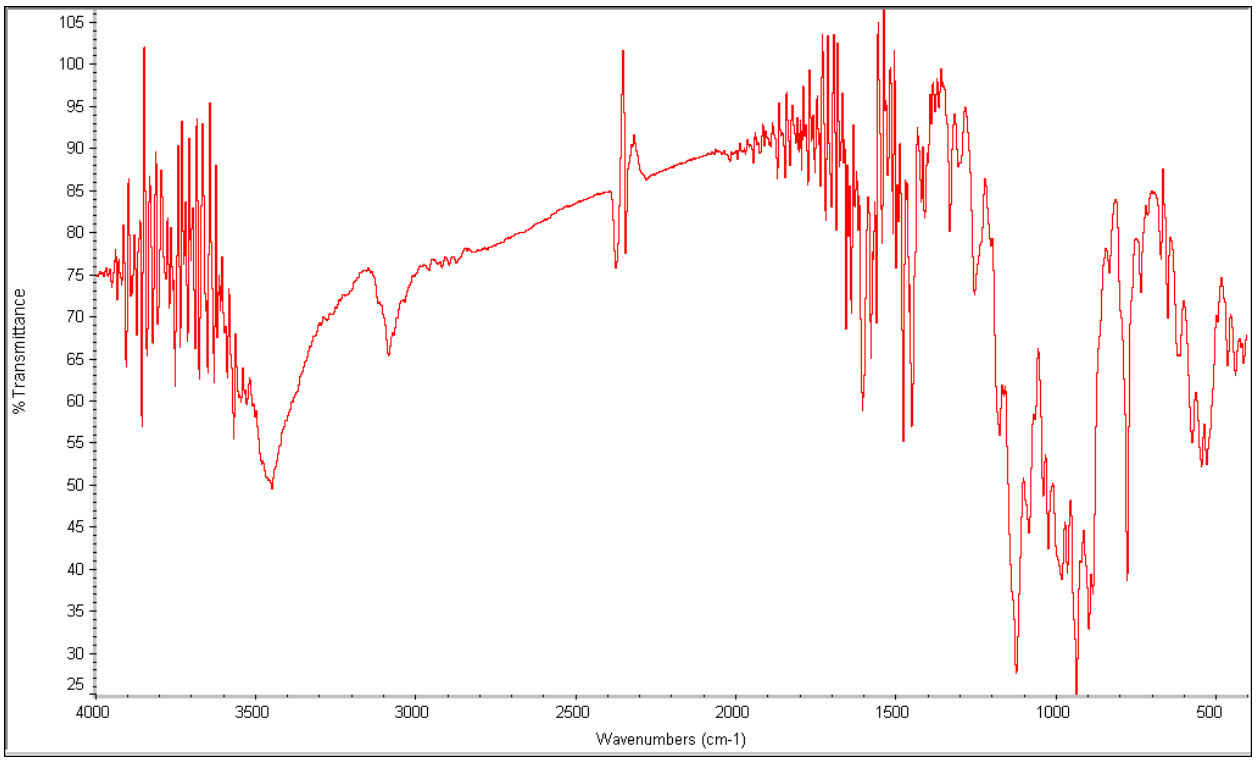


Figure S41. Infrared spectra for **13**·H_2O.

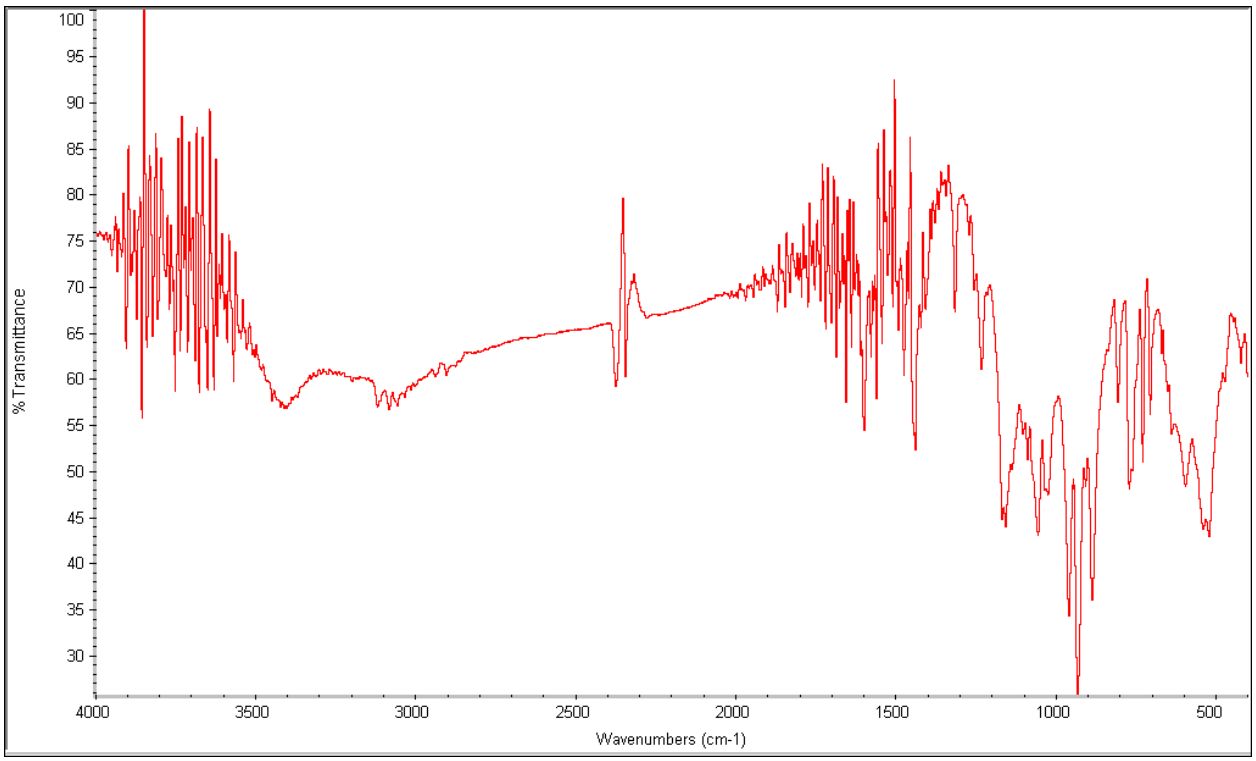


Figure S42. Infrared spectra for **14**.

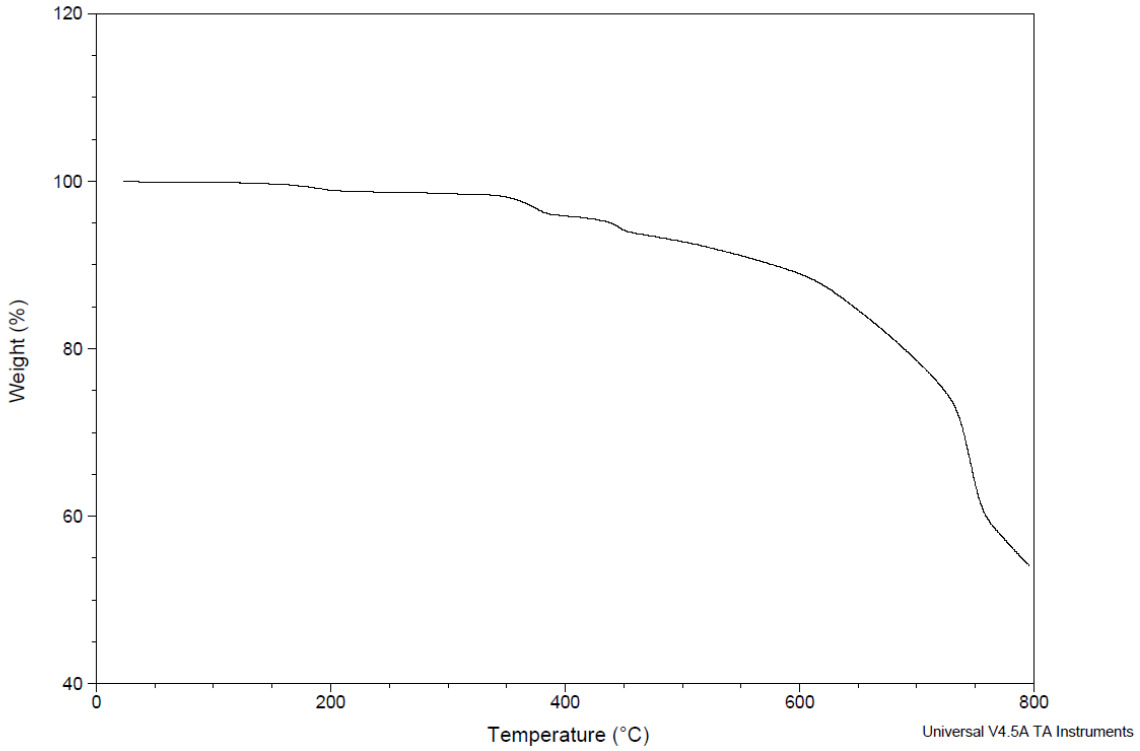


Figure S43. Thermogravimetric analysis profile for compound **1**·H_2O in the 25-800^{o}C range.

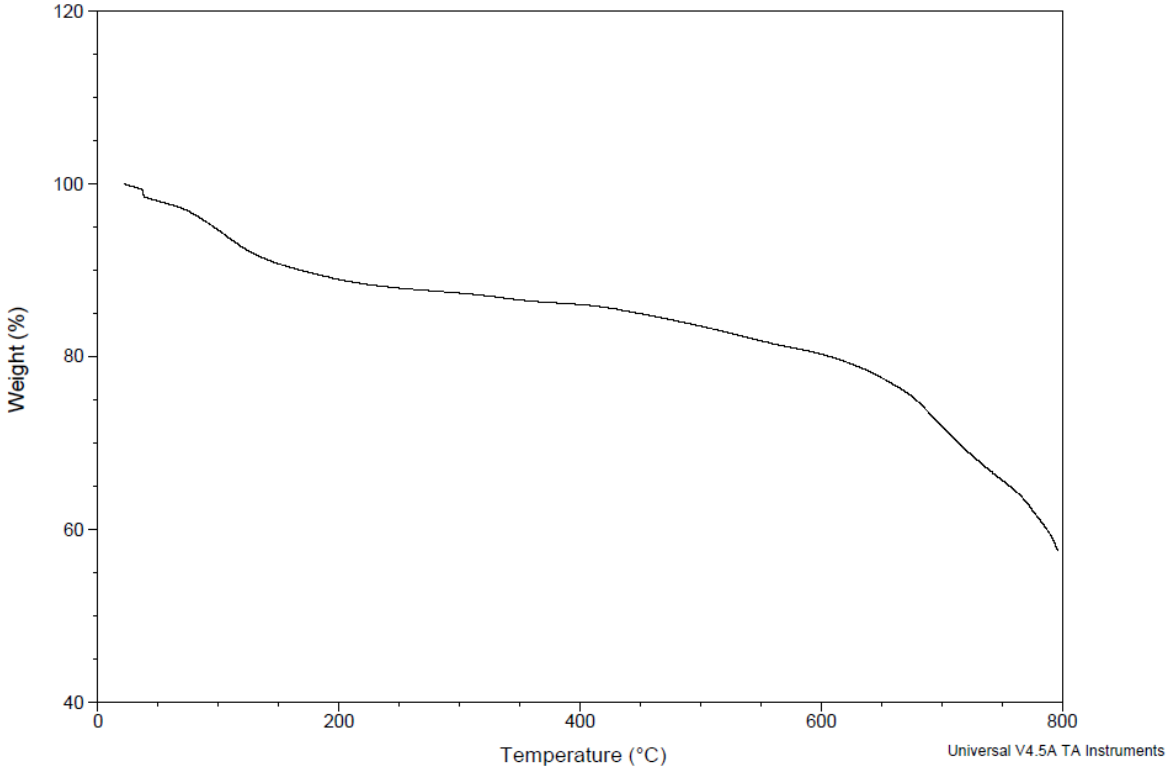


Figure S44. Thermogravimetric analysis profile for compound **2** in the 25-800^{o}C range.

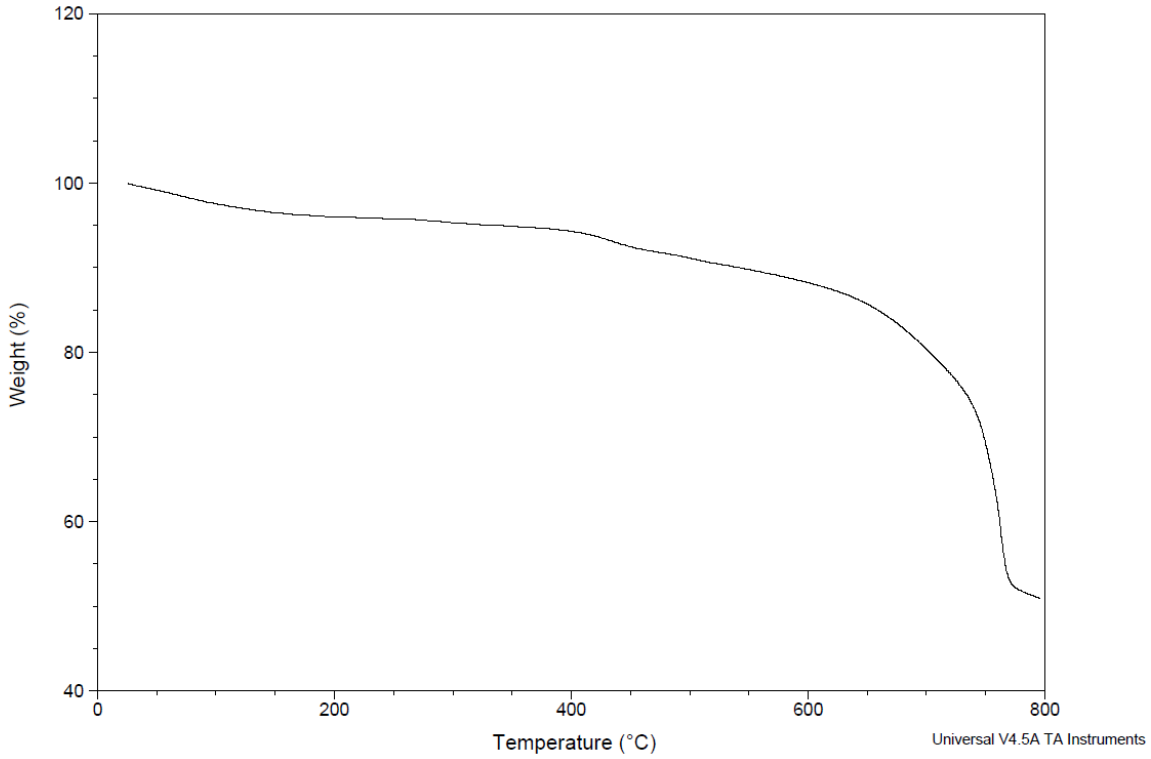


Figure S45. Thermogravimetric analysis profile for compound **3**·H_2O in the 25-800°C range.

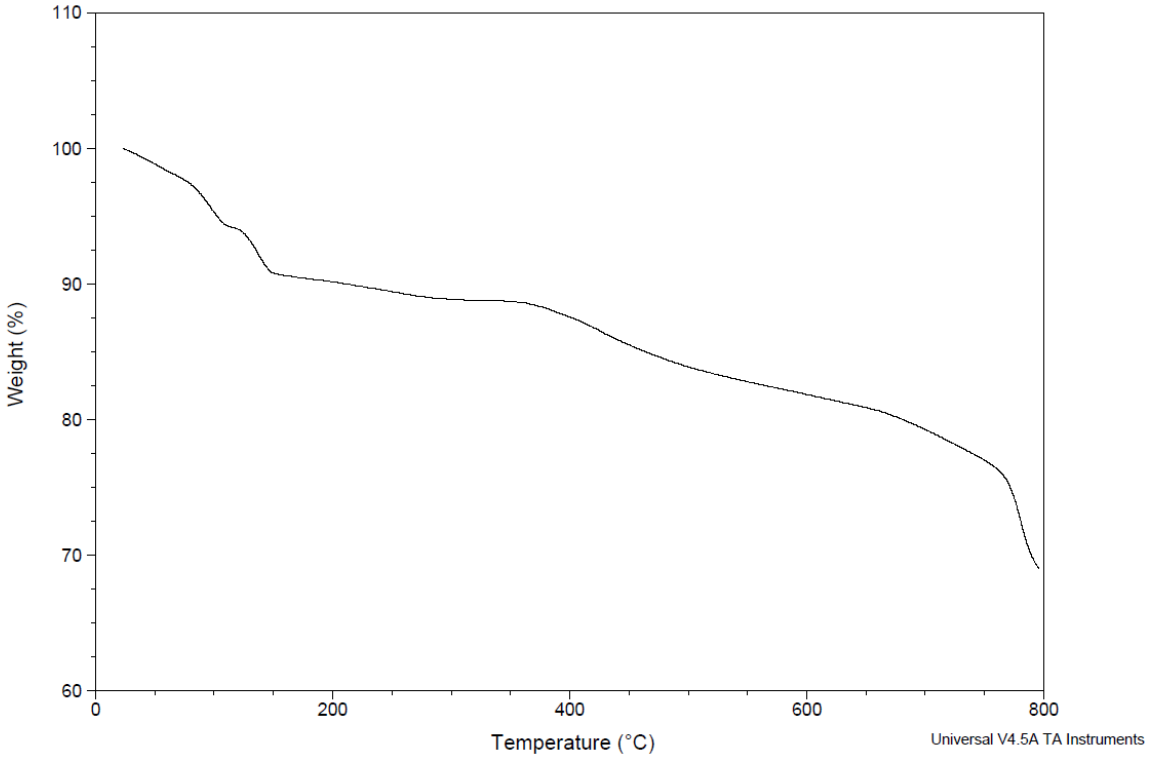


Figure S46. Thermogravimetric analysis profile for compound **4**·6H_2O in the 25-800°C range.

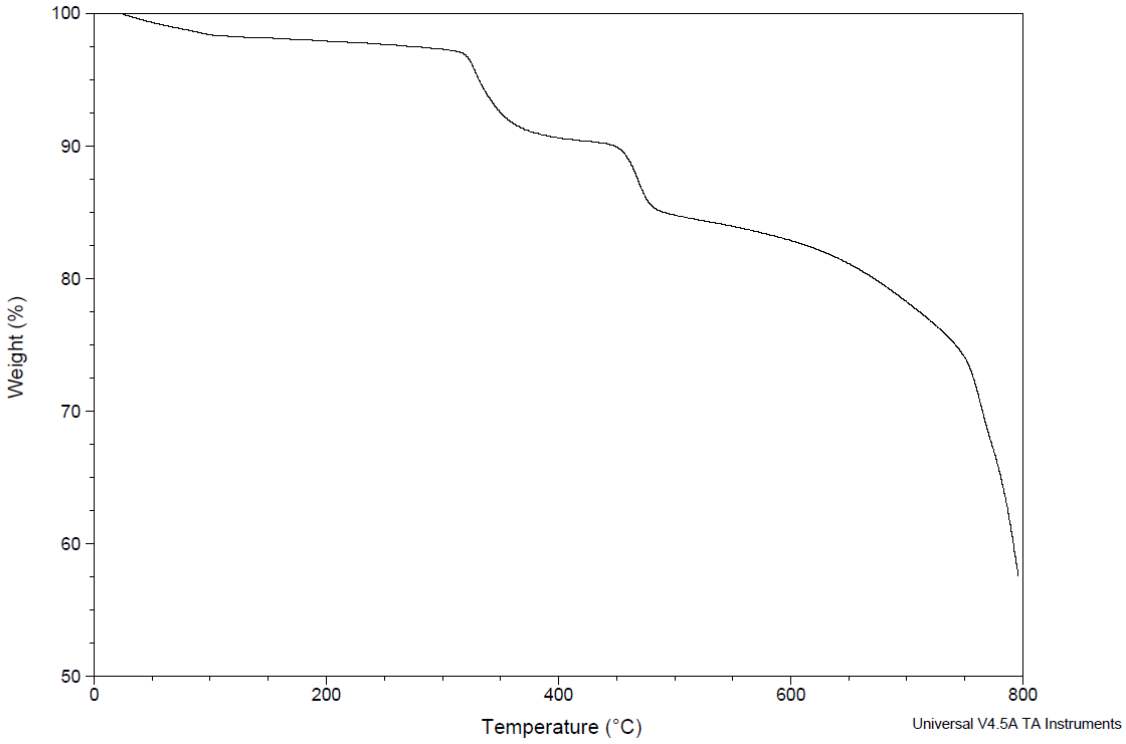


Figure S47. Thermogravimetric analysis profile for compound **5**·6H_2O in the 25-800^oC range.

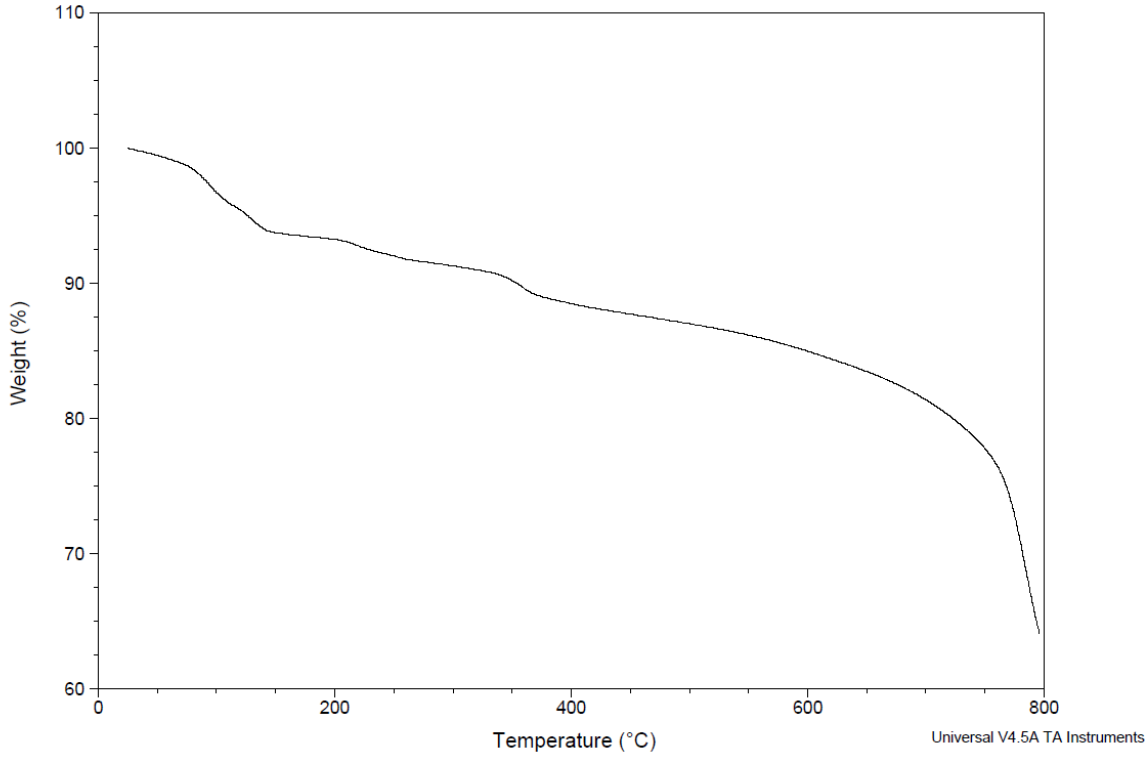


Figure S48. Thermogravimetric analysis profile for compound **6**·4H_2O in the 25-800^oC range.

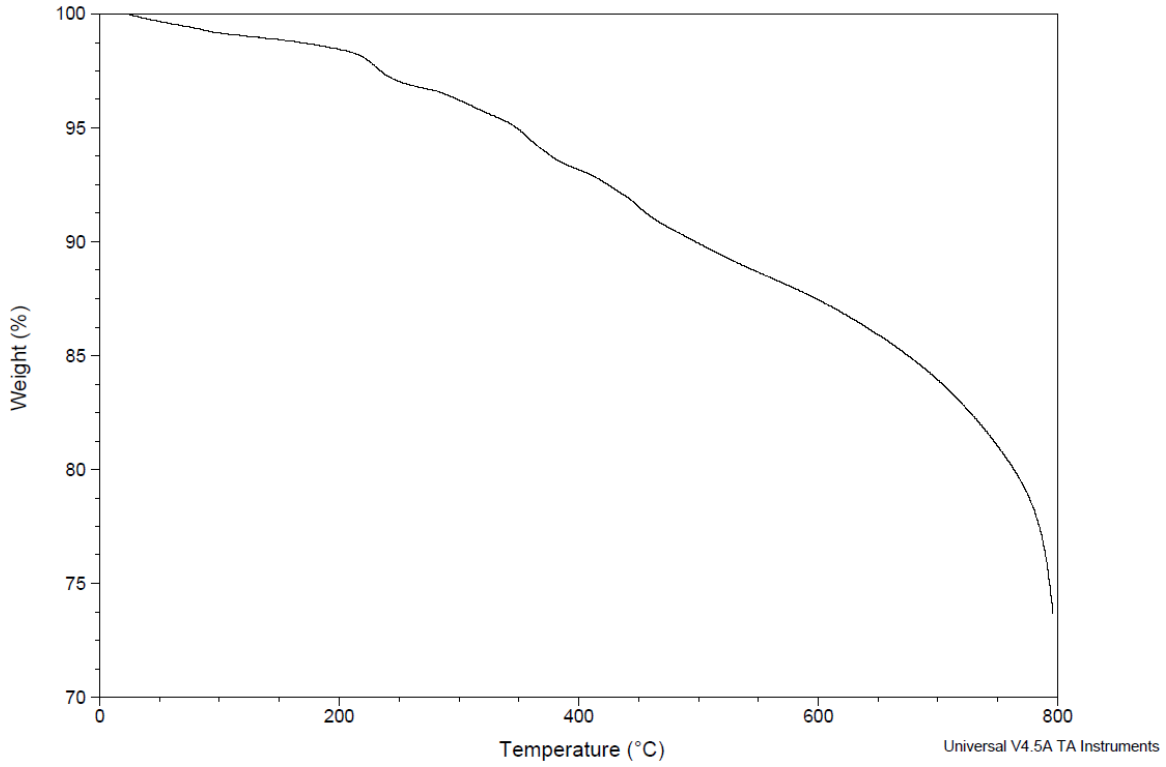


Figure S49. Thermogravimetric analysis profile for compound **7** in the 25-800°C range.

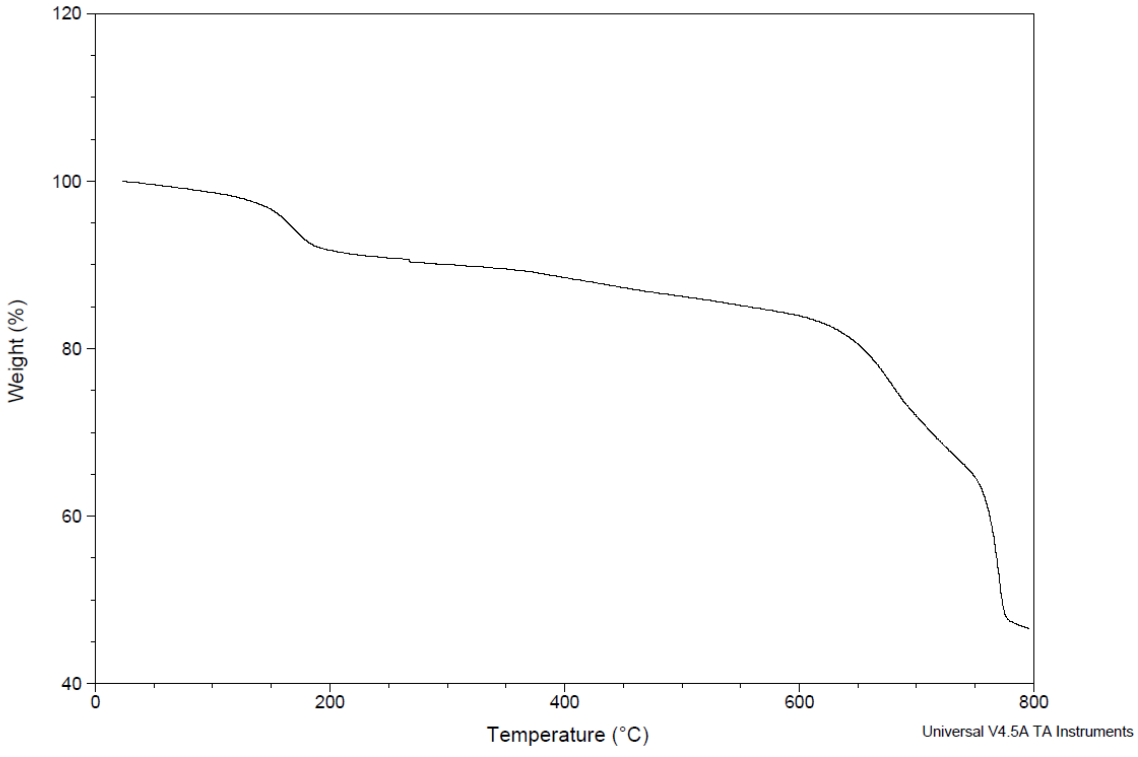


Figure S50. Thermogravimetric analysis profile for compound **8**·4H_2O in the 25-800°C range.

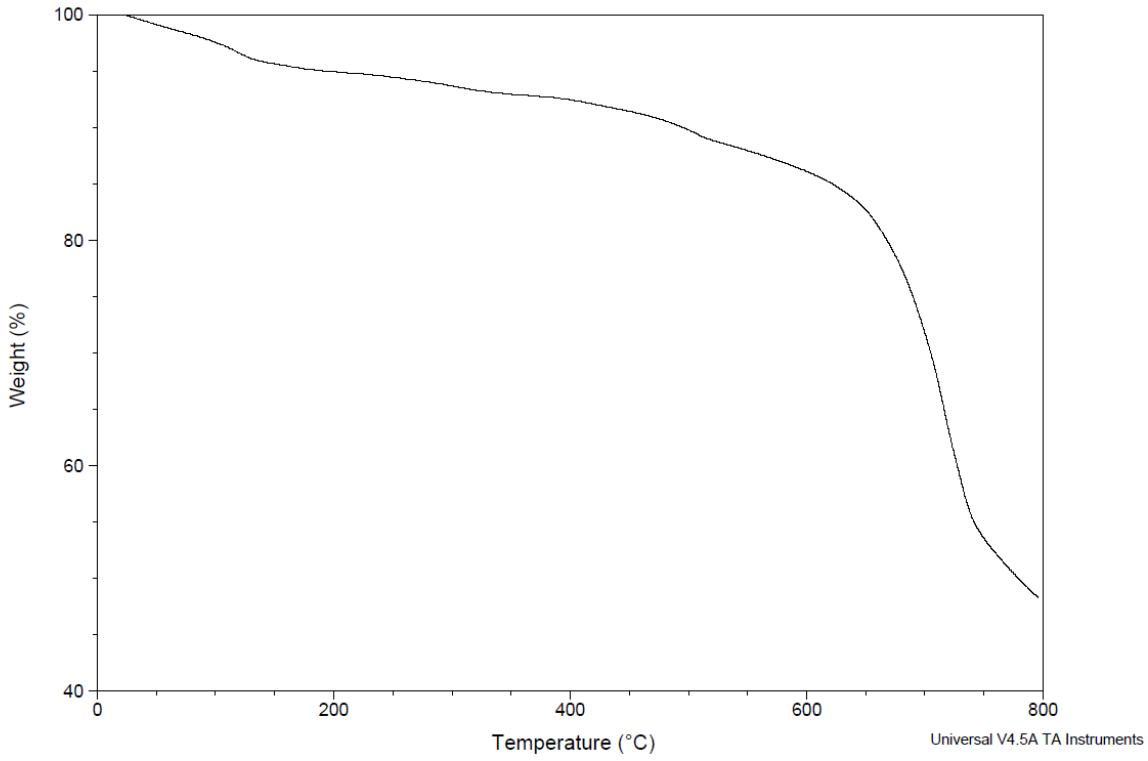


Figure S51. Thermogravimetric analysis profile for compound **9**·6H_2O in the 25-800^oC range.

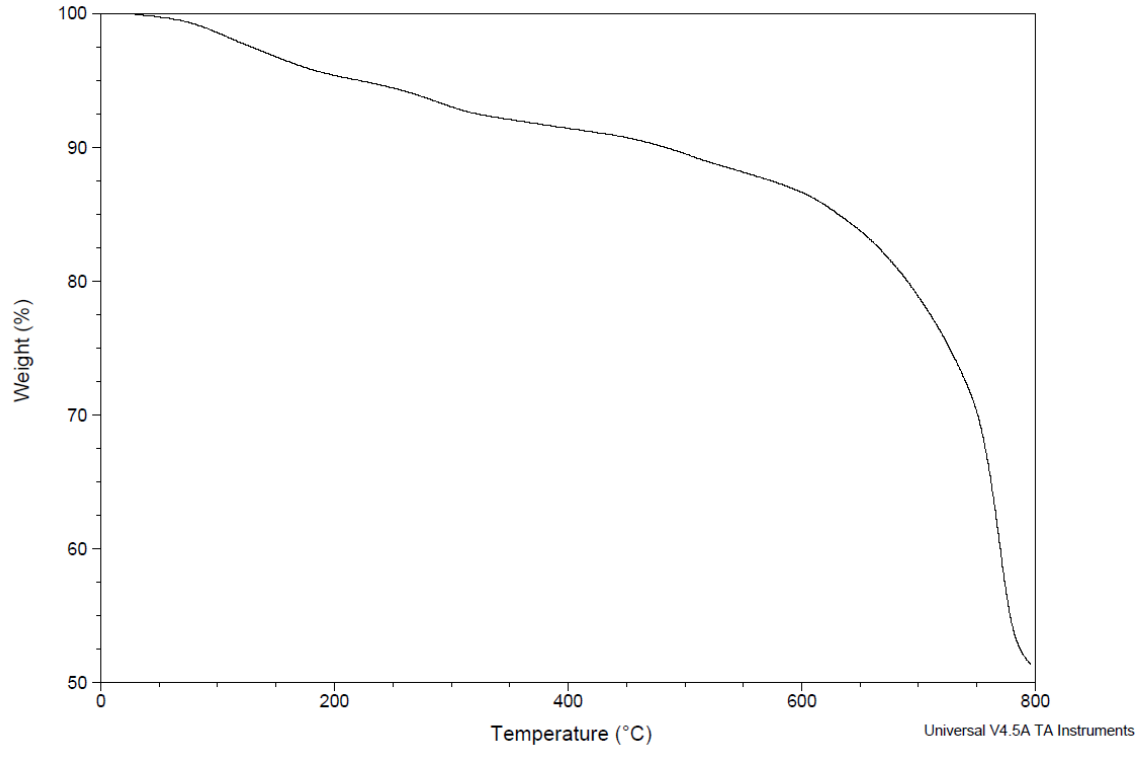


Figure S52. Thermogravimetric analysis profile for compound **10**·4H_2O in the 25-800^oC range.

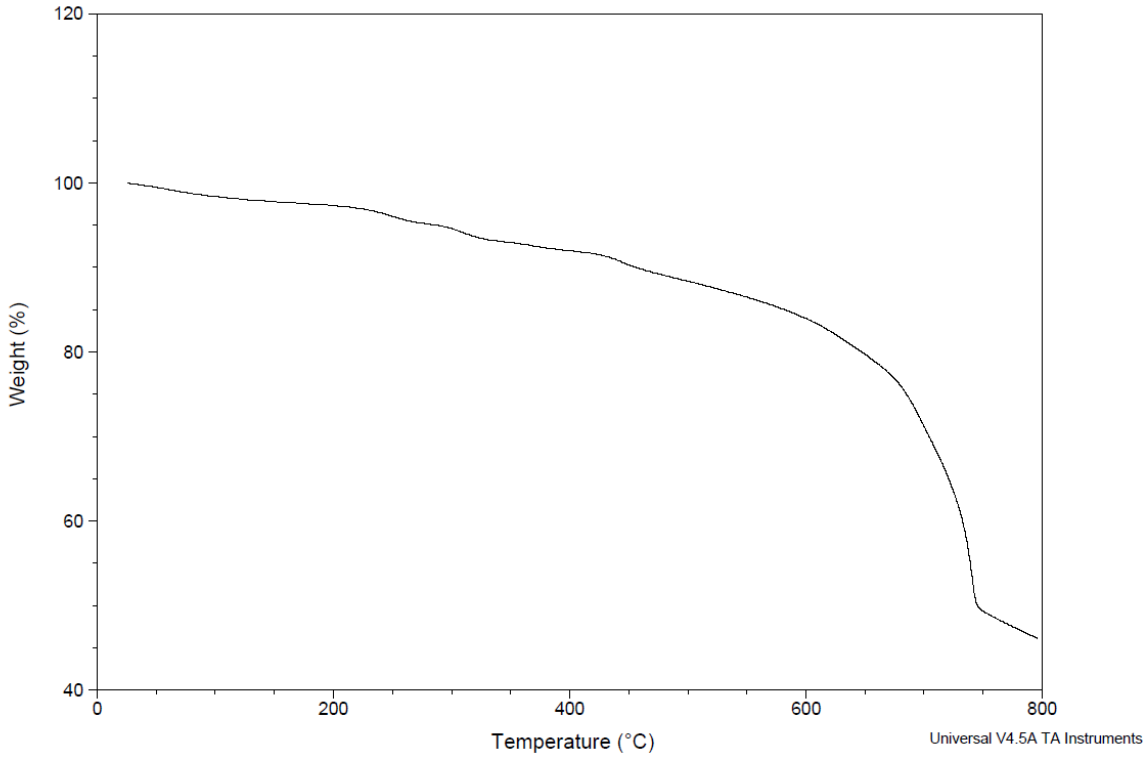


Figure S53. Thermogravimetric analysis profile for compound **11**·2H_2O in the 25-800^oC range.

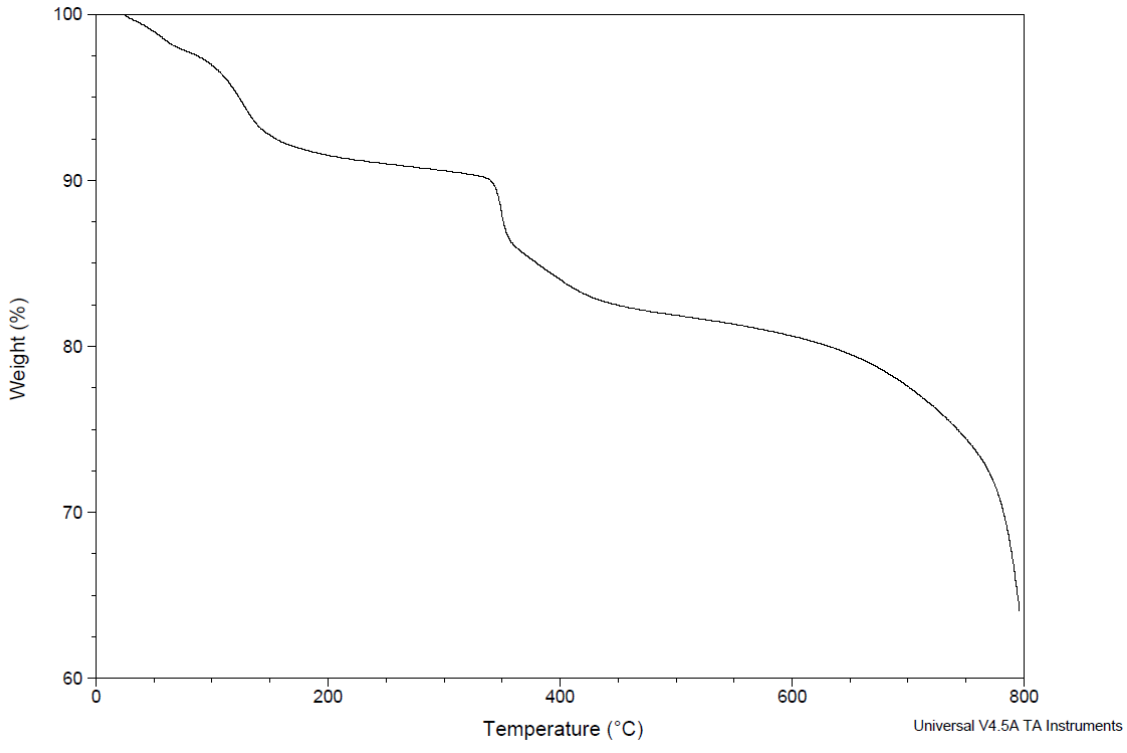


Figure S54. Thermogravimetric analysis profile for compound **12**·H_2O in the 25-800^oC range.

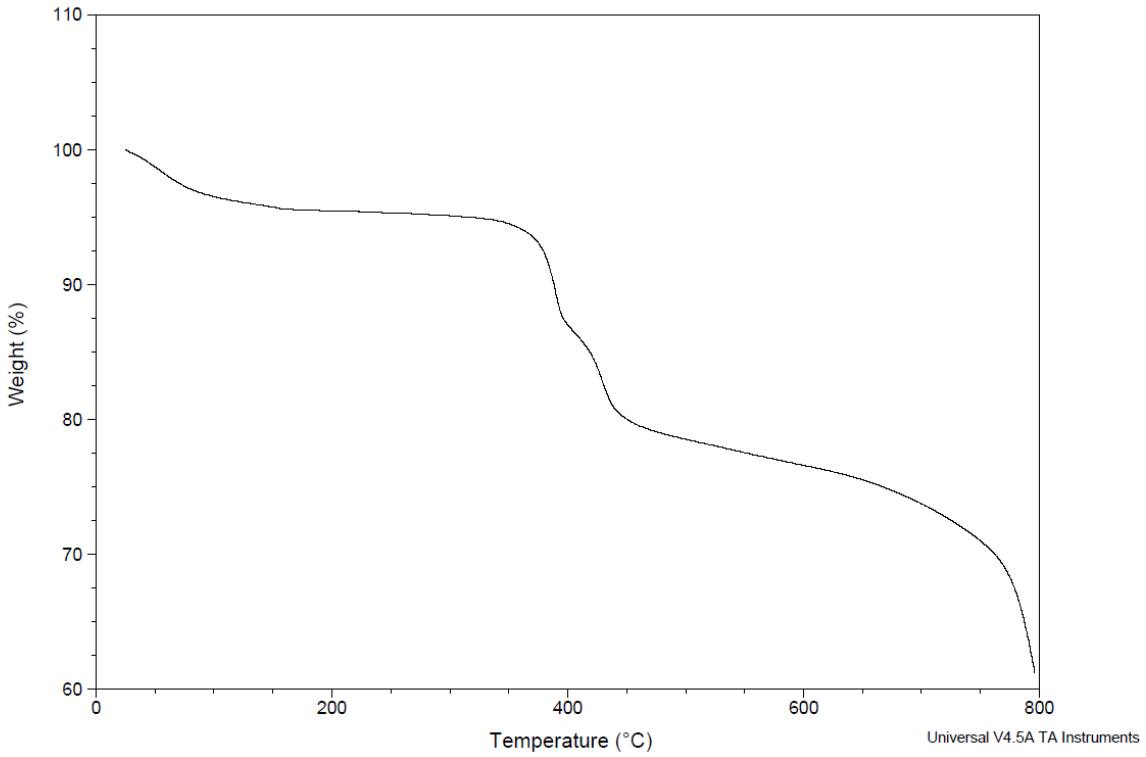


Figure S55. Thermogravimetric analysis profile for compound **13**·H_2O in the 25-800°C range.

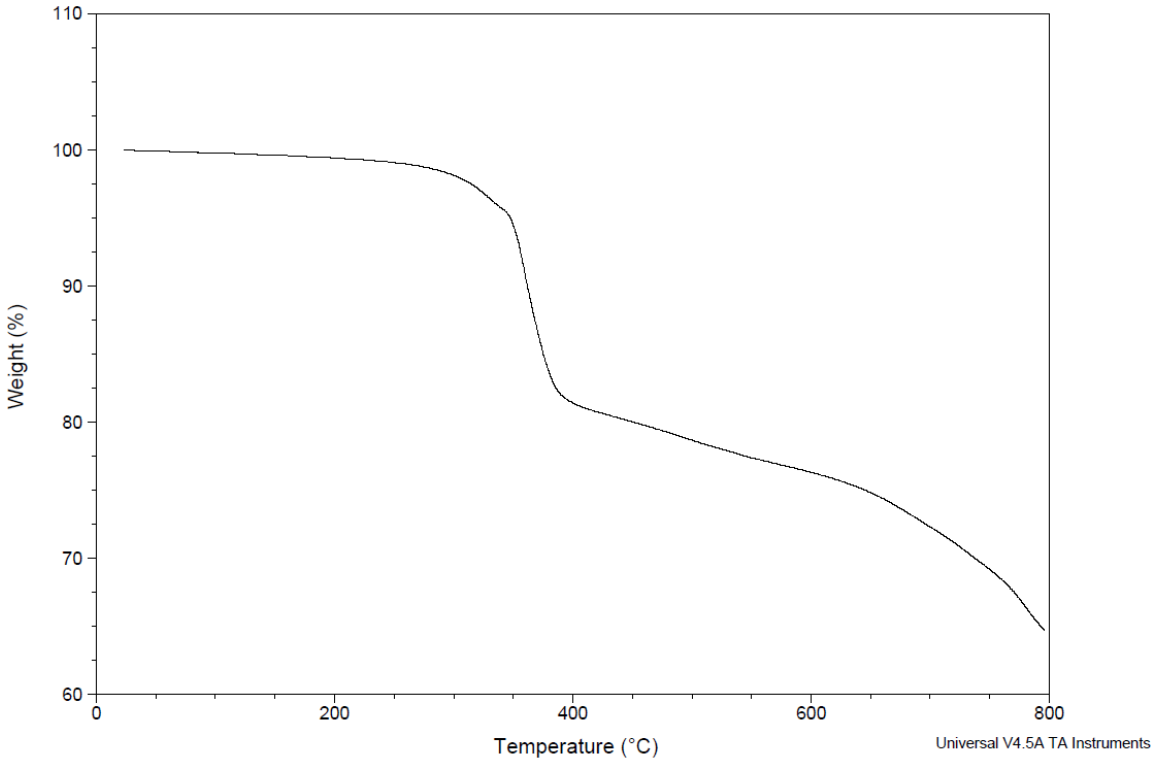
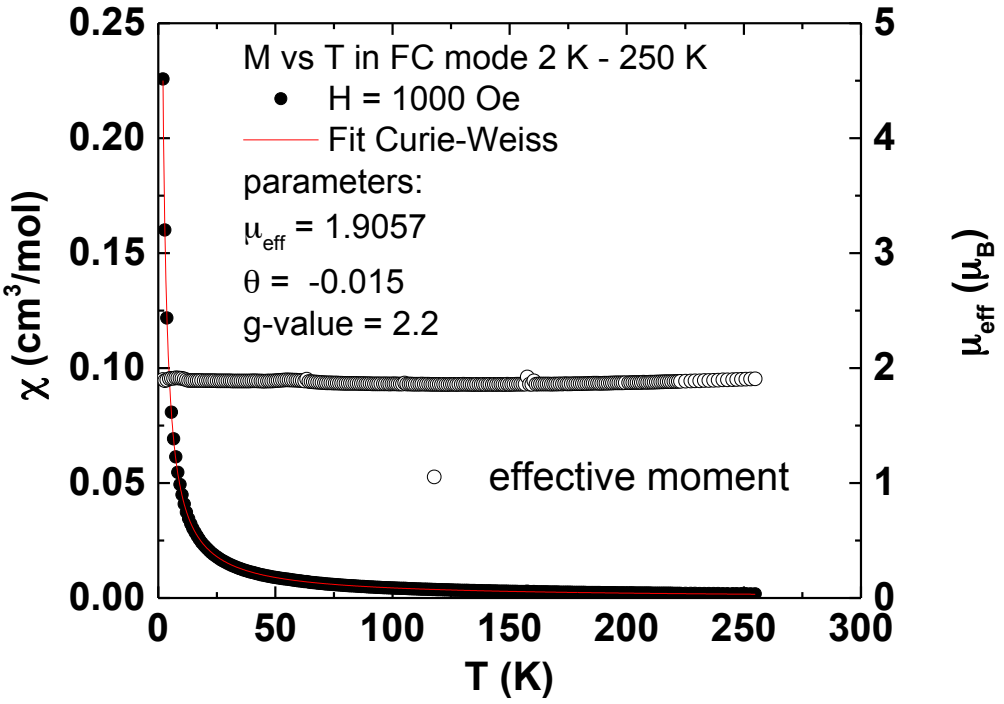
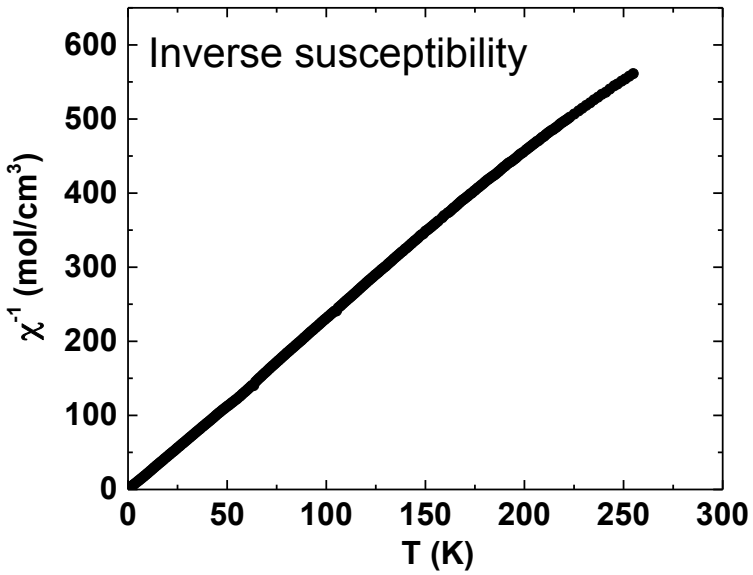


Figure S56. Thermogravimetric analysis profile for compound **14** in the 25-800°C range.

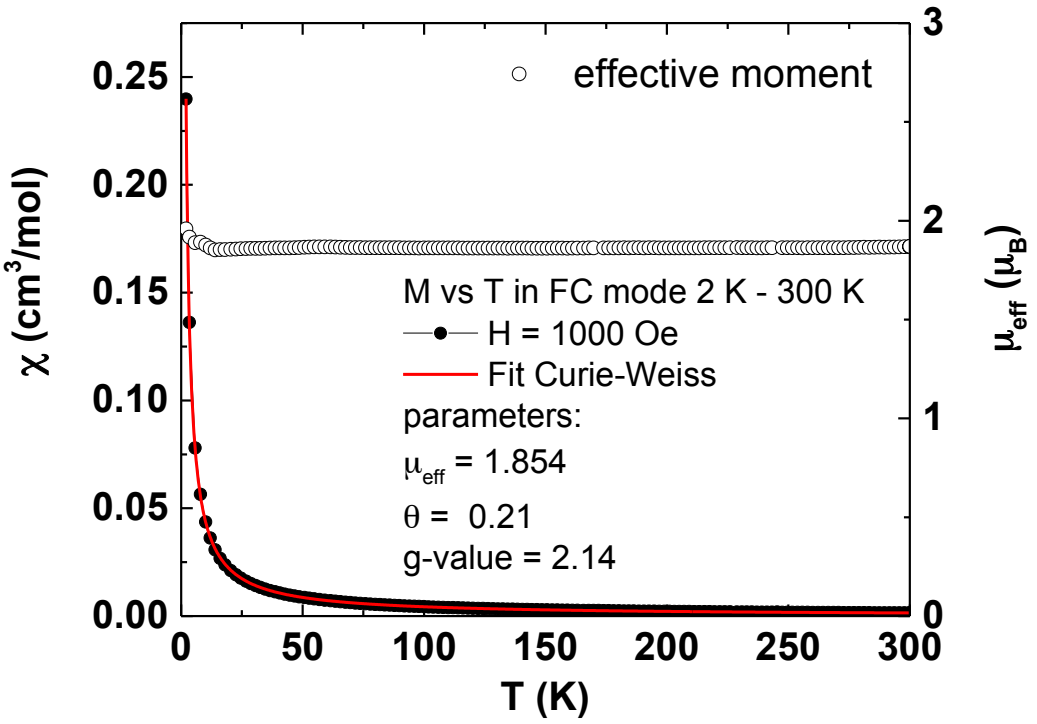


(a)

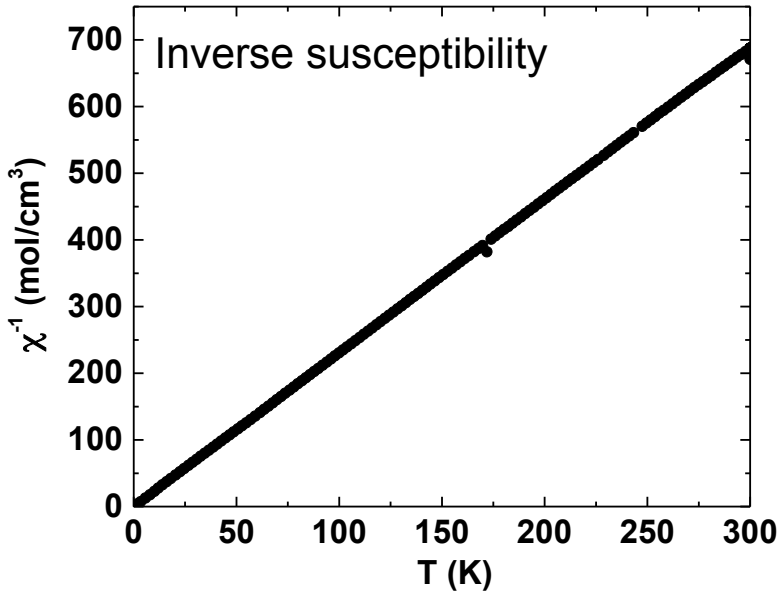


(b)

Figure S57. (a) The temperature dependence of the magnetic susceptibility χ (filled circles) and of the effective magnetic moment μ_{eff} (open circles) of **5**. The red line is the fit to equation 1. (b) A plot of the inverse susceptibility, 1/χ, as a function of temperature.

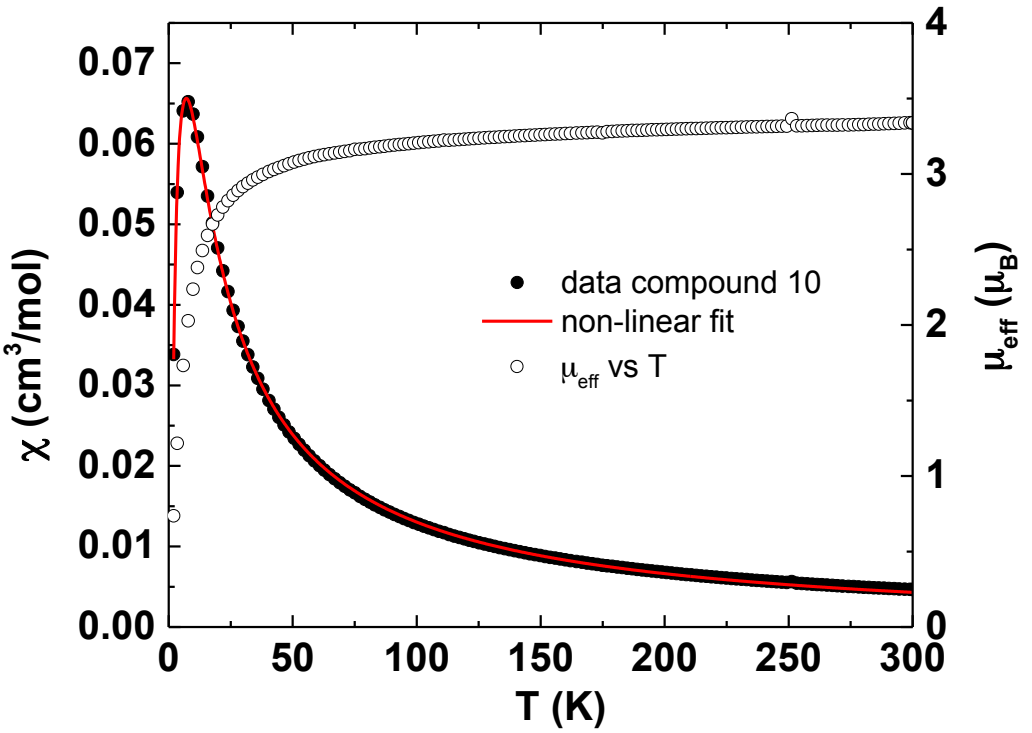


(a)

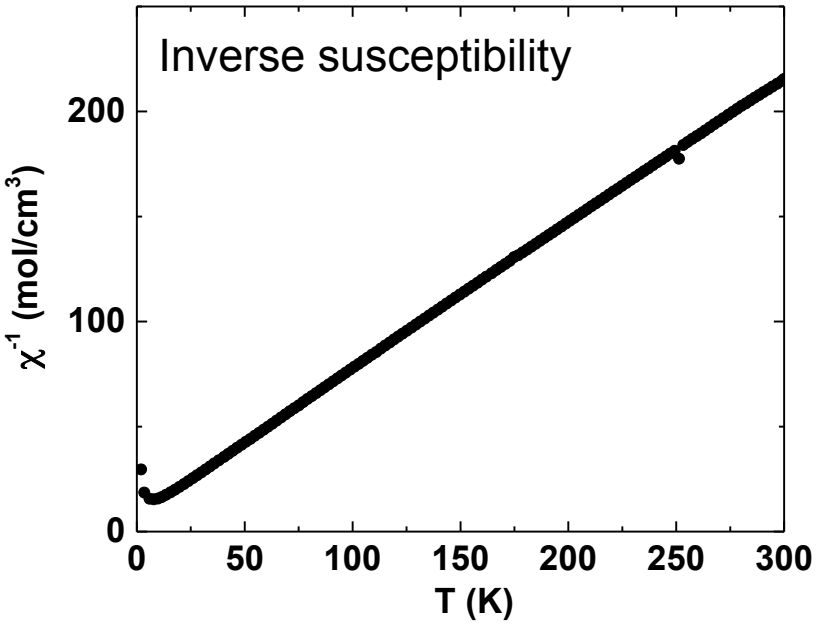


(b)

Figure S58. (a) The temperature dependence of the magnetic susceptibility χ (filled circles) and of the effective magnetic moment μ_{eff} (open circles) of **14**. The red line is the fit to equation 1. (b) A plot of the inverse susceptibility, 1/χ, as a function of temperature.

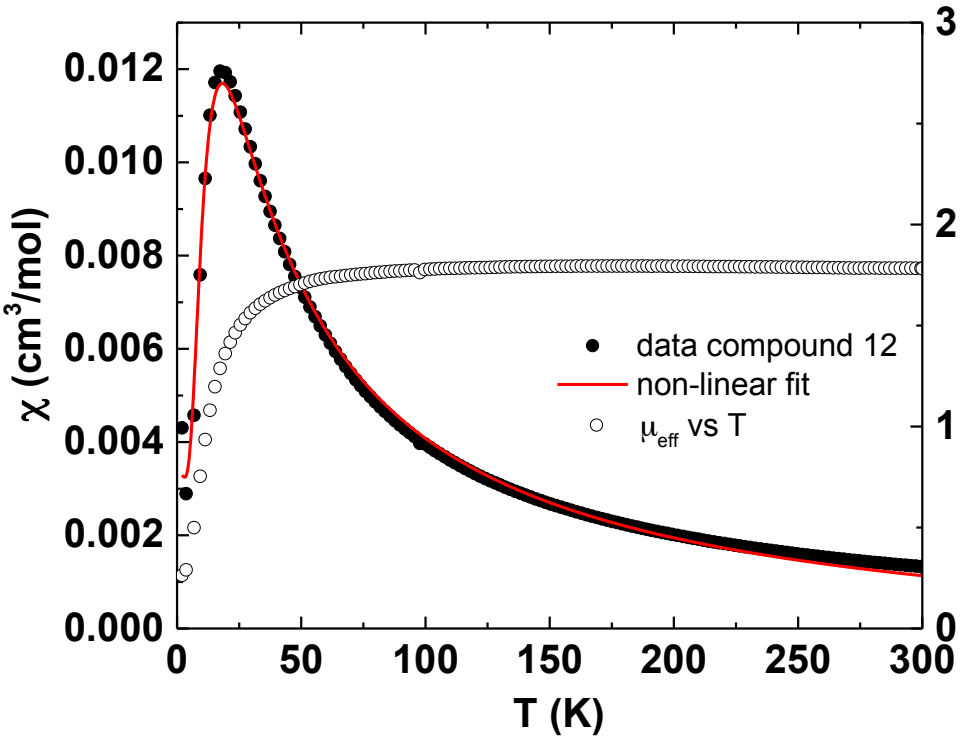


(a)

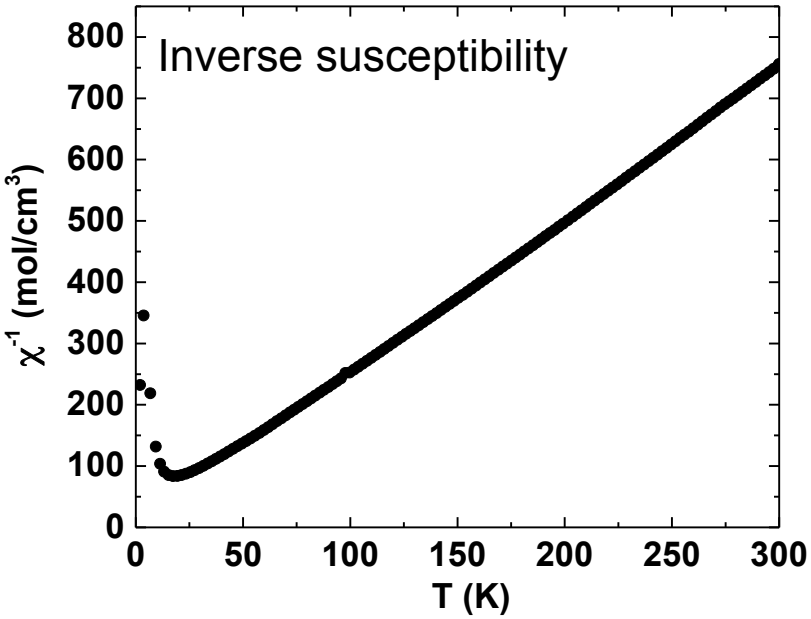


(b)

Figure S59. (a) The temperature dependence of the magnetic susceptibility χ (filled circles) and of the effective magnetic moment μ_{eff} (open circles) of **10**. The red line is the fit to equation 4. (b) A plot of the inverse susceptibility, 1/χ, as a function of temperature.



(a)



(b)

Figure S60. (a) The temperature dependence of the magnetic susceptibility χ (filled circles) and of the effective magnetic moment μ_{eff} (open circles) of **12**. The red line is the fit to equation 6. (b) A plot of the inverse susceptibility, 1/χ, as a function of temperature.

Table S1. Sample and crystal data for $(H_2\text{-}4,4'dpa)_2[Mo_5O_{15}(1,2\text{-}O_3PC_8H_8PO_3)\cdot H_2O$ ($1\cdot H_2O$).

Identification code	$(H_2\text{-}4,4'dpa)_2[Mo_5O_{15}(1,2\text{-}O_3PC_8H_8PO_3)\cdot H_2O$	
Chemical formula	$C_{28}H_{32}Mo_5N_6O_{22}P_2$	
Formula weight	1346.23	
Temperature	90(2) K	
Wavelength	0.71073 Å	
Crystal size	0.186 x 0.250 x 0.585 mm	
Crystal system	Monoclinic	
Space group	P 21/c	
Unit cell dimensions	a = 11.4959(7) Å	α = 90°
	b = 19.4686(12) Å	β = 105.4640(10)°
	c = 18.3104(11) Å	γ = 90°
Volume	3949.7(4) $Å^3$	
Z	4	
Density (calculated)	2.264 g/cm^3	
Absorption coefficient	1.721 mm^{-1}	
F(000)	2632	

Table S2. Data collection and structure refinement for $(H_2\text{-}4,4'dpa)_2[Mo_5O_{15}(1,2\text{-}O_3PC_8H_8PO_3)\cdot H_2O$ $(1\cdot H_2O)$.

Theta range for data collection	1.56 to 25.68°	
Index ranges	-13<=h<=14, -23<=k<=23, -22<=l<=22	
Reflections collected	29843	
Independent reflections	7495 [R(int) = 0.0123]	
Coverage of independent reflections	99.9%	
Max. and min. transmission	0.7400 and 0.4330	
Refinement method	Full-matrix least-squares on F^2	
Refinement program	SHELXL-2013 (Sheldrick, 2013)	
Function minimized	$\Sigma\, w(F_o^2 - F_c^2)^2$	
Data / restraints / parameters	7495 / 8 / 598	
Goodness-of-fit on F^2	0.930	
Δ/σ_{max}	0.003	
Final R indices	7165 data; I>2σ(I)	R1 = 0.0154, wR2 = 0.0394
	all data	R1 = 0.0165, wR2 = 0.0401
Weighting scheme	$w=1/[\sigma^2(F_o^2)+(0.0178P)^2+8.8933P]$ where $P=(F_o^2+2F_c^2)/3$	
Largest diff. peak and hole	1.286 and -0.353 eÅ^{-3}	
R.M.S. deviation from mean	0.066 eÅ^{-3}	

Table S3. Atomic coordinates and equivalent isotropic atomic displacement parameters **(Å^2) for** $(H_2$-4,4'dpa$)_2[Mo_5O_{15}(1,2\text{-}O_3PC_8H_8PO_3)\cdot H_2O$ ($1\cdot H_2O$).

U(eq) is defined as one third of the trace of the orthogonalized U_{ij} tensor.

	x/a	**y/b**	**z/c**	**U(eq)**
Mo1	0.60957(2)	0.67461(2)	0.34370(2)	0.00997(4)
Mo2	0.64208(2)	0.82537(2)	0.43846(2)	0.00893(4)
Mo3	0.96519(2)	0.86512(2)	0.48018(2)	0.00901(4)
Mo4	0.10245(2)	0.71388(2)	0.47612(2)	0.01001(4)
Mo5	0.88814(2)	0.60093(2)	0.38125(2)	0.01049(4)
P1	0.83474(4)	0.70650(3)	0.51705(3)	0.00882(10)
P2	0.84865(4)	0.77302(3)	0.32856(3)	0.00912(10)
O1	0.54275(13)	0.60336(8)	0.36913(9)	0.0168(3)
O2	0.52195(13)	0.69634(8)	0.25593(8)	0.0172(3)
O3	0.55743(12)	0.73885(7)	0.40812(8)	0.0120(3)
O4	0.73120(12)	0.76371(7)	0.35408(8)	0.0099(3)
O5	0.73829(12)	0.63075(7)	0.31114(8)	0.0119(3)
O6	0.78137(12)	0.65473(7)	0.45308(8)	0.0105(3)
O7	0.73765(12)	0.75280(7)	0.53090(8)	0.0107(3)
O8	0.56713(12)	0.85447(7)	0.50233(8)	0.0133(3)
O9	0.57575(13)	0.87331(7)	0.36019(8)	0.0137(3)
O10	0.79251(12)	0.86962(7)	0.46651(8)	0.0108(3)
O11	0.00722(13)	0.87143(8)	0.57679(8)	0.0147(3)

	x/a	y/b	z/c	U(eq)
O12	0.00007(13)	0.94363(8)	0.45147(8)	0.0146(3)
O13	0.90586(12)	0.84077(7)	0.35816(8)	0.0109(3)
O14	0.93493(12)	0.74982(7)	0.49697(8)	0.0102(3)
O15	0.10185(12)	0.81218(7)	0.46379(8)	0.0110(3)
O16	0.17570(13)	0.70690(8)	0.56938(8)	0.0153(3)
O17	0.21059(13)	0.69891(8)	0.42857(9)	0.0172(3)
O18	0.02074(12)	0.62625(7)	0.46217(8)	0.0121(3)
O19	0.93637(12)	0.71270(7)	0.35500(8)	0.0114(3)
O20	0.95343(13)	0.57468(8)	0.31174(8)	0.0158(3)
O21	0.84375(13)	0.52470(8)	0.41231(8)	0.0160(3)
O90	0.1354(2)	0.84473(13)	0.78029(12)	0.0440(5)
N1	0.17093(16)	0.65397(10)	0.83799(10)	0.0147(4)
N2	0.29731(16)	0.58754(9)	0.65910(10)	0.0129(3)
N3	0.50990(17)	0.66055(11)	0.52712(11)	0.0196(4)
N4	0.23395(17)	0.89869(10)	0.67951(10)	0.0177(4)
N5	0.35751(16)	0.93131(9)	0.49125(10)	0.0134(4)
N6	0.28538(16)	0.07672(9)	0.32250(10)	0.0144(4)
C1	0.91177(18)	0.66165(11)	0.60232(11)	0.0126(4)
C2	0.82881(18)	0.62250(11)	0.63861(11)	0.0133(4)
C3	0.8003(2)	0.55493(12)	0.61434(12)	0.0205(5)
C4	0.7203(2)	0.51619(12)	0.64149(13)	0.0246(5)

	x/a	y/b	z/c	U(eq)
C5	0.77801(18)	0.65113(11)	0.69348(11)	0.0123(4)
C6	0.69587(19)	0.61125(12)	0.71946(12)	0.0169(4)
C7	0.6660(2)	0.54523(12)	0.69334(12)	0.0213(5)
C8	0.80858(19)	0.77710(11)	0.22547(11)	0.0142(4)
C9	0.11041(19)	0.60605(11)	0.78948(12)	0.0145(4)
C10	0.15270(18)	0.58451(11)	0.73012(12)	0.0133(4)
C11	0.27405(19)	0.68116(11)	0.83008(12)	0.0162(4)
C12	0.25935(18)	0.61302(11)	0.71902(11)	0.0122(4)
C13	0.32102(19)	0.66196(11)	0.77166(12)	0.0141(4)
C14	0.37309(18)	0.61452(11)	0.61944(12)	0.0130(4)
C15	0.3876(2)	0.57456(12)	0.55839(12)	0.0185(4)
C16	0.43322(19)	0.67769(11)	0.63308(12)	0.0149(4)
C17	0.4559(2)	0.59913(13)	0.51317(13)	0.0222(5)
C18	0.50060(19)	0.69907(12)	0.58563(12)	0.0170(4)
C19	0.28432(19)	0.84742(12)	0.65036(12)	0.0173(4)
C20	0.21965(19)	0.96125(12)	0.64817(12)	0.0163(4)
C21	0.32503(19)	0.85788(11)	0.58765(12)	0.0153(4)
C22	0.25921(18)	0.97553(11)	0.58553(11)	0.0131(4)
C23	0.31226(17)	0.92277(11)	0.55379(11)	0.0123(4)
C24	0.32989(18)	0.98099(10)	0.43639(11)	0.0116(4)
C25	0.22412(18)	0.02037(10)	0.42003(11)	0.0116(4)

	x/a	y/b	z/c	U(eq)
C26	0.41063(18)	0.99002(11)	0.39126(12)	0.0139(4)
C27	0.20538(18)	0.06824(11)	0.36350(11)	0.0135(4)
C28	0.38539(19)	0.03791(11)	0.33491(12)	0.0154(4)

Table S4. Bond lengths (Å) for $(H_2\text{-}4,4'dpa)_2[Mo_5O_{15}(1,2\text{-}O_3PC_8H_8PO_3)\cdot H_2O$ ($1\cdot H_2O$).

Mo1-O2	1.7067(15)	Mo1-O1	1.7094(15)
Mo1-O3	1.9214(14)	Mo1-O5	1.9352(14)
Mo1-O4	2.2041(14)	Mo1-O6	2.4397(14)
Mo2-O9	1.7099(14)	Mo2-O8	1.7229(14)
Mo2-O10	1.8770(14)	Mo2-O3	1.9504(14)
Mo2-O7	2.2531(14)	Mo2-O4	2.3921(13)
Mo3-O12	1.6989(15)	Mo3-O11	1.7093(14)
Mo3-O10	1.9354(14)	Mo3-O15	1.9677(14)
Mo3-O13	2.2066(14)	Mo3-O14	2.3046(14)
Mo4-O16	1.6961(15)	Mo4-O17	1.7221(14)
Mo4-O15	1.9269(14)	Mo4-O18	1.9313(14)
Mo4-O14	2.1763(13)	Mo4-O19	2.5113(14)
Mo5-O21	1.7149(15)	Mo5-O20	1.7186(14)
Mo5-O18	1.8862(14)	Mo5-O5	1.9426(14)
Mo5-O6	2.2792(13)	Mo5-O19	2.3273(14)
P1-O7	1.5078(14)	P1-O6	1.5430(14)
P1-O14	1.5488(14)	P1-C1	1.802(2)
P2-O13	1.5087(15)	P2-O19	1.5394(15)
P2-O4	1.5526(14)	P2-C8	1.821(2)
O90-H90A	0.870(18)	O90-H90B	0.919(18)
N1-C11	1.341(3)	N1-C9	1.347(3)

N1-H1'	0.863(17)	N2-C12	1.377(3)
N2-C14	1.378(3)	N2-H2'	0.851(17)
N3-C18	1.335(3)	N3-C17	1.340(3)
N3-H3'	0.859(17)	N4-C19	1.335(3)
N4-C20	1.338(3)	N4-H4'	0.859(17)
N5-C24	1.369(3)	N5-C23	1.388(3)
N5-H5'	0.851(17)	N6-C28	1.344(3)
N6-C27	1.343(3)	N6-H6'	0.872(17)
C1-C2	1.507(3)	C1-H1A	0.99
C1-H1B	0.99	C2-C3	1.399(3)
C2-C5	1.405(3)	C3-C4	1.381(3)
C3-H3	0.95	C4-C7	1.388(3)
C4-H4	0.95	C5-C6	1.400(3)
C5-C8	1.520(3)	C6-C7	1.383(3)
C6-H6	0.95	C7-H7	0.95
C8-C5	1.520(3)	C8-H8A	0.99
C8-H8B	0.99	C9-C10	1.370(3)
C9-H9	0.95	C10-C12	1.409(3)
C10-H10	0.95	C11-C13	1.372(3)
C11-H11	0.95	C12-C13	1.405(3)
C13-H13	0.95	C14-C16	1.400(3)
C14-C15	1.408(3)	C15-C17	1.370(3)

C15-H15	0.95	C16-C18	1.374(3)
C16-H16	0.95	C17-H17	0.95
C18-H18	0.95	C19-C21	1.366(3)
C19-H19	0.95	C20-C22	1.370(3)
C20-H20	0.95	C21-C23	1.398(3)
C21-H21	0.95	C22-C23	1.398(3)
C22-H22	0.95	C24-C25	1.401(3)
C24-C26	1.409(3)	C25-C27	1.367(3)
C25-H25	0.95	C26-C28	1.363(3)
C26-H26	0.95	C27-H27	0.95
C28-H28	0.95		

Table S5. Bond angles (°) for $(H_2\text{-}4,4'dpa)_2[Mo_5O_{15}(1,2\text{-}O_3PC_8H_8PO_3)\cdot H_2O$ $(1\cdot H_2O)$.

O2-Mo1-O1	105.21(7)	O2-Mo1-O3	102.38(7)
O1-Mo1-O3	97.16(6)	O2-Mo1-O5	97.30(7)
O1-Mo1-O5	99.32(7)	O3-Mo1-O5	149.96(6)
O2-Mo1-O4	95.03(6)	O1-Mo1-O4	159.38(6)
O3-Mo1-O4	74.30(5)	O5-Mo1-O4	81.62(5)
O2-Mo1-O6	163.26(6)	O1-Mo1-O6	88.39(6)
O3-Mo1-O6	85.18(5)	O5-Mo1-O6	70.42(5)
O4-Mo1-O6	72.42(5)	O9-Mo2-O8	101.39(7)
O9-Mo2-O10	98.69(7)	O8-Mo2-O10	104.74(6)
O9-Mo2-O3	98.95(7)	O8-Mo2-O3	100.11(6)
O10-Mo2-O3	145.85(6)	O9-Mo2-O7	172.34(6)
O8-Mo2-O7	85.89(6)	O10-Mo2-O7	81.55(6)
O3-Mo2-O7	77.22(6)	O9-Mo2-O4	84.53(6)
O8-Mo2-O4	168.90(6)	O10-Mo2-O4	83.40(5)
O3-Mo2-O4	69.50(5)	O7-Mo2-O4	87.91(5)
O12-Mo3-O11	103.33(7)	O12-Mo3-O10	103.75(6)
O11-Mo3-O10	97.34(6)	O12-Mo3-O15	99.22(6)
O11-Mo3-O15	100.29(6)	O10-Mo3-O15	146.83(6)
O12-Mo3-O13	85.10(6)	O11-Mo3-O13	171.51(6)
O10-Mo3-O13	81.33(5)	O15-Mo3-O13	77.11(5)
O12-Mo3-O14	166.94(6)	O11-Mo3-O14	86.74(6)

O10-Mo3-O14	82.84(5)	O15-Mo3-O14	70.44(5)
O13-Mo3-O14	84.78(5)	O16-Mo4-O17	105.31(7)
O16-Mo4-O15	100.54(7)	O17-Mo4-O15	95.13(7)
O16-Mo4-O18	98.96(7)	O17-Mo4-O18	100.07(7)
O15-Mo4-O18	151.09(6)	O16-Mo4-O14	94.25(6)
O17-Mo4-O14	159.22(6)	O15-Mo4-O14	74.07(5)
O18-Mo4-O14	83.41(6)	O16-Mo4-O19	160.89(6)
O17-Mo4-O19	91.91(6)	O15-Mo4-O19	85.83(5)
O18-Mo4-O19	69.31(5)	O14-Mo4-O19	69.99(5)
O21-Mo5-O20	102.45(7)	O21-Mo5-O18	102.43(7)
O20-Mo5-O18	103.79(7)	O21-Mo5-O5	100.84(7)
O20-Mo5-O5	94.87(6)	O18-Mo5-O5	146.05(6)
O21-Mo5-O6	87.29(6)	O20-Mo5-O6	166.59(6)
O18-Mo5-O6	82.72(5)	O5-Mo5-O6	74.03(5)
O21-Mo5-O19	170.41(6)	O20-Mo5-O19	87.13(6)
O18-Mo5-O19	74.38(5)	O5-Mo5-O19	78.58(5)
O6-Mo5-O19	83.34(5)	O7-P1-O6	110.84(8)
O7-P1-O14	110.19(8)	O6-P1-O14	110.23(8)
O7-P1-C1	111.02(9)	O6-P1-C1	110.20(9)
O14-P1-C1	104.17(9)	O13-P2-O19	111.57(8)
O13-P2-O4	108.95(8)	O19-P2-O4	111.45(8)
O13-P2-C8	107.44(9)	O19-P2-C8	108.82(9)

O4-P2-C8	108.49(9)	Mo1-O3-Mo2	121.56(7)
P2-O4-Mo1	130.42(8)	P2-O4-Mo2	133.82(8)
Mo1-O4-Mo2	94.56(5)	Mo1-O5-Mo5	122.99(7)
P1-O6-Mo5	124.89(8)	P1-O6-Mo1	126.19(8)
Mo5-O6-Mo1	92.40(5)	P1-O7-Mo2	118.88(8)
Mo2-O10-Mo3	148.54(8)	P2-O13-Mo3	122.61(8)
P1-O14-Mo4	127.99(8)	P1-O14-Mo3	135.25(8)
Mo4-O14-Mo3	96.76(5)	Mo4-O15-Mo3	118.71(7)
Mo5-O18-Mo4	126.69(7)	P2-O19-Mo5	127.30(8)
P2-O19-Mo4	123.42(8)	Mo5-O19-Mo4	89.60(5)
H90A-O90-H90B	86.(3)	C11-N1-C9	121.22(18)
C11-N1-H1'	117.(2)	C9-N1-H1'	122.(2)
C12-N2-C14	131.43(19)	C12-N2-H2'	112.6(17)
C14-N2-H2'	114.5(17)	C18-N3-C17	121.46(19)
C18-N3-H3'	117.8(19)	C17-N3-H3'	120.3(19)
C19-N4-C20	121.76(19)	C19-N4-H4'	118.(2)
C20-N4-H4'	120.(2)	C24-N5-C23	128.69(18)
C24-N5-H5'	116.1(19)	C23-N5-H5'	114.5(19)
C28-N6-C27	121.26(19)	C28-N6-H6'	117.1(18)
C27-N6-H6'	121.0(18)	C2-C1-P1	113.87(14)
C2-C1-H1A	108.8	P1-C1-H1A	108.8
C2-C1-H1B	108.8	P1-C1-H1B	108.8

H1A-C1-H1B	107.7	C3-C2-C5	119.6(2)
C3-C2-C1	117.23(19)	C5-C2-C1	123.19(19)
C4-C3-C2	121.4(2)	C4-C3-H3	119.3
C2-C3-H3	119.3	C3-C4-C7	119.2(2)
C3-C4-H4	120.4	C7-C4-H4	120.4
C6-C5-C2	118.0(2)	C6-C5-C8	119.11(18)
C2-C5-C8	122.88(18)	C7-C6-C5	121.7(2)
C7-C6-H6	119.1	C5-C6-H6	119.1
C6-C7-C4	120.0(2)	C6-C7-H7	120.0
C4-C7-H7	120.0	C5-C8-P2	114.09(14)
C5-C8-H8A	108.7	P2-C8-H8A	108.7
C5-C8-H8B	108.7	P2-C8-H8B	108.7
H8A-C8-H8B	107.6	N1-C9-C10	120.35(19)
N1-C9-H9	119.8	C10-C9-H9	119.8
C9-C10-C12	120.0(2)	C9-C10-H10	120.0
C12-C10-H10	120.0	N1-C11-C13	121.3(2)
N1-C11-H11	119.3	C13-C11-H11	119.3
N2-C12-C13	125.57(19)	N2-C12-C10	116.48(19)
C13-C12-C10	117.87(19)	C11-C13-C12	119.2(2)
C11-C13-H13	120.4	C12-C13-H13	120.4
N2-C14-C16	126.30(19)	N2-C14-C15	115.66(19)
C16-C14-C15	118.01(19)	C17-C15-C14	119.7(2)

C17-C15-H15	120.2	C14-C15-H15	120.2
C18-C16-C14	119.2(2)	C18-C16-H16	120.4
C14-C16-H16	120.4	N3-C17-C15	120.5(2)
N3-C17-H17	119.8	C15-C17-H17	119.8
N3-C18-C16	121.1(2)	N3-C18-H18	119.4
C16-C18-H18	119.4	N4-C19-C21	120.5(2)
N4-C19-H19	119.8	C21-C19-H19	119.8
N4-C20-C22	120.9(2)	N4-C20-H20	119.6
C22-C20-H20	119.6	C19-C21-C23	119.3(2)
C19-C21-H21	120.4	C23-C21-H21	120.4
C20-C22-C23	118.6(2)	C20-C22-H22	120.7
C23-C22-H22	120.7	N5-C23-C21	117.13(19)
N5-C23-C22	123.78(19)	C21-C23-C22	119.06(19)
N5-C24-C25	124.17(19)	N5-C24-C26	117.28(18)
C25-C24-C26	118.46(19)	C27-C25-C24	119.19(19)
C27-C25-H25	120.4	C24-C25-H25	120.4
C28-C26-C24	119.26(19)	C28-C26-H26	120.4
C24-C26-H26	120.4	N6-C27-C25	120.92(19)
N6-C27-H27	119.5	C25-C27-H27	119.5
N6-C28-C26	120.83(19)	N6-C28-H28	119.6
C26-C28-H28	119.6		

Table S6. Torsion angles (°) for $(H_2\text{-}4,4'dpa)_2[Mo_5O_{15}(1,2\text{-}O_3PC_8H_8PO_3)\cdot H_2O$ $(1\cdot H_2O)$.

O13-P2-O4-Mo1	-172.75(9)	O19-P2-O4-Mo1	-49.20(12)
C8-P2-O4-Mo1	70.58(12)	O13-P2-O4-Mo2	-8.53(13)
O19-P2-O4-Mo2	115.02(11)	C8-P2-O4-Mo2	-125.20(11)
O7-P1-O6-Mo5	156.88(8)	O14-P1-O6-Mo5	34.60(11)
C1-P1-O6-Mo5	-79.82(11)	O7-P1-O6-Mo1	31.89(11)
O14-P1-O6-Mo1	-90.38(10)	C1-P1-O6-Mo1	155.19(9)
O6-P1-O7-Mo2	-64.83(10)	O14-P1-O7-Mo2	57.47(10)
C1-P1-O7-Mo2	172.35(9)	O9-Mo2-O10-Mo3	-114.87(15)
O8-Mo2-O10-Mo3	140.85(15)	O3-Mo2-O10-Mo3	5.5(2)
O7-Mo2-O10-Mo3	57.40(15)	O4-Mo2-O10-Mo3	-31.46(15)
O19-P2-O13-Mo3	-63.26(11)	O4-P2-O13-Mo3	60.21(11)
C8-P2-O13-Mo3	177.54(9)	O7-P1-O14-Mo4	176.89(9)
O6-P1-O14-Mo4	-60.45(12)	C1-P1-O14-Mo4	57.74(12)
O7-P1-O14-Mo3	-2.39(14)	O6-P1-O14-Mo3	120.27(11)
C1-P1-O14-Mo3	-121.53(12)	O21-Mo5-O18-Mo4	-169.18(9)
O20-Mo5-O18-Mo4	84.48(10)	O5-Mo5-O18-Mo4	-36.93(16)
O6-Mo5-O18-Mo4	-83.59(9)	O19-Mo5-O18-Mo4	1.50(8)
O13-P2-O19-Mo5	147.75(9)	O4-P2-O19-Mo5	25.71(12)
C8-P2-O19-Mo5	-93.88(11)	O13-P2-O19-Mo4	28.26(11)
O4-P2-O19-Mo4	-93.78(9)	C8-P2-O19-Mo4	146.63(9)
O7-P1-C1-C2	54.06(17)	O6-P1-C1-C2	-69.14(16)

O14-P1-C1-C2	172.64(14)	P1-C1-C2-C3	88.3(2)
P1-C1-C2-C5	-90.3(2)	C5-C2-C3-C4	2.0(3)
C1-C2-C3-C4	-176.7(2)	C2-C3-C4-C7	0.7(4)
C3-C2-C5-C6	-2.8(3)	C1-C2-C5-C6	175.83(19)
C3-C2-C5-C8	177.15(19)	C1-C2-C5-C8	-4.3(3)
C2-C5-C6-C7	0.9(3)	C8-C5-C6-C7	-179.0(2)
C5-C6-C7-C4	1.8(3)	C3-C4-C7-C6	-2.6(3)
O13-P2-C8-C5	-30.04(17)	O19-P2-C8-C5	-150.99(14)
O4-P2-C8-C5	87.59(16)	C11-N1-C9-C10	-0.7(3)
N1-C9-C10-C12	-0.7(3)	C9-N1-C11-C13	0.9(3)
C14-N2-C12-C13	-24.2(3)	C14-N2-C12-C10	159.3(2)
C9-C10-C12-N2	178.59(19)	C9-C10-C12-C13	1.8(3)
N1-C11-C13-C12	0.3(3)	N2-C12-C13-C11	-178.0(2)
C10-C12-C13-C11	-1.6(3)	C12-N2-C14-C16	0.9(4)
C12-N2-C14-C15	-177.4(2)	N2-C14-C15-C17	176.4(2)
C16-C14-C15-C17	-2.1(3)	N2-C14-C16-C18	-176.4(2)
C15-C14-C16-C18	1.8(3)	C18-N3-C17-C15	1.0(4)
C14-C15-C17-N3	0.7(4)	C17-N3-C18-C16	-1.2(3)
C14-C16-C18-N3	-0.2(3)	C20-N4-C19-C21	1.1(3)
C19-N4-C20-C22	-1.6(3)	N4-C19-C21-C23	-0.7(3)
N4-C20-C22-C23	1.6(3)	C24-N5-C23-C21	160.6(2)
C24-N5-C23-C22	-21.6(3)	C19-C21-C23-N5	178.56(19)

C19-C21-C23-C22	0.7(3)	C20-C22-C23-N5	-178.85(19)
C20-C22-C23-C21	-1.1(3)	C23-N5-C24-C25	-20.7(3)
C23-N5-C24-C26	162.8(2)	N5-C24-C25-C27	-179.45(19)
C26-C24-C25-C27	-3.0(3)	N5-C24-C26-C28	178.63(19)
C25-C24-C26-C28	1.9(3)	C28-N6-C27-C25	0.9(3)
C24-C25-C27-N6	1.6(3)	C27-N6-C28-C26	-2.0(3)
C24-C26-C28-N6	0.6(3)		

Table S7. Anisotropic atomic displacement parameters ($Å^2$) for $(H_2\text{-}4,4'dpa)_2[Mo_5O_{15}(1,2\text{-}O_3PC_8H_8PO_3)\cdot H_2O$ ($1\cdot H_2O$).

The anisotropic atomic displacement factor exponent takes the form: $-2\pi^2[h^2 a^{*2} U_{11} + ... + 2 h k a^* b^* U_{12}]$

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
Mo1	0.00964(8)	0.01110(9)	0.00978(8)	-0.00202(6)	0.00365(6)	-0.00216(6)
Mo2	0.00743(8)	0.00936(8)	0.01063(8)	-0.00080(6)	0.00351(6)	0.00032(6)
Mo3	0.00785(8)	0.00928(8)	0.01038(8)	-0.00175(6)	0.00324(6)	-0.00085(6)
Mo4	0.00837(8)	0.01080(9)	0.01146(8)	0.00015(6)	0.00369(6)	0.00100(6)
Mo5	0.01236(8)	0.00898(9)	0.01100(8)	-0.00158(6)	0.00462(6)	-0.00007(6)
P1	0.0096(2)	0.0095(2)	0.0080(2)	-0.00005(19)	0.00341(18)	-0.00013(19)
P2	0.0094(2)	0.0103(2)	0.0086(2)	0.00027(19)	0.00391(18)	-0.00046(19)
O1	0.0180(8)	0.0151(8)	0.0200(8)	-0.0039(6)	0.0100(6)	-0.0054(6)
O2	0.0142(7)	0.0225(8)	0.0133(7)	-0.0018(6)	0.0011(6)	-0.0014(6)
O3	0.0093(7)	0.0139(7)	0.0136(7)	-0.0021(6)	0.0045(5)	-0.0014(5)
O4	0.0099(6)	0.0102(7)	0.0104(7)	-0.0006(5)	0.0042(5)	-0.0004(5)
O5	0.0133(7)	0.0129(7)	0.0103(7)	-0.0031(6)	0.0043(5)	-0.0004(6)
O6	0.0120(7)	0.0101(7)	0.0103(7)	-0.0005(5)	0.0045(5)	-0.0007(5)
O7	0.0112(7)	0.0115(7)	0.0104(7)	-0.0003(5)	0.0048(5)	0.0001(5)
O8	0.0113(7)	0.0144(7)	0.0152(7)	-0.0019(6)	0.0055(6)	0.0006(6)
O9	0.0121(7)	0.0145(7)	0.0149(7)	0.0006(6)	0.0044(6)	0.0028(6)
O10	0.0096(7)	0.0098(7)	0.0133(7)	-0.0021(5)	0.0038(5)	-0.0005(5)
O11	0.0128(7)	0.0177(8)	0.0136(7)	-0.0037(6)	0.0036(6)	-0.0010(6)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
O12	0.0137(7)	0.0124(7)	0.0182(7)	-0.0017(6)	0.0053(6)	-0.0021(6)
O13	0.0109(7)	0.0111(7)	0.0113(7)	0.0000(5)	0.0039(5)	-0.0010(5)
O14	0.0095(6)	0.0108(7)	0.0113(7)	-0.0005(5)	0.0041(5)	0.0001(5)
O15	0.0091(7)	0.0116(7)	0.0128(7)	-0.0007(6)	0.0039(5)	-0.0003(5)
O16	0.0129(7)	0.0155(8)	0.0163(7)	0.0019(6)	0.0015(6)	0.0010(6)
O17	0.0157(7)	0.0162(8)	0.0228(8)	-0.0007(6)	0.0105(6)	0.0014(6)
O18	0.0130(7)	0.0102(7)	0.0134(7)	0.0004(6)	0.0038(6)	0.0021(5)
O19	0.0118(7)	0.0117(7)	0.0117(7)	0.0000(6)	0.0049(5)	0.0000(5)
O20	0.0171(7)	0.0147(7)	0.0170(7)	-0.0028(6)	0.0069(6)	0.0006(6)
O21	0.0191(8)	0.0127(7)	0.0167(7)	-0.0010(6)	0.0055(6)	-0.0010(6)
O90	0.0427(12)	0.0583(15)	0.0411(12)	0.0191(10)	0.0291(10)	0.0165(11)
N1	0.0177(9)	0.0154(9)	0.0132(9)	0.0006(7)	0.0081(7)	0.0038(7)
N2	0.0150(9)	0.0115(9)	0.0140(9)	-0.0015(7)	0.0070(7)	-0.0016(7)
N3	0.0184(9)	0.0255(10)	0.0185(10)	0.0033(8)	0.0112(8)	-0.0016(8)
N4	0.0165(9)	0.0246(10)	0.0126(9)	0.0050(8)	0.0050(7)	0.0003(8)
N5	0.0140(9)	0.0126(9)	0.0143(9)	0.0020(7)	0.0051(7)	0.0046(7)
N6	0.0178(9)	0.0148(9)	0.0108(8)	0.0030(7)	0.0040(7)	0.0002(7)
C1	0.0134(10)	0.0155(10)	0.0093(9)	0.0010(8)	0.0036(8)	0.0035(8)
C2	0.0142(10)	0.0161(11)	0.0082(9)	0.0031(8)	0.0008(8)	0.0024(8)
C3	0.0294(12)	0.0180(11)	0.0141(10)	-0.0019(9)	0.0058(9)	0.0007(9)
C4	0.0359(14)	0.0174(12)	0.0173(11)	-0.0012(9)	0.0014(10)	-0.0081(10)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
C5	0.0127(10)	0.0144(10)	0.0084(9)	0.0009(8)	0.0003(7)	0.0006(8)
C6	0.0159(10)	0.0236(12)	0.0105(10)	0.0027(9)	0.0024(8)	0.0003(9)
C7	0.0215(11)	0.0248(12)	0.0149(11)	0.0045(9)	0.0003(9)	-0.0092(10)
C8	0.0194(10)	0.0149(10)	0.0094(9)	-0.0003(8)	0.0058(8)	-0.0009(8)
C9	0.0146(10)	0.0129(10)	0.0174(10)	0.0037(8)	0.0068(8)	0.0019(8)
C10	0.0148(10)	0.0118(10)	0.0138(10)	0.0011(8)	0.0047(8)	0.0002(8)
C11	0.0175(10)	0.0157(11)	0.0142(10)	-0.0006(8)	0.0024(8)	0.0007(8)
C12	0.0133(10)	0.0115(10)	0.0122(10)	0.0040(8)	0.0040(8)	0.0043(8)
C13	0.0128(10)	0.0148(10)	0.0147(10)	0.0015(8)	0.0036(8)	0.0000(8)
C14	0.0102(9)	0.0157(10)	0.0135(10)	0.0037(8)	0.0040(8)	0.0029(8)
C15	0.0205(11)	0.0182(11)	0.0188(11)	-0.0026(9)	0.0087(9)	-0.0030(9)
C16	0.0148(10)	0.0150(10)	0.0161(10)	0.0002(8)	0.0060(8)	0.0024(8)
C17	0.0246(12)	0.0268(13)	0.0190(11)	-0.0039(10)	0.0121(9)	-0.0034(10)
C18	0.0132(10)	0.0161(11)	0.0216(11)	0.0037(9)	0.0045(8)	0.0008(8)
C19	0.0147(10)	0.0172(11)	0.0182(11)	0.0068(9)	0.0011(8)	0.0001(8)
C20	0.0157(10)	0.0196(11)	0.0129(10)	-0.0003(8)	0.0026(8)	0.0019(9)
C21	0.0137(10)	0.0133(10)	0.0174(10)	0.0016(8)	0.0017(8)	0.0012(8)
C22	0.0132(10)	0.0121(10)	0.0126(10)	0.0010(8)	0.0012(8)	-0.0004(8)
C23	0.0087(9)	0.0153(10)	0.0113(10)	0.0014(8)	0.0000(7)	-0.0020(8)
C24	0.0142(10)	0.0100(10)	0.0094(9)	-0.0028(8)	0.0012(8)	-0.0019(8)
C25	0.0116(9)	0.0138(10)	0.0095(9)	-0.0023(8)	0.0031(7)	-0.0017(8)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
C26	0.0117(10)	0.0149(10)	0.0154(10)	-0.0016(8)	0.0039(8)	0.0021(8)
C27	0.0132(10)	0.0141(10)	0.0130(10)	-0.0013(8)	0.0033(8)	0.0014(8)
C28	0.0165(10)	0.0170(11)	0.0145(10)	-0.0018(8)	0.0075(8)	-0.0021(8)

Table S8. Hydrogen atomic coordinates and isotropic atomic displacement parameters ($Å^2$) for $(H_2\text{-}4,4'dpa)_2[Mo_5O_{15}(1,2\text{-}O_3PC_8H_8PO_3)\cdot H_2O$ ($1\cdot H_2O$).

	x/a	y/b	z/c	U(eq)
H90A	0.069(2)	0.8638(18)	0.784(2)	0.053
H90B	0.174(3)	0.8801(15)	0.8103(18)	0.053
H1A	0.9702	0.6291	0.5902	0.015
H1B	0.9581	0.6953	0.6393	0.015
H3	0.8369	0.5353	0.5784	0.025
H4	0.7026	0.4702	0.6249	0.03
H6	0.6597	0.6301	0.7560	0.02
H7	0.6083	0.5197	0.7109	0.026
H8A	0.8768	0.7591	0.2077	0.017
H8B	0.7383	0.7468	0.2050	0.017
H9	0.0380	0.5871	0.7965	0.017
H10	0.1101	0.5503	0.6964	0.016
H11	0.3154	0.7144	0.8656	0.019
H13	0.3943	0.6815	0.7669	0.017
H15	0.3502	0.5308	0.5486	0.022
H16	0.4275	0.7054	0.6747	0.018
H17	0.4650	0.5724	0.4716	0.027
H18	0.5413	0.7420	0.5946	0.02
H19	0.2919	0.8033	0.6734	0.021

	x/a	y/b	z/c	U(eq)
H20	0.1816	0.9963	0.6696	0.02
H21	0.3616	0.8215	0.5673	0.018
H22	0.2508	1.0203	0.5641	0.016
H25	0.1661	1.0139	0.4479	0.014
H26	0.4819	0.9630	0.4000	0.017
H27	0.1348	1.0959	0.3531	0.016
H28	0.4392	1.0439	0.3040	0.018
H2'	0.255(2)	0.5535(11)	0.6377(14)	0.018(6)
H6'	0.269(2)	1.1029(12)	0.2824(12)	0.023(7)
H1'	0.145(2)	0.6685(14)	0.8754(13)	0.028(7)
H5'	0.418(2)	0.9059(13)	0.4913(16)	0.027(7)
H4'	0.206(2)	0.8900(15)	0.7176(13)	0.031(8)
H3'	0.544(2)	0.6784(14)	0.4952(14)	0.026(7)

Table S9. Sample and crystal data for $(H_2$4,4'-dpa$)_2[Mo_5O_{15}(1,3\text{-}O_3PC_8H_8PO_3)]$ (**2**).

Identification code	$(H_2$4,4'-dpa$)_2[Mo_5O_{15}(1,3\text{-}O_3PC_8H_8PO_3)]$	
Chemical formula	$C_{28}H_{30}Mo_5N_6O_{21}P_2$	
Formula weight	1328.22	
Temperature	90(2) K	
Wavelength	0.71073 Å	
Crystal size	0.080 x 0.154 x 0.265 mm	
Crystal system	Trigonal	
Space group	P 32	
Unit cell dimensions	a = 11.1778(7) Å	α = 90°
	b = 11.1778(7) Å	β = 90°
	c = 27.6410(17) Å	γ = 120°
Volume	2990.9(4) $Å^3$	
Z	3	
Density (calculated)	2.212 g/cm^3	
Absorption coefficient	1.701 mm^{-1}	
F(000)	1944	

Table S10. Data collection and structure refinement for $(H_2$4,4'-dpa$)_2$ $[Mo_5O_{15}(1,3\text{-}O_3PC_8H_8PO_3)]$ (**2**).

Theta range for data collection	2.10 to 27.09°	
Index ranges	-14<=h<=13, -13<=k<=14, -35<=l<=35	
Reflections collected	32751	
Independent reflections	8572 [R(int) = 0.0209]	
Coverage of independent reflections	100.0%	
Max. and min. transmission	0.8760 and 0.6610	
Refinement method	Full-matrix least-squares on F^2	
Refinement program	SHELXL-2013 (Sheldrick, 2013)	
Function minimized	$\Sigma\, w(F_o^2 - F_c^2)^2$	
Data / restraints / parameters	8572 / 1 / 560	
Goodness-of-fit on F^2	1.049	
Δ/σ_{max}	0.001	
Final R indices	8346 data; I>2σ(I)	R1 = 0.0338, wR2 = 0.0902
	all data	R1 = 0.0350, wR2 = 0.0908
Weighting scheme	$w=1/[\sigma^2(F_o^2)+(0.0509P)^2+12.8222P]$ where $P=(F_o^2+2F_c^2)/3$	
Absolute structure parameter	0.0(0)	
Largest diff. peak and hole	1.133 and -0.560 eÅ^{-3}	
R.M.S. deviation from mean	0.143 eÅ^{-3}	

Table S11. Atomic coordinates and equivalent isotropic atomic displacement parameters ($Å^2$) for $(H_2$4,4'-dpa$)_2[Mo_5O_{15}(1,3\text{-}O_3PC_8H_8PO_3)]$ (**2**).

U(eq) is defined as one third of the trace of the orthogonalized U_{ij} tensor.

	x/a	**y/b**	**z/c**	**U(eq)**
Mo1	0.15857(7)	0.71566(7)	0.31185(3)	0.01639(15)
Mo2	0.94565(8)	0.81303(7)	0.36164(3)	0.01747(16)
Mo3	0.67841(7)	0.53954(8)	0.41701(3)	0.01796(16)
Mo4	0.06339(7)	0.39100(7)	0.35212(3)	0.01630(15)
Mo5	0.70357(7)	0.25655(7)	0.39677(2)	0.01626(15)
P1	0.8103(2)	0.4909(2)	0.30721(7)	0.0146(4)
P2	0.0028(2)	0.5839(2)	0.42716(7)	0.0132(4)
O1	0.3067(7)	0.8529(7)	0.3339(2)	0.0273(15)
O2	0.1982(7)	0.7119(7)	0.2516(2)	0.0262(14)
O3	0.9598(6)	0.5185(6)	0.3086(2)	0.0162(12)
O4	0.1967(6)	0.5754(6)	0.3373(2)	0.0167(11)
O5	0.0399(7)	0.3137(7)	0.2967(2)	0.0218(13)
O6	0.1558(7)	0.3306(7)	0.3818(2)	0.0220(13)
O7	0.8809(6)	0.2773(6)	0.3777(2)	0.0174(12)
O8	0.0770(6)	0.5019(6)	0.4239(2)	0.0156(11)
O9	0.7012(7)	0.2063(7)	0.4553(2)	0.0236(13)
O10	0.5943(6)	0.3470(7)	0.4013(2)	0.0213(13)
O11	0.7122(6)	0.3391(6)	0.3187(2)	0.0184(12)

	x/a	y/b	z/c	U(eq)
O12	0.8451(6)	0.4852(6)	0.4219(2)	0.0149(11)
O13	0.5506(7)	0.5660(8)	0.3945(3)	0.0277(15)
O14	0.0561(6)	0.6982(6)	0.3885(2)	0.0155(11)
O15	0.5899(7)	0.1041(7)	0.3696(2)	0.0256(14)
O16	0.8530(7)	0.8724(7)	0.3297(2)	0.0258(14)
O17	0.6559(7)	0.5327(7)	0.4779(2)	0.0295(15)
O18	0.7903(6)	0.5861(6)	0.3427(2)	0.0138(11)
O19	0.8328(6)	0.7290(6)	0.4166(2)	0.0185(12)
O20	0.0728(7)	0.9592(7)	0.3880(2)	0.0250(14)
O21	0.0409(7)	0.7919(6)	0.3064(2)	0.0192(12)
N1	0.8333(11)	0.7765(9)	0.1264(3)	0.039(2)
N2	0.0388(8)	0.0141(8)	0.2461(3)	0.0221(16)
N3	0.4392(8)	0.2125(8)	0.2963(3)	0.0219(16)
N4	0.3991(9)	0.9184(9)	0.4390(3)	0.0320(19)
N5	0.3503(10)	0.1309(10)	0.5521(3)	0.034(2)
N6	0.3886(11)	0.5122(11)	0.5707(4)	0.045(3)
C1	0.7740(9)	0.5304(9)	0.2476(3)	0.0168(16)
C2	0.0306(9)	0.6687(8)	0.4853(3)	0.0150(15)
C3	0.0164(8)	0.5813(8)	0.5285(3)	0.0139(15)
C4	0.8955(9)	0.4524(9)	0.5362(3)	0.0164(16)
C5	0.8863(8)	0.3694(9)	0.5756(3)	0.0172(16)

	x/a	y/b	z/c	U(eq)
C6	0.9979(9)	0.4175(9)	0.6077(3)	0.0198(17)
C7	0.1128(9)	0.5454(9)	0.6014(3)	0.0207(17)
C8	0.1243(9)	0.6282(9)	0.5622(3)	0.0186(17)
C9	0.9712(13)	0.8579(12)	0.1230(4)	0.037(3)
C10	0.7645(12)	0.7763(10)	0.1656(4)	0.034(2)
C11	0.0438(12)	0.9424(11)	0.1613(4)	0.035(2)
C12	0.9781(10)	0.9392(9)	0.2045(3)	0.0238(19)
C13	0.8322(11)	0.8525(10)	0.2066(4)	0.029(2)
C14	0.1759(9)	0.0794(8)	0.2610(3)	0.0180(16)
C15	0.2811(11)	0.0671(10)	0.2379(3)	0.0258(19)
C16	0.2073(9)	0.1579(9)	0.3026(3)	0.0196(17)
C17	0.4107(10)	0.1381(10)	0.2565(4)	0.027(2)
C18	0.3429(9)	0.2259(9)	0.3205(3)	0.0206(17)
C19	0.3388(10)	0.9954(10)	0.4300(4)	0.028(2)
C20	0.4364(11)	0.9066(11)	0.4834(4)	0.031(2)
C21	0.3194(10)	0.0669(10)	0.4656(4)	0.030(2)
C22	0.4231(11)	0.9787(11)	0.5210(4)	0.030(2)
C23	0.3644(11)	0.0617(10)	0.5125(4)	0.028(2)
C24	0.3573(10)	0.2558(11)	0.5552(4)	0.031(2)
C25	0.3817(12)	0.3449(12)	0.5168(5)	0.038(3)
C26	0.3463(13)	0.2984(14)	0.6027(5)	0.043(3)

	x/a	**y/b**	**z/c**	**U(eq)**
C27	0.3966(12)	0.4746(12)	0.5252(5)	0.044(3)
C28	0.3634(12)	0.4262(14)	0.6094(5)	0.045(3)

Table S12. Bond lengths (Å) for $(H_2$4,4'-dpa$)_2[Mo_5O_{15}(1,3\text{-}O_3PC_8H_8PO_3)]$ (**2**).

Mo1-O1	1.711(7)	Mo1-O2	1.728(6)
Mo1-O21	1.898(6)	Mo1-O4	1.950(6)
Mo1-O3	2.214(6)	Mo1-O14	2.370(6)
Mo2-O20	1.702(7)	Mo2-O16	1.726(7)
Mo2-O19	1.897(6)	Mo2-O21	1.941(6)
Mo2-O14	2.304(6)	Mo2-O18	2.306(6)
Mo3-O17	1.698(7)	Mo3-O13	1.714(6)
Mo3-O10	1.919(6)	Mo3-O19	1.952(6)
Mo3-O12	2.234(6)	Mo3-O18	2.326(6)
Mo4-O6	1.700(6)	Mo4-O5	1.714(6)
Mo4-O4	1.888(6)	Mo4-O7	1.919(6)
Mo4-O8	2.304(6)	Mo5-O15	1.708(6)
Mo5-O9	1.710(7)	Mo5-O10	1.940(6)
Mo5-O7	1.950(6)	Mo5-O11	2.330(6)
Mo5-O12	2.339(6)	P1-O11	1.524(6)
P1-O3	1.541(6)	P1-O18	1.543(6)
P1-C1	1.803(8)	P2-O8	1.516(6)
P2-O14	1.539(6)	P2-O12	1.550(6)
P2-C2	1.811(8)	N1-C10	1.328(16)
N1-C9	1.345(16)	N1-H1'	0.88
N2-C12	1.382(12)	N2-C14	1.391(11)

N2-H2'	0.88	N3-C17	1.319(13)
N3-C18	1.337(12)	N3-H3'	0.88
N4-C20	1.323(14)	N4-C19	1.357(14)
N4-H4'	0.88	N5-C24	1.362(14)
N5-C23	1.394(13)	N5-H5'	0.88
N6-C27	1.343(18)	N6-C28	1.370(19)
N6-H6'	0.88	C1-C5	1.540(11)
C1-H1A	0.99	C1-H1B	0.99
C2-C3	1.501(11)	C2-H2A	0.99
C2-H2B	0.99	C3-C8	1.402(12)
C3-C4	1.415(12)	C4-C5	1.399(12)
C4-H4	0.95	C5-C6	1.401(12)
C5-C1	1.540(11)	C6-C7	1.374(12)
C6-H6	0.95	C7-C8	1.390(12)
C7-H7	0.95	C8-H8	0.95
C9-C11	1.380(15)	C9-H9	0.95
C10-C13	1.393(14)	C10-H10	0.95
C11-C12	1.394(15)	C11-H11	0.95
C12-C13	1.422(14)	C13-H13	0.95
C14-C16	1.380(12)	C14-C15	1.405(13)
C15-C17	1.357(14)	C15-H15	0.95
C16-C18	1.403(12)	C16-H16	0.95

C17-H17	0.95	C18-H18	0.95
C19-C21	1.351(15)	C19-H19	0.95
C20-C22	1.369(15)	C20-H20	0.95
C21-C23	1.403(15)	C21-H21	0.95
C22-C23	1.398(15)	C22-H22	0.95
C24-C25	1.388(16)	C24-C26	1.421(16)
C25-C27	1.394(17)	C25-H25	0.95
C26-C28	1.356(19)	C26-H26	0.95
C27-H27	0.95	C28-H28	0.95

Table S13. Bond angles (°) for $(H_2$4,4'-dpa$)_2[Mo_5O_{15}(1,3\text{-}O_3PC_8H_8PO_3)]$ (**2**).

O1-Mo1-O2	102.7(3)	O1-Mo1-O21	101.7(3)
O2-Mo1-O21	100.3(3)	O1-Mo1-O4	95.2(3)
O2-Mo1-O4	99.6(3)	O21-Mo1-O4	150.3(2)
O1-Mo1-O3	161.2(3)	O2-Mo1-O3	94.5(3)
O21-Mo1-O3	82.4(2)	O4-Mo1-O3	74.2(2)
O1-Mo1-O14	88.1(3)	O2-Mo1-O14	168.0(3)
O21-Mo1-O14	72.1(2)	O4-Mo1-O14	84.4(2)
O3-Mo1-O14	75.6(2)	O20-Mo2-O16	102.9(3)
O20-Mo2-O19	99.3(3)	O16-Mo2-O19	102.8(3)
O20-Mo2-O21	102.0(3)	O16-Mo2-O21	96.7(3)
O19-Mo2-O21	147.0(2)	O20-Mo2-O14	88.5(3)
O16-Mo2-O14	166.1(3)	O19-Mo2-O14	82.8(2)
O21-Mo2-O14	72.9(2)	O20-Mo2-O18	164.0(3)
O16-Mo2-O18	92.3(3)	O19-Mo2-O18	71.9(2)
O21-Mo2-O18	81.1(2)	O14-Mo2-O18	77.3(2)
O17-Mo3-O13	104.5(3)	O17-Mo3-O10	101.3(3)
O13-Mo3-O10	97.3(3)	O17-Mo3-O19	94.8(3)
O13-Mo3-O19	99.9(3)	O10-Mo3-O19	152.5(3)
O17-Mo3-O12	93.0(3)	O13-Mo3-O12	161.6(3)
O10-Mo3-O12	73.5(2)	O19-Mo3-O12	83.7(2)
O17-Mo3-O18	159.6(3)	O13-Mo3-O18	92.4(3)

O10-Mo3-O18	87.6(2)	O19-Mo3-O18	70.6(2)
O12-Mo3-O18	71.7(2)	O6-Mo4-O5	101.6(3)
O6-Mo4-O4	103.5(3)	O5-Mo4-O4	101.2(3)
O6-Mo4-O7	102.4(3)	O5-Mo4-O7	98.7(3)
O4-Mo4-O7	143.1(3)	O6-Mo4-O8	84.7(3)
O5-Mo4-O8	173.5(3)	O4-Mo4-O8	78.2(2)
O7-Mo4-O8	78.6(2)	O15-Mo5-O9	102.0(3)
O15-Mo5-O10	101.5(3)	O9-Mo5-O10	101.6(3)
O15-Mo5-O7	101.8(3)	O9-Mo5-O7	98.3(3)
O10-Mo5-O7	145.3(3)	O15-Mo5-O11	81.4(3)
O9-Mo5-O11	176.4(3)	O10-Mo5-O11	76.4(2)
O7-Mo5-O11	82.2(2)	O15-Mo5-O12	168.4(3)
O9-Mo5-O12	88.2(3)	O10-Mo5-O12	70.7(2)
O7-Mo5-O12	81.9(2)	O11-Mo5-O12	88.3(2)
O11-P1-O3	108.8(3)	O11-P1-O18	111.4(3)
O3-P1-O18	111.3(3)	O11-P1-C1	109.8(4)
O3-P1-C1	109.1(4)	O18-P1-C1	106.5(4)
O8-P2-O14	111.1(3)	O8-P2-O12	109.6(3)
O14-P2-O12	110.9(3)	O8-P2-C2	111.5(4)
O14-P2-C2	106.7(4)	O12-P2-C2	107.0(4)
P1-O3-Mo1	130.4(3)	Mo4-O4-Mo1	125.9(3)
Mo4-O7-Mo5	149.2(3)	P2-O8-Mo4	117.8(3)

Mo3-O10-Mo5	121.1(3)	P1-O11-Mo5	119.9(3)
P2-O12-Mo3	128.2(3)	P2-O12-Mo5	135.5(3)
Mo3-O12-Mo5	94.5(2)	P2-O14-Mo2	126.4(3)
P2-O14-Mo1	131.3(3)	Mo2-O14-Mo1	92.1(2)
P1-O18-Mo2	127.9(3)	P1-O18-Mo3	130.8(3)
Mo2-O18-Mo3	94.0(2)	Mo2-O19-Mo3	123.3(3)
Mo1-O21-Mo2	122.6(3)	C10-N1-C9	121.7(9)
C10-N1-H1'	119.1	C9-N1-H1'	119.1
C12-N2-C14	130.0(9)	C12-N2-H2'	115.0
C14-N2-H2'	115.0	C17-N3-C18	122.2(8)
C17-N3-H3'	118.9	C18-N3-H3'	118.9
C20-N4-C19	121.0(9)	C20-N4-H4'	119.5
C19-N4-H4'	119.5	C24-N5-C23	131.0(9)
C24-N5-H5'	114.5	C23-N5-H5'	114.5
C27-N6-C28	122.2(11)	C27-N6-H6'	118.9
C28-N6-H6'	118.9	C5-C1-P1	114.5(6)
C5-C1-H1A	108.6	P1-C1-H1A	108.6
C5-C1-H1B	108.6	P1-C1-H1B	108.6
H1A-C1-H1B	107.6	C3-C2-P2	115.5(6)
C3-C2-H2A	108.4	P2-C2-H2A	108.4
C3-C2-H2B	108.4	P2-C2-H2B	108.4
H2A-C2-H2B	107.5	C8-C3-C4	119.0(7)

C8-C3-C2	119.7(7)	C4-C3-C2	121.3(7)
C5-C4-C3	120.6(7)	C5-C4-H4	119.7
C3-C4-H4	119.7	C4-C5-C6	118.8(8)
C4-C5-C1	118.9(8)	C6-C5-C1	122.2(8)
C7-C6-C5	120.6(8)	C7-C6-H6	119.7
C5-C6-H6	119.7	C6-C7-C8	121.2(8)
C6-C7-H7	119.4	C8-C7-H7	119.4
C7-C8-C3	119.6(8)	C7-C8-H8	120.2
C3-C8-H8	120.2	N1-C9-C11	119.5(11)
N1-C9-H9	120.3	C11-C9-H9	120.3
N1-C10-C13	121.7(11)	N1-C10-H10	119.2
C13-C10-H10	119.2	C9-C11-C12	121.2(10)
C9-C11-H11	119.4	C12-C11-H11	119.4
N2-C12-C11	127.2(9)	N2-C12-C13	115.3(9)
C11-C12-C13	117.5(9)	C10-C13-C12	118.1(11)
C10-C13-H13	120.9	C12-C13-H13	120.9
C16-C14-N2	116.4(8)	C16-C14-C15	118.8(8)
N2-C14-C15	124.7(8)	C17-C15-C14	118.1(8)
C17-C15-H15	120.9	C14-C15-H15	120.9
C14-C16-C18	120.0(9)	C14-C16-H16	120.0
C18-C16-H16	120.0	N3-C17-C15	122.4(9)
N3-C17-H17	118.8	C15-C17-H17	118.8

N3-C18-C16	118.4(8)	N3-C18-H18	120.8
C16-C18-H18	120.8	C21-C19-N4	121.2(9)
C21-C19-H19	119.4	N4-C19-H19	119.4
N4-C20-C22	120.9(10)	N4-C20-H20	119.6
C22-C20-H20	119.6	C19-C21-C23	118.8(10)
C19-C21-H21	120.6	C23-C21-H21	120.6
C20-C22-C23	119.2(10)	C20-C22-H22	120.4
C23-C22-H22	120.4	N5-C23-C22	117.2(9)
N5-C23-C21	124.1(10)	C22-C23-C21	118.7(9)
N5-C24-C25	125.2(10)	N5-C24-C26	115.6(11)
C25-C24-C26	119.0(11)	C24-C25-C27	119.8(12)
C24-C25-H25	120.1	C27-C25-H25	120.1
C28-C26-C24	119.3(13)	C28-C26-H26	120.3
C24-C26-H26	120.3	N6-C27-C25	119.4(13)
N6-C27-H27	120.3	C25-C27-H27	120.3
C26-C28-N6	120.3(12)	C26-C28-H28	119.9
N6-C28-H28	119.9		

Table S14. Torsion angles (°) for $(H_2$4,4'-dpa$)_2[Mo_5O_{15}(1,3\text{-}O_3PC_8H_8PO_3)]$ (**2**).

O11-P1-O3-Mo1	-164.9(4)	O18-P1-O3-Mo1	-41.8(5)
C1-P1-O3-Mo1	75.4(5)	O6-Mo4-O4-Mo1	-170.2(4)
O5-Mo4-O4-Mo1	84.8(4)	O7-Mo4-O4-Mo1	-36.6(6)
O8-Mo4-O4-Mo1	-88.6(4)	O14-P2-O8-Mo4	-59.0(4)
O12-P2-O8-Mo4	63.9(4)	C2-P2-O8-Mo4	-177.8(3)
O3-P1-O11-Mo5	70.2(4)	O18-P1-O11-Mo5	-52.8(5)
C1-P1-O11-Mo5	-170.5(4)	O8-P2-O12-Mo3	-174.3(4)
O14-P2-O12-Mo3	-51.3(5)	C2-P2-O12-Mo3	64.7(5)
O8-P2-O12-Mo5	-13.7(6)	O14-P2-O12-Mo5	109.3(5)
C2-P2-O12-Mo5	-134.7(5)	O8-P2-O14-Mo2	161.6(4)
O12-P2-O14-Mo2	39.4(5)	C2-P2-O14-Mo2	-76.8(5)
O8-P2-O14-Mo1	26.4(5)	O12-P2-O14-Mo1	-95.7(5)
C2-P2-O14-Mo1	148.1(4)	O11-P1-O18-Mo2	157.8(4)
O3-P1-O18-Mo2	36.2(5)	C1-P1-O18-Mo2	-82.6(5)
O11-P1-O18-Mo3	15.6(5)	O3-P1-O18-Mo3	-106.0(4)
C1-P1-O18-Mo3	135.2(4)	O20-Mo2-O19-Mo3	-169.8(4)
O16-Mo2-O19-Mo3	84.6(4)	O21-Mo2-O19-Mo3	-40.1(7)
O14-Mo2-O19-Mo3	-82.5(4)	O18-Mo2-O19-Mo3	-3.6(3)
O1-Mo1-O21-Mo2	-79.2(4)	O2-Mo1-O21-Mo2	175.4(4)
O4-Mo1-O21-Mo2	44.0(7)	O3-Mo1-O21-Mo2	82.2(4)
O14-Mo1-O21-Mo2	4.9(3)	O11-P1-C1-C5	64.9(7)

O3-P1-C1-C5	-176.0(6)	O18-P1-C1-C5	-55.8(7)
O8-P2-C2-C3	-45.1(7)	O14-P2-C2-C3	-166.5(6)
O12-P2-C2-C3	74.8(7)	P2-C2-C3-C8	126.2(7)
P2-C2-C3-C4	-54.4(10)	C8-C3-C4-C5	-3.4(12)
C2-C3-C4-C5	177.2(8)	C3-C4-C5-C6	1.3(12)
C3-C4-C5-C1	-176.3(7)	C4-C5-C6-C7	1.7(13)
C1-C5-C6-C7	179.2(8)	C5-C6-C7-C8	-2.6(14)
C6-C7-C8-C3	0.4(14)	C4-C3-C8-C7	2.5(12)
C2-C3-C8-C7	-178.1(8)	C10-N1-C9-C11	0.9(17)
C9-N1-C10-C13	-4.9(16)	N1-C9-C11-C12	3.7(18)
C14-N2-C12-C11	-19.5(16)	C14-N2-C12-C13	162.0(9)
C9-C11-C12-N2	177.5(10)	C9-C11-C12-C13	-4.0(16)
N1-C10-C13-C12	4.2(15)	N2-C12-C13-C10	178.8(8)
C11-C12-C13-C10	0.2(14)	C12-N2-C14-C16	174.0(8)
C12-N2-C14-C15	-8.2(14)	C16-C14-C15-C17	-2.7(13)
N2-C14-C15-C17	179.5(9)	N2-C14-C16-C18	179.3(8)
C15-C14-C16-C18	1.3(13)	C18-N3-C17-C15	-1.6(15)
C14-C15-C17-N3	2.9(15)	C17-N3-C18-C16	0.0(13)
C14-C16-C18-N3	0.1(13)	C20-N4-C19-C21	2.5(15)
C19-N4-C20-C22	-4.1(16)	N4-C19-C21-C23	0.8(15)
N4-C20-C22-C23	2.4(16)	C24-N5-C23-C22	147.2(11)
C24-N5-C23-C21	-35.1(18)	C20-C22-C23-N5	178.7(10)

C20-C22-C23-C21	0.8(15)	C19-C21-C23-N5	179.9(10)
C19-C21-C23-C22	-2.4(15)	C23-N5-C24-C25	0.1(19)
C23-N5-C24-C26	-176.2(11)	N5-C24-C25-C27	-175.3(10)
C26-C24-C25-C27	0.8(16)	N5-C24-C26-C28	174.8(11)
C25-C24-C26-C28	-1.7(17)	C28-N6-C27-C25	-1.3(17)
C24-C25-C27-N6	0.7(17)	C24-C26-C28-N6	1.1(18)
C27-N6-C28-C26	0.4(18)		

Table S15. Anisotropic atomic displacement parameters ($Å^2$) for $(H_2$4,4'-dpa$)_2[Mo_5O_{15}$(1,3-$O_3PC_8H_8PO_3$)] (**2**).

The anisotropic atomic displacement factor exponent takes the form: $-2\pi^2[h^2 a^{*2} U_{11} + ... + 2 h k a^* b^* U_{12}]$

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
Mo1	0.0161(3)	0.0140(3)	0.0137(3)	0.0011(3)	0.0021(3)	0.0035(3)
Mo2	0.0213(4)	0.0154(3)	0.0163(3)	-0.0014(3)	-0.0038(3)	0.0096(3)
Mo3	0.0146(3)	0.0225(4)	0.0176(3)	-0.0043(3)	-0.0003(3)	0.0099(3)
Mo4	0.0160(3)	0.0174(3)	0.0172(3)	0.0018(3)	0.0031(3)	0.0097(3)
Mo5	0.0131(3)	0.0148(3)	0.0156(3)	0.0003(3)	0.0024(3)	0.0030(3)
P1	0.0128(10)	0.0156(10)	0.0141(9)	-0.0015(7)	-0.0022(7)	0.0062(8)
P2	0.0124(9)	0.0138(9)	0.0119(9)	0.0006(7)	-0.0001(7)	0.0053(8)
O1	0.025(3)	0.020(3)	0.020(3)	-0.001(2)	0.006(3)	-0.002(3)
O2	0.035(4)	0.023(3)	0.014(3)	0.002(2)	0.002(3)	0.009(3)
O3	0.014(3)	0.016(3)	0.019(3)	0.001(2)	-0.003(2)	0.008(2)
O4	0.010(3)	0.019(3)	0.018(3)	0.002(2)	0.000(2)	0.005(2)
O5	0.021(3)	0.027(3)	0.020(3)	-0.001(2)	0.000(2)	0.014(3)
O6	0.024(3)	0.021(3)	0.025(3)	-0.001(2)	-0.001(3)	0.014(3)
O7	0.015(3)	0.013(3)	0.022(3)	0.001(2)	0.004(2)	0.005(2)
O8	0.016(3)	0.015(3)	0.016(3)	0.001(2)	0.001(2)	0.008(2)
O9	0.024(3)	0.018(3)	0.021(3)	0.001(2)	0.004(3)	0.004(3)
O10	0.013(3)	0.026(3)	0.020(3)	-0.004(2)	0.001(2)	0.006(3)
O11	0.014(3)	0.019(3)	0.020(3)	-0.002(2)	-0.001(2)	0.006(2)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
O12	0.013(3)	0.016(3)	0.015(3)	0.003(2)	0.000(2)	0.006(2)
O13	0.020(3)	0.039(4)	0.033(4)	-0.008(3)	-0.003(3)	0.021(3)
O14	0.019(3)	0.015(3)	0.014(3)	0.004(2)	0.003(2)	0.010(2)
O15	0.024(3)	0.018(3)	0.025(3)	-0.005(3)	0.004(3)	0.003(3)
O16	0.032(4)	0.021(3)	0.028(3)	-0.004(3)	-0.008(3)	0.016(3)
O17	0.028(4)	0.033(4)	0.020(3)	-0.008(3)	0.001(3)	0.009(3)
O18	0.015(3)	0.012(3)	0.017(3)	0.000(2)	-0.002(2)	0.009(2)
O19	0.022(3)	0.015(3)	0.017(3)	-0.006(2)	-0.003(2)	0.008(2)
O20	0.031(4)	0.018(3)	0.025(3)	-0.004(3)	-0.005(3)	0.012(3)
O21	0.027(3)	0.017(3)	0.014(3)	0.003(2)	-0.001(2)	0.011(3)
N1	0.051(6)	0.025(4)	0.030(5)	-0.003(4)	-0.016(4)	0.011(4)
N2	0.021(4)	0.022(4)	0.024(4)	-0.003(3)	-0.008(3)	0.011(3)
N3	0.011(3)	0.023(4)	0.027(4)	0.004(3)	-0.001(3)	0.005(3)
N4	0.032(5)	0.027(4)	0.035(5)	-0.007(4)	0.000(4)	0.013(4)
N5	0.041(5)	0.030(5)	0.028(4)	0.002(4)	0.007(4)	0.016(4)
N6	0.033(5)	0.039(6)	0.075(8)	-0.022(5)	-0.016(5)	0.027(5)
C1	0.019(4)	0.023(4)	0.013(4)	-0.001(3)	-0.005(3)	0.013(3)
C2	0.017(4)	0.014(4)	0.011(3)	0.002(3)	-0.001(3)	0.005(3)
C3	0.015(4)	0.014(4)	0.012(4)	0.001(3)	0.004(3)	0.007(3)
C4	0.016(4)	0.024(4)	0.012(4)	0.000(3)	-0.001(3)	0.011(3)
C5	0.012(4)	0.016(4)	0.021(4)	0.000(3)	0.004(3)	0.005(3)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
C6	0.023(4)	0.020(4)	0.015(4)	0.004(3)	0.000(3)	0.010(4)
C7	0.013(4)	0.022(4)	0.021(4)	-0.002(3)	-0.004(3)	0.004(3)
C8	0.015(4)	0.017(4)	0.020(4)	0.002(3)	0.004(3)	0.005(3)
C9	0.049(7)	0.034(6)	0.022(5)	-0.004(4)	-0.005(4)	0.015(5)
C10	0.035(6)	0.018(5)	0.046(6)	-0.002(4)	-0.013(5)	0.011(4)
C11	0.038(6)	0.024(5)	0.027(5)	0.002(4)	-0.003(4)	0.004(5)
C12	0.030(5)	0.018(4)	0.020(4)	0.005(3)	-0.005(4)	0.010(4)
C13	0.029(5)	0.020(4)	0.039(5)	-0.002(4)	-0.016(4)	0.013(4)
C14	0.016(4)	0.007(4)	0.022(4)	0.005(3)	0.002(3)	-0.001(3)
C15	0.034(5)	0.021(5)	0.020(4)	-0.002(3)	0.006(4)	0.012(4)
C16	0.020(4)	0.015(4)	0.023(4)	0.002(3)	-0.002(3)	0.008(3)
C17	0.024(5)	0.025(5)	0.033(5)	0.001(4)	0.008(4)	0.014(4)
C18	0.021(4)	0.023(4)	0.018(4)	-0.002(3)	-0.004(3)	0.011(4)
C19	0.021(5)	0.029(5)	0.022(4)	0.001(4)	-0.001(4)	0.003(4)
C20	0.030(5)	0.024(5)	0.038(6)	-0.004(4)	-0.002(4)	0.013(4)
C21	0.019(5)	0.022(5)	0.041(6)	0.001(4)	-0.003(4)	0.004(4)
C22	0.029(5)	0.025(5)	0.026(5)	0.001(4)	0.001(4)	0.006(4)
C23	0.031(5)	0.021(5)	0.027(5)	0.002(4)	0.008(4)	0.010(4)
C24	0.015(4)	0.025(5)	0.041(6)	-0.009(4)	0.002(4)	0.000(4)
C25	0.030(6)	0.037(6)	0.050(7)	-0.001(5)	0.001(5)	0.019(5)
C26	0.043(7)	0.052(7)	0.040(6)	-0.011(5)	-0.015(5)	0.028(6)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
C27	0.029(6)	0.027(6)	0.071(9)	-0.004(5)	0.001(6)	0.011(5)
C28	0.031(6)	0.054(8)	0.051(7)	-0.023(6)	-0.015(5)	0.021(6)

Table S16. Hydrogen atomic coordinates and isotropic atomic displacement parameters ($Å^2$) for $(H_2$4,4'-dpa$)_2[Mo_5O_{15}(1,3\text{-}O_3PC_8H_8PO_3)]$ (**2**).

	x/a	y/b	z/c	U(eq)
H1'	-0.2124	-0.2781	0.1021	0.046
H2'	-0.0182	0.0216	0.2662	0.027
H3'	0.5245	0.2549	0.3073	0.026
H4'	0.4137	-0.1244	0.4150	0.038
H5'	0.3341	0.0863	0.5797	0.041
H6'	0.3998	0.5950	0.5759	0.054
H1A	0.8452	0.6261	0.2393	0.02
H1B	0.7816	0.4679	0.2241	0.02
H2A	1.1245	0.7507	0.4855	0.018
H2B	0.9639	0.7020	0.4886	0.018
H4	0.8198	0.4219	0.5146	0.02
H6	0.9941	0.3612	0.6341	0.024
H7	1.1859	0.5778	0.6243	0.025
H8	1.2047	0.7161	0.5583	0.022
H9	0.0182	-0.1427	0.0944	0.045
H10	-0.3335	-0.2773	0.1656	0.041
H11	0.1404	0.0038	0.1582	0.042
H13	-0.2177	-0.1532	0.2352	0.035
H15	0.2621	0.0107	0.2100	0.031

	x/a	y/b	z/c	U(eq)
H16	0.1370	0.1658	0.3191	0.024
H17	0.4835	0.1339	0.2403	0.032
H18	0.3658	0.2801	0.3491	0.025
H19	0.3096	-0.0007	0.3980	0.034
H20	0.4729	-0.1528	0.4892	0.037
H21	0.2760	0.1196	0.4589	0.036
H22	0.4533	-0.0276	0.5525	0.036
H25	0.3881	0.3176	0.4848	0.046
H26	0.3272	0.2380	0.6294	0.052
H27	0.4123	0.5359	0.4991	0.053
H28	0.3579	0.4561	0.6411	0.054

Table S17. Sample and crystal data for $[Co(terpy)_2](H_3O)[Mo_5O_{15}(1,4\text{-}O_3PC_8H_8PO_3)] \cdot H_2O$ (**3**·H_2O).

Identification code	$[Co(terpy)_2](H_3O)[Mo_5O_{15}(1,4\text{-}O_3PC_8H_8PO_3)] \cdot H_2O$	
Chemical formula	$C_{38}H_{35}CoMo_5N_6O_{23}P_2$	
Formula weight	1544.29	
Temperature	90(2) K	
Wavelength	0.71073 Å	
Crystal size	0.059 x 0.099 x 0.291 mm	
Crystal system	Orthorhombic	
Space group	P n a 21	
Unit cell dimensions	a = 23.327(3) Å	α = 90°
	b = 21.115(3) Å	β = 90°
	c = 9.7462(14) Å	γ = 90°
Volume	4800.5(11) $Å^3$	
Z	4	
Density (calculated)	2.137 g/cm^3	
Absorption coefficient	1.763 mm^{-1}	
F(000)	3024	

Table S18. Data collection and structure refinement for $[Co(terpy)_2](H_3O)[Mo_5O_{15}(1,4\text{-}O_3PC_8H_8PO_3)]\cdot H_2O$ (**3**·H_2O).

Theta range for data collection	2.00 to 27.12°	
Index ranges	-29<=h<=29, -27<=k<=27, -12<=l<=12	
Reflections collected	56796	
Independent reflections	10565 [R(int) = 0.0631]	
Coverage of independent reflections	99.7%	
Absorption correction	multi-scan	
Max. and min. transmission	0.9030 and 0.6280	
Refinement method	Full-matrix least-squares on F^2	
Refinement program	SHELXL-2013 (Sheldrick, 2013)	
Function minimized	$\Sigma\, w(F_o^2 - F_c^2)^2$	
Data / restraints / parameters	10565 / 1 / 677	
Goodness-of-fit on F^2	1.030	
Δ/σ_{max}	0.003	
Final R indices	9133 data; I>2σ(I)	R1 = 0.0395, wR2 = 0.0864
	all data	R1 = 0.0494, wR2 = 0.0903
Weighting scheme	$w=1/[\sigma^2(F_o^2)+(0.0484P)^2+0.5799P]$ where $P=(F_o^2+2F_c^2)/3$	
Absolute structure parameter	0.5(0)	
Largest diff. peak and hole	1.212 and -0.970 eÅ^{-3}	
R.M.S. deviation from mean	0.126 eÅ^{-3}	

Table S19. Atomic coordinates and equivalent isotropic atomic displacement parameters ($Å^2$) for $[Co(terpy)_2](H_3O)[Mo_5O_{15}(1,4\text{-}O_3PC_8H_8PO_3)]\cdot H_2O$ (**3**·H_2O).

U(eq) is defined as one third of the trace of the orthogonalized U_{ij} tensor.

	x/a	**y/b**	**z/c**	**U(eq)**
Mo1	0.75696(3)	0.92314(4)	0.32527(8)	0.01853(18)
Mo2	0.61693(3)	0.93485(4)	0.23037(7)	0.01616(17)
Mo3	0.61298(3)	0.07301(4)	0.99838(7)	0.01656(17)
Mo4	0.75036(3)	0.09118(4)	0.89464(8)	0.01780(17)
Mo5	0.83816(3)	0.00636(4)	0.10279(10)	0.01834(16)
Co1	0.49721(5)	0.26827(4)	0.36109(15)	0.0127(2)
P1	0.71081(10)	0.07357(10)	0.2376(2)	0.0153(5)
P2	0.78928(10)	0.43757(10)	0.4819(2)	0.0142(5)
O1	0.6986(3)	0.0042(3)	0.2806(6)	0.0142(13)
O2	0.6802(3)	0.8874(3)	0.2978(6)	0.0198(14)
O3	0.7550(3)	0.9340(3)	0.5004(7)	0.0276(16)
O4	0.7927(3)	0.8530(3)	0.3053(7)	0.0245(15)
O5	0.8162(3)	0.9839(3)	0.2878(6)	0.0201(14)
O6	0.5942(3)	0.9653(3)	0.3914(7)	0.0246(14)
O7	0.5651(3)	0.8789(3)	0.2036(7)	0.0233(15)
O8	0.5926(2)	0.0036(3)	0.1123(8)	0.0188(11)
O9	0.5895(3)	0.0428(3)	0.8353(7)	0.0216(14)
O10	0.5607(3)	0.1266(3)	0.0274(7)	0.0267(16)

	x/a	y/b	z/c	U(eq)
O11	0.6551(3)	0.1029(3)	0.1858(6)	0.0181(13)
O12	0.6732(3)	0.1234(3)	0.9288(6)	0.0201(14)
O13	0.7454(3)	0.0814(3)	0.7207(7)	0.0264(16)
O14	0.7858(3)	0.1616(3)	0.9133(7)	0.0268(15)
O15	0.8116(3)	0.0305(3)	0.9223(6)	0.0212(14)
O16	0.7594(3)	0.0756(3)	0.1288(6)	0.0173(14)
O17	0.8828(3)	0.9458(3)	0.0597(8)	0.0308(17)
O18	0.8831(3)	0.0678(3)	0.1388(8)	0.0289(17)
O19	0.7403(3)	0.4362(3)	0.5879(7)	0.0186(13)
O20	0.8042(3)	0.5068(3)	0.4405(6)	0.0160(14)
O21	0.8427(3)	0.4062(3)	0.5386(6)	0.0171(13)
C1	0.7342(4)	0.1175(4)	0.3871(9)	0.0203(19)
C2	0.7412(4)	0.1891(4)	0.3697(9)	0.0174(19)
C3	0.7807(4)	0.2152(4)	0.2761(10)	0.021(2)
C4	0.7114(4)	0.2315(5)	0.4493(10)	0.021(2)
C5	0.7892(4)	0.2804(4)	0.2660(10)	0.020(2)
C6	0.7177(4)	0.2974(4)	0.4421(9)	0.0186(19)
C7	0.7576(4)	0.3234(4)	0.3473(9)	0.0197(19)
C8	0.7660(4)	0.3942(4)	0.3322(9)	0.0178(18)
N1	0.4266(3)	0.2770(3)	0.4651(7)	0.0153(16)
N2	0.4940(3)	0.3565(3)	0.3582(9)	0.0160(14)

	x/a	y/b	z/c	U(eq)
N3	0.5671(3)	0.2817(3)	0.2574(7)	0.0136(15)
N4	0.5405(3)	0.2601(3)	0.5308(7)	0.0154(16)
N5	0.4991(3)	0.1802(3)	0.3643(9)	0.0138(12)
N6	0.4545(3)	0.2534(3)	0.1930(7)	0.0149(16)
C9	0.3937(4)	0.2304(4)	0.5168(9)	0.0168(19)
C10	0.3457(4)	0.2427(4)	0.5954(9)	0.0176(18)
C11	0.3306(4)	0.3057(5)	0.6220(9)	0.023(2)
C12	0.3640(4)	0.3531(4)	0.5687(9)	0.020(2)
C13	0.4115(4)	0.3389(4)	0.4883(9)	0.0188(19)
C14	0.4500(4)	0.3850(4)	0.4233(8)	0.0181(19)
C15	0.4465(4)	0.4512(4)	0.4226(10)	0.024(2)
C16	0.4882(4)	0.4842(4)	0.3517(12)	0.032(2)
C17	0.5335(4)	0.4535(4)	0.2847(9)	0.025(2)
C18	0.5356(4)	0.3876(4)	0.2884(9)	0.0172(19)
C19	0.5775(4)	0.3444(4)	0.2269(9)	0.0173(18)
C20	0.6238(4)	0.3620(5)	0.1503(10)	0.024(2)
C21	0.6605(4)	0.3142(4)	0.0970(11)	0.023(2)
C22	0.6022(4)	0.2375(4)	0.2084(9)	0.0148(18)
C23	0.6500(4)	0.2522(4)	0.1257(8)	0.019(2)
C24	0.5623(4)	0.3065(5)	0.6070(10)	0.025(2)
C25	0.5928(4)	0.2942(5)	0.7268(11)	0.029(2)

	x/a	y/b	z/c	U(eq)
C26	0.6007(5)	0.2322(5)	0.7678(11)	0.034(3)
C27	0.5788(5)	0.1841(6)	0.6879(12)	0.036(3)
C28	0.5486(4)	0.1986(4)	0.5701(9)	0.0173(19)
C29	0.5244(4)	0.1518(4)	0.4728(10)	0.0189(19)
C30	0.5247(4)	0.0867(4)	0.4812(12)	0.025(2)
C31	0.4998(4)	0.0526(4)	0.3756(14)	0.030(2)
C32	0.4733(4)	0.0819(4)	0.2656(12)	0.025(2)
C33	0.4739(4)	0.1479(4)	0.2621(10)	0.0168(19)
C34	0.4487(4)	0.1912(4)	0.1612(10)	0.018(2)
C35	0.4200(4)	0.1717(5)	0.0411(9)	0.024(2)
C36	0.3966(5)	0.2173(5)	0.9578(10)	0.032(3)
C37	0.4032(4)	0.2807(5)	0.9894(11)	0.031(2)
C38	0.4325(4)	0.2980(4)	0.1093(10)	0.021(2)
O90	0.6491(3)	0.0016(5)	0.6209(11)	0.062(2)
O91	0.8432(3)	0.0018(3)	0.6504(6)	0.0317(16)

Table S20. Bond lengths (Å) for $[Co(terpy)_2](H_3O)[Mo_5O_{15}(1,4\text{-}O_3PC_8H_8PO_3)] \cdot H_2O$ (**3**·H_2O).

Mo1-O4	1.710(6)	Mo1-O3	1.723(7)
Mo1-O5	1.920(7)	Mo1-O2	1.962(6)
Mo1-O1	2.230(6)	Mo1-O19	2.331(6)
Mo2-O7	1.711(6)	Mo2-O6	1.777(6)
Mo2-O2	1.900(6)	Mo2-O8	1.937(7)
Mo2-O21	2.179(6)	Mo2-O1	2.453(6)
Mo3-O10	1.688(7)	Mo3-O9	1.798(6)
Mo3-O12	1.888(6)	Mo3-O8	1.900(7)
Mo3-O11	2.169(6)	Mo3-O20	2.450(6)
Mo4-O14	1.711(6)	Mo4-O13	1.712(7)
Mo4-O15	1.938(7)	Mo4-O12	1.954(6)
Mo4-O20	2.236(6)	Mo4-O16	2.316(6)
Mo5-O17	1.700(7)	Mo5-O18	1.704(6)
Mo5-O5	1.934(6)	Mo5-O15	1.933(6)
Mo5-O19	2.358(6)	Mo5-O16	2.362(7)
Co1-N5	1.860(6)	Co1-N2	1.864(6)
Co1-N3	1.938(7)	Co1-N1	1.942(8)
Co1-N6	1.943(7)	Co1-N4	1.946(7)
P1-O11	1.525(6)	P1-O1	1.550(6)
P1-O16	1.553(7)	P1-C1	1.812(9)
P2-O21	1.515(6)	P2-O19	1.541(7)

P2-O20	1.555(6)	P2-C8	1.806(9)
O19-Mo1	2.331(6)	O19-Mo5	2.358(6)
O20-Mo4	2.236(6)	O20-Mo3	2.451(6)
O21-Mo2	2.179(6)	C1-C2	1.529(12)
C1-H1A	0.97	C1-H1B	0.97
C2-C4	1.373(12)	C2-C3	1.409(13)
C3-C5	1.395(12)	C3-H3	0.93
C4-C6	1.401(13)	C4-H4	0.93
C5-C7	1.412(12)	C5-H5	0.93
C6-C7	1.422(12)	C6-H6	0.93
C7-C8	1.516(12)	C8-H8A	0.97
C8-H8B	0.97	N1-C9	1.346(11)
N1-C13	1.373(11)	N2-C14	1.349(11)
N2-C18	1.354(12)	N3-C22	1.329(11)
N3-C19	1.379(11)	N4-C24	1.330(12)
N4-C28	1.367(11)	N5-C33	1.343(12)
N5-C29	1.352(12)	N6-C38	1.347(11)
N6-C34	1.355(11)	C9-C10	1.383(13)
C9-H9	0.93	C10-C11	1.401(12)
C10-H10	0.93	C11-C12	1.369(13)
C11-H11	0.93	C12-C13	1.389(12)
C12-H12	0.93	C13-C14	1.466(12)

C14-C15	1.400(12)	C15-C16	1.382(13)
C15-H15	0.93	C16-C17	1.401(14)
C16-H16	0.93	C17-C18	1.393(12)
C17-H17	0.93	C18-C19	1.466(12)
C19-C20	1.364(13)	C20-C21	1.422(13)
C20-H20	0.93	C21-C23	1.362(12)
C21-H21	0.93	C22-C23	1.411(12)
C22-H22	0.93	C23-H23	0.93
C24-C25	1.392(13)	C24-H24	0.93
C25-C26	1.381(15)	C25-H25	0.93
C26-C27	1.378(16)	C26-H26	0.93
C27-C28	1.383(14)	C27-H27	0.93
C28-C29	1.481(13)	C29-C30	1.377(13)
C30-C31	1.384(16)	C30-H30	0.93
C31-C32	1.383(16)	C31-H31	0.93
C32-C33	1.395(12)	C32-H32	0.93
C33-C34	1.465(13)	C34-C35	1.410(13)
C35-C36	1.374(14)	C35-H35	0.93
C36-C37	1.382(15)	C36-H36	0.93
C37-C38	1.402(13)	C37-H37	0.93
C38-H38	0.93	O90-H90A	0.8488
O90-H90B	0.8575		

Table S21. Bond angles (°) for $[Co(terpy)_2](H_3O)[Mo_5O_{15}(1,4\text{-}O_3PC_8H_8PO_3)] \cdot H_2O$ (**3**·H_2O).

O4-Mo1-O3	103.9(3)	O4-Mo1-O5	101.9(3)
O3-Mo1-O5	96.8(3)	O4-Mo1-O2	95.6(3)
O3-Mo1-O2	99.4(3)	O5-Mo1-O2	152.6(3)
O4-Mo1-O1	160.0(3)	O3-Mo1-O1	94.3(3)
O5-Mo1-O1	83.6(3)	O2-Mo1-O1	73.2(2)
O4-Mo1-O19	88.6(3)	O3-Mo1-O19	165.5(3)
O5-Mo1-O19	73.3(2)	O2-Mo1-O19	86.3(2)
O1-Mo1-O19	74.4(2)	O7-Mo2-O6	100.0(3)
O7-Mo2-O2	103.8(3)	O6-Mo2-O2	96.7(3)
O7-Mo2-O8	102.7(3)	O6-Mo2-O8	99.6(3)
O2-Mo2-O8	145.9(2)	O7-Mo2-O21	89.0(3)
O6-Mo2-O21	170.8(3)	O2-Mo2-O21	79.3(2)
O8-Mo2-O21	79.9(3)	O7-Mo2-O1	172.8(3)
O6-Mo2-O1	80.7(3)	O2-Mo2-O1	69.1(2)
O8-Mo2-O1	84.2(2)	O21-Mo2-O1	90.0(2)
O10-Mo3-O9	99.6(3)	O10-Mo3-O12	102.6(3)
O9-Mo3-O12	96.3(3)	O10-Mo3-O8	103.8(3)
O9-Mo3-O8	99.6(3)	O12-Mo3-O8	146.2(2)
O10-Mo3-O11	89.5(3)	O9-Mo3-O11	170.4(3)
O12-Mo3-O11	78.5(2)	O8-Mo3-O11	81.1(3)
O10-Mo3-O20	172.2(3)	O9-Mo3-O20	80.4(2)

O12-Mo3-O20	69.7(2)	O8-Mo3-O20	83.8(2)
O11-Mo3-O20	90.1(2)	O14-Mo4-O13	104.0(3)
O14-Mo4-O15	101.7(3)	O13-Mo4-O15	96.2(3)
O14-Mo4-O12	97.1(3)	O13-Mo4-O12	98.6(3)
O15-Mo4-O12	152.4(3)	O14-Mo4-O20	161.2(3)
O13-Mo4-O20	93.7(3)	O15-Mo4-O20	82.3(2)
O12-Mo4-O20	73.7(2)	O14-Mo4-O16	88.5(3)
O13-Mo4-O16	164.9(3)	O15-Mo4-O16	72.6(2)
O12-Mo4-O16	88.0(2)	O20-Mo4-O16	75.0(2)
O17-Mo5-O18	104.3(3)	O17-Mo5-O5	102.0(3)
O18-Mo5-O5	99.1(3)	O17-Mo5-O15	99.8(3)
O18-Mo5-O15	100.6(3)	O5-Mo5-O15	145.9(2)
O17-Mo5-O19	89.3(3)	O18-Mo5-O19	165.4(3)
O5-Mo5-O19	72.4(2)	O15-Mo5-O19	82.0(2)
O17-Mo5-O16	165.4(3)	O18-Mo5-O16	89.1(3)
O5-Mo5-O16	81.1(2)	O15-Mo5-O16	71.6(2)
O19-Mo5-O16	78.00(17)	N5-Co1-N2	179.1(3)
N5-Co1-N3	97.7(3)	N2-Co1-N3	83.1(3)
N5-Co1-N1	96.1(3)	N2-Co1-N1	83.1(3)
N3-Co1-N1	166.2(3)	N5-Co1-N6	82.2(3)
N2-Co1-N6	97.4(3)	N3-Co1-N6	90.9(3)
N1-Co1-N6	91.2(3)	N5-Co1-N4	83.4(3)

N2-Co1-N4	97.0(3)	N3-Co1-N4	91.1(3)
N1-Co1-N4	90.3(3)	N6-Co1-N4	165.6(3)
O11-P1-O1	108.5(4)	O11-P1-O16	112.6(4)
O1-P1-O16	110.2(3)	O11-P1-C1	108.3(4)
O1-P1-C1	108.8(4)	O16-P1-C1	108.4(4)
O21-P2-O19	111.0(4)	O21-P2-O20	108.7(4)
O19-P2-O20	110.9(3)	O21-P2-C8	108.7(4)
O19-P2-C8	108.0(4)	O20-P2-C8	109.5(4)
P1-O1-Mo1	131.8(4)	P1-O1-Mo2	130.8(3)
Mo1-O1-Mo2	93.14(19)	Mo2-O2-Mo1	123.6(3)
Mo1-O5-Mo5	122.2(3)	Mo3-O8-Mo2	148.4(3)
P1-O11-Mo3	123.1(4)	Mo3-O12-Mo4	123.4(3)
Mo5-O15-Mo4	122.5(3)	P1-O16-Mo4	127.6(4)
P1-O16-Mo5	128.6(4)	Mo4-O16-Mo5	93.0(2)
P2-O19-Mo1	130.3(4)	P2-O19-Mo5	127.3(4)
Mo1-O19-Mo5	92.0(2)	P2-O20-Mo4	132.3(4)
P2-O20-Mo3	130.8(3)	Mo4-O20-Mo3	92.3(2)
P2-O21-Mo2	123.2(3)	C2-C1-P1	116.7(6)
C2-C1-H1A	108.1	P1-C1-H1A	108.1
C2-C1-H1B	108.1	P1-C1-H1B	108.1
H1A-C1-H1B	107.3	C4-C2-C3	116.2(8)
C4-C2-C1	121.8(8)	C3-C2-C1	121.9(8)

C5-C3-C2	121.7(8)	C5-C3-H3	119.2
C2-C3-H3	119.2	C2-C4-C6	124.5(9)
C2-C4-H4	117.8	C6-C4-H4	117.8
C3-C5-C7	121.4(9)	C3-C5-H5	119.3
C7-C5-H5	119.3	C4-C6-C7	119.0(8)
C4-C6-H6	120.5	C7-C6-H6	120.5
C5-C7-C6	117.3(8)	C5-C7-C8	120.8(8)
C6-C7-C8	121.9(8)	C7-C8-P2	117.4(6)
C7-C8-H8A	107.9	P2-C8-H8A	107.9
C7-C8-H8B	107.9	P2-C8-H8B	107.9
H8A-C8-H8B	107.2	C9-N1-C13	119.2(8)
C9-N1-Co1	127.5(6)	C13-N1-Co1	113.2(6)
C14-N2-C18	124.4(7)	C14-N2-Co1	117.9(6)
C18-N2-Co1	117.6(6)	C22-N3-C19	119.2(8)
C22-N3-Co1	127.1(6)	C19-N3-Co1	113.6(6)
C24-N4-C28	119.4(8)	C24-N4-Co1	127.4(6)
C28-N4-Co1	113.2(6)	C33-N5-C29	123.1(6)
C33-N5-Co1	119.0(6)	C29-N5-Co1	117.9(6)
C38-N6-C34	120.0(8)	C38-N6-Co1	126.3(6)
C34-N6-Co1	113.6(6)	N1-C9-C10	122.2(8)
N1-C9-H9	118.9	C10-C9-H9	118.9
C9-C10-C11	118.9(9)	C9-C10-H10	120.5

C11-C10-H10	120.5	C12-C11-C10	118.7(9)
C12-C11-H11	120.6	C10-C11-H11	120.6
C11-C12-C13	120.7(9)	C11-C12-H12	119.6
C13-C12-H12	119.6	N1-C13-C12	120.1(8)
N1-C13-C14	113.8(8)	C12-C13-C14	126.1(8)
N2-C14-C15	119.1(8)	N2-C14-C13	111.9(7)
C15-C14-C13	129.0(8)	C16-C15-C14	117.8(9)
C16-C15-H15	121.1	C14-C15-H15	121.1
C15-C16-C17	122.0(8)	C15-C16-H16	119.0
C17-C16-H16	119.0	C18-C17-C16	118.4(9)
C18-C17-H17	120.8	C16-C17-H17	120.8
N2-C18-C17	118.3(8)	N2-C18-C19	112.4(8)
C17-C18-C19	129.3(8)	C20-C19-N3	121.3(8)
C20-C19-C18	125.6(8)	N3-C19-C18	113.1(8)
C19-C20-C21	118.8(9)	C19-C20-H20	120.6
C21-C20-H20	120.6	C23-C21-C20	120.0(9)
C23-C21-H21	120.0	C20-C21-H21	120.0
N3-C22-C23	122.6(8)	N3-C22-H22	118.7
C23-C22-H22	118.7	C21-C23-C22	118.0(9)
C21-C23-H23	121.0	C22-C23-H23	121.0
N4-C24-C25	121.7(9)	N4-C24-H24	119.1
C25-C24-H24	119.1	C26-C25-C24	119.2(9)

C26-C25-H25	120.4	C24-C25-H25	120.4
C27-C26-C25	119.1(10)	C27-C26-H26	120.5
C25-C26-H26	120.5	C26-C27-C28	119.6(10)
C26-C27-H27	120.2	C28-C27-H27	120.2
N4-C28-C27	120.9(9)	N4-C28-C29	113.8(7)
C27-C28-C29	125.3(9)	N5-C29-C30	119.5(9)
N5-C29-C28	111.7(7)	C30-C29-C28	128.8(9)
C29-C30-C31	118.2(9)	C29-C30-H30	120.9
C31-C30-H30	120.9	C30-C31-C32	122.1(8)
C30-C31-H31	118.9	C32-C31-H31	118.9
C31-C32-C33	117.5(9)	C31-C32-H32	121.2
C33-C32-H32	121.2	N5-C33-C32	119.6(9)
N5-C33-C34	110.9(7)	C32-C33-C34	129.5(9)
N6-C34-C35	121.4(9)	N6-C34-C33	114.3(8)
C35-C34-C33	124.4(8)	C36-C35-C34	118.3(9)
C36-C35-H35	120.9	C34-C35-H35	120.9
C35-C36-C37	120.3(9)	C35-C36-H36	119.9
C37-C36-H36	119.9	C36-C37-C38	119.4(10)
C36-C37-H37	120.3	C38-C37-H37	120.3
N6-C38-C37	120.6(9)	N6-C38-H38	119.7
C37-C38-H38	119.7	H90A-O90-H90B	110.4

Table S22. Torsion angles (°) for $[Co(terpy)_2](H_3O)[Mo_5O_{15}(1,4\text{-}O_3PC_8H_8PO_3)] \cdot H_2O$ (**3**·H_2O).

O11-P1-O1-Mo1	165.7(4)	O16-P1-O1-Mo1	41.9(6)
C1-P1-O1-Mo1	-76.7(5)	O11-P1-O1-Mo2	15.5(5)
O16-P1-O1-Mo2	-108.2(5)	C1-P1-O1-Mo2	133.1(5)
O7-Mo2-O2-Mo1	171.6(4)	O6-Mo2-O2-Mo1	-86.3(4)
O8-Mo2-O2-Mo1	31.8(7)	O21-Mo2-O2-Mo1	85.2(4)
O1-Mo2-O2-Mo1	-9.0(3)	O1-P1-O11-Mo3	-67.5(5)
O16-P1-O11-Mo3	54.8(5)	C1-P1-O11-Mo3	174.6(4)
O10-Mo3-O12-Mo4	172.4(4)	O9-Mo3-O12-Mo4	-86.3(4)
O8-Mo3-O12-Mo4	31.5(7)	O11-Mo3-O12-Mo4	85.6(4)
O20-Mo3-O12-Mo4	-8.9(3)	O11-P1-O16-Mo4	-17.8(5)
O1-P1-O16-Mo4	103.5(4)	C1-P1-O16-Mo4	-137.6(4)
O11-P1-O16-Mo5	-152.3(4)	O1-P1-O16-Mo5	-31.0(5)
C1-P1-O16-Mo5	87.9(5)	O21-P2-O19-Mo1	20.3(6)
O20-P2-O19-Mo1	-100.7(5)	C8-P2-O19-Mo1	139.3(4)
O21-P2-O19-Mo5	154.9(4)	O20-P2-O19-Mo5	34.0(5)
C8-P2-O19-Mo5	-86.0(5)	O21-P2-O20-Mo4	-164.1(4)
O19-P2-O20-Mo4	-41.8(6)	C8-P2-O20-Mo4	77.2(5)
O21-P2-O20-Mo3	-15.2(5)	O19-P2-O20-Mo3	107.1(5)
C8-P2-O20-Mo3	-133.8(5)	O19-P2-O21-Mo2	-55.3(5)
O20-P2-O21-Mo2	67.0(5)	C8-P2-O21-Mo2	-173.9(4)
O11-P1-C1-C2	-54.9(8)	O1-P1-C1-C2	-172.6(6)

O16-P1-C1-C2	67.6(8)	P1-C1-C2-C4	121.7(8)
P1-C1-C2-C3	-61.6(10)	C4-C2-C3-C5	0.4(12)
C1-C2-C3-C5	-176.4(8)	C3-C2-C4-C6	0.4(13)
C1-C2-C4-C6	177.3(8)	C2-C3-C5-C7	-1.3(13)
C2-C4-C6-C7	-0.4(13)	C3-C5-C7-C6	1.3(13)
C3-C5-C7-C8	-178.2(8)	C4-C6-C7-C5	-0.5(12)
C4-C6-C7-C8	179.0(8)	C5-C7-C8-P2	-120.4(8)
C6-C7-C8-P2	60.1(10)	O21-P2-C8-C7	52.8(7)
O19-P2-C8-C7	-67.7(7)	O20-P2-C8-C7	171.4(6)
N3-Co1-N2-C14	-178.9(7)	N1-Co1-N2-C14	0.8(7)
N6-Co1-N2-C14	91.1(7)	N4-Co1-N2-C14	-88.6(7)
N3-Co1-N2-C18	2.4(7)	N1-Co1-N2-C18	-177.8(7)
N6-Co1-N2-C18	-87.6(7)	N4-Co1-N2-C18	92.7(7)
N3-Co1-N5-C33	-89.4(7)	N1-Co1-N5-C33	90.8(7)
N6-Co1-N5-C33	0.4(7)	N4-Co1-N5-C33	-179.6(7)
N3-Co1-N5-C29	92.8(7)	N1-Co1-N5-C29	-87.0(7)
N6-Co1-N5-C29	-177.4(7)	N4-Co1-N5-C29	2.6(7)
C13-N1-C9-C10	-1.4(13)	Co1-N1-C9-C10	176.7(6)
N1-C9-C10-C11	0.1(13)	C9-C10-C11-C12	0.2(13)
C10-C11-C12-C13	0.8(14)	C9-N1-C13-C12	2.4(13)
Co1-N1-C13-C12	-175.9(7)	C9-N1-C13-C14	-178.4(7)
Co1-N1-C13-C14	3.3(9)	C11-C12-C13-N1	-2.1(14)

C11-C12-C13-C14	178.7(9)	C18-N2-C14-C15	-1.3(14)
Co1-N2-C14-C15	-179.9(6)	C18-N2-C14-C13	179.3(8)
Co1-N2-C14-C13	0.7(10)	N1-C13-C14-N2	-2.6(11)
C12-C13-C14-N2	176.5(9)	N1-C13-C14-C15	178.1(8)
C12-C13-C14-C15	-2.7(15)	N2-C14-C15-C16	1.5(13)
C13-C14-C15-C16	-179.3(9)	C14-C15-C16-C17	-1.4(15)
C15-C16-C17-C18	1.1(15)	C14-N2-C18-C17	1.0(14)
Co1-N2-C18-C17	179.6(6)	C14-N2-C18-C19	-179.3(8)
Co1-N2-C18-C19	-0.7(10)	C16-C17-C18-N2	-0.8(13)
C16-C17-C18-C19	179.6(9)	C22-N3-C19-C20	-1.5(12)
Co1-N3-C19-C20	-177.6(7)	C22-N3-C19-C18	-179.7(7)
Co1-N3-C19-C18	4.2(9)	N2-C18-C19-C20	179.6(9)
C17-C18-C19-C20	-0.7(15)	N2-C18-C19-N3	-2.3(10)
C17-C18-C19-N3	177.3(9)	N3-C19-C20-C21	2.4(14)
C18-C19-C20-C21	-179.7(9)	C19-C20-C21-C23	-1.7(15)
C19-N3-C22-C23	-0.1(12)	Co1-N3-C22-C23	175.5(6)
C20-C21-C23-C22	0.3(14)	N3-C22-C23-C21	0.7(13)
C28-N4-C24-C25	-0.5(14)	Co1-N4-C24-C25	-179.8(7)
N4-C24-C25-C26	0.0(16)	C24-C25-C26-C27	0.8(17)
C25-C26-C27-C28	-1.1(17)	C24-N4-C28-C27	0.2(14)
Co1-N4-C28-C27	179.6(8)	C24-N4-C28-C29	-177.3(8)
Co1-N4-C28-C29	2.1(10)	C26-C27-C28-N4	0.7(16)

C26-C27-C28-C29	177.9(10)	C33-N5-C29-C30	-0.1(15)
Co1-N5-C29-C30	177.6(7)	C33-N5-C29-C28	-179.7(7)
Co1-N5-C29-C28	-2.1(10)	N4-C28-C29-N5	-0.1(11)
C27-C28-C29-N5	-177.5(9)	N4-C28-C29-C30	-179.7(9)
C27-C28-C29-C30	2.9(16)	N5-C29-C30-C31	1.2(14)
C28-C29-C30-C31	-179.2(9)	C29-C30-C31-C32	-2.0(16)
C30-C31-C32-C33	1.7(16)	C29-N5-C33-C32	-0.3(15)
Co1-N5-C33-C32	-177.9(7)	C29-N5-C33-C34	178.2(7)
Co1-N5-C33-C34	0.6(10)	C31-C32-C33-N5	-0.5(14)
C31-C32-C33-C34	-178.7(9)	C38-N6-C34-C35	0.2(13)
Co1-N6-C34-C35	-178.6(7)	C38-N6-C34-C33	-179.2(8)
Co1-N6-C34-C33	2.1(10)	N5-C33-C34-N6	-1.7(11)
C32-C33-C34-N6	176.6(9)	N5-C33-C34-C35	178.9(9)
C32-C33-C34-C35	-2.8(16)	N6-C34-C35-C36	-1.6(14)
C33-C34-C35-C36	177.7(9)	C34-C35-C36-C37	2.4(15)
C35-C36-C37-C38	-1.9(16)	C34-N6-C38-C37	0.4(13)
Co1-N6-C38-C37	179.0(7)	C36-C37-C38-N6	0.5(15)

Table S23. Anisotropic atomic displacement parameters ($Å^2$) for $[Co(terpy)_2](H_3O)[Mo_5O_{15}(1,4\text{-}O_3PC_8H_8PO_3)] \cdot H_2O$ (**3**·H_2O).

The anisotropic atomic displacement factor exponent takes the form: $-2\pi^2[h^2 a^{*2} U_{11} + ... + 2 h k a^* b^* U_{12}]$

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
Mo1	0.0209(4)	0.0205(4)	0.0142(4)	0.0043(3)	0.0020(3)	0.0080(3)
Mo2	0.0159(4)	0.0157(4)	0.0170(4)	0.0009(3)	0.0026(3)	0.0008(3)
Mo3	0.0177(4)	0.0145(4)	0.0174(4)	0.0010(3)	-0.0032(3)	-0.0007(3)
Mo4	0.0204(4)	0.0194(4)	0.0135(3)	0.0034(3)	-0.0021(3)	-0.0081(3)
Mo5	0.0147(3)	0.0219(4)	0.0185(3)	0.0013(3)	-0.0013(4)	-0.0013(4)
Co1	0.0166(5)	0.0088(5)	0.0127(4)	-0.0002(4)	-0.0049(3)	0.0009(5)
P1	0.0194(12)	0.0123(11)	0.0141(10)	-0.0011(9)	-0.0018(9)	-0.0005(9)
P2	0.0165(12)	0.0131(11)	0.0129(10)	-0.0005(9)	-0.0038(9)	0.0001(9)
O1	0.016(3)	0.011(3)	0.015(3)	0.001(2)	-0.001(2)	0.002(2)
O2	0.028(4)	0.009(3)	0.022(3)	0.002(2)	0.003(3)	0.004(2)
O3	0.031(4)	0.037(4)	0.015(3)	0.005(3)	0.004(3)	0.012(3)
O4	0.026(3)	0.023(4)	0.025(3)	0.005(3)	0.005(3)	0.015(3)
O5	0.020(3)	0.025(4)	0.015(3)	-0.003(3)	-0.005(2)	0.000(3)
O6	0.027(4)	0.029(4)	0.018(3)	-0.002(3)	0.005(3)	0.006(3)
O7	0.021(4)	0.021(4)	0.028(4)	-0.001(3)	0.005(3)	-0.003(3)
O8	0.016(2)	0.019(3)	0.022(2)	-0.001(2)	0.002(3)	0.005(3)
O9	0.020(3)	0.020(3)	0.025(3)	0.000(3)	-0.007(3)	-0.003(3)
O10	0.029(4)	0.025(4)	0.027(4)	0.008(3)	-0.006(3)	0.001(3)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
O11	0.022(3)	0.015(3)	0.017(3)	-0.001(2)	-0.003(3)	0.004(3)
O12	0.019(3)	0.020(3)	0.022(3)	0.003(3)	-0.006(3)	-0.005(3)
O13	0.033(4)	0.029(4)	0.017(3)	0.007(3)	-0.005(3)	-0.012(3)
O14	0.029(4)	0.027(4)	0.025(3)	0.008(3)	-0.002(3)	-0.012(3)
O15	0.018(3)	0.026(4)	0.020(3)	0.002(3)	0.008(3)	-0.009(3)
O16	0.021(3)	0.018(3)	0.013(3)	-0.002(2)	-0.004(2)	-0.008(3)
O17	0.021(4)	0.037(4)	0.034(4)	0.008(3)	0.006(3)	0.003(3)
O18	0.027(4)	0.028(4)	0.032(4)	0.002(3)	-0.009(3)	-0.011(3)
O19	0.020(3)	0.021(3)	0.015(3)	0.001(3)	0.001(3)	-0.003(3)
O20	0.016(3)	0.014(3)	0.018(3)	-0.001(2)	0.000(2)	0.004(2)
O21	0.020(3)	0.013(3)	0.018(3)	-0.001(2)	-0.008(2)	0.001(3)
C1	0.025(5)	0.021(5)	0.015(4)	-0.003(4)	-0.007(4)	-0.004(4)
C2	0.018(4)	0.022(5)	0.012(4)	-0.002(3)	-0.008(3)	0.000(4)
C3	0.022(5)	0.017(5)	0.023(5)	-0.016(4)	-0.006(4)	0.001(4)
C4	0.018(5)	0.028(5)	0.017(4)	-0.003(4)	-0.001(4)	-0.003(4)
C5	0.014(4)	0.024(5)	0.020(5)	-0.001(4)	-0.002(4)	-0.007(4)
C6	0.020(5)	0.022(5)	0.013(4)	0.001(3)	-0.001(3)	-0.002(4)
C7	0.014(4)	0.023(5)	0.021(5)	-0.012(4)	0.002(4)	-0.005(4)
C8	0.019(4)	0.024(5)	0.011(4)	-0.001(4)	0.004(4)	0.000(4)
N1	0.018(4)	0.010(4)	0.017(4)	-0.003(3)	-0.007(3)	0.007(3)
N2	0.022(4)	0.012(3)	0.015(3)	-0.001(3)	-0.004(2)	-0.003(3)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
N3	0.016(4)	0.016(4)	0.009(3)	0.003(3)	-0.005(3)	0.000(3)
N4	0.016(4)	0.013(4)	0.017(4)	0.007(3)	-0.007(3)	-0.008(3)
N5	0.012(3)	0.009(3)	0.020(3)	0.003(3)	-0.001(2)	-0.003(3)
N6	0.015(4)	0.019(4)	0.011(3)	0.000(3)	-0.002(3)	0.004(3)
C9	0.024(5)	0.008(4)	0.018(4)	0.000(3)	-0.009(4)	0.000(3)
C10	0.028(5)	0.010(4)	0.015(4)	-0.002(4)	-0.003(4)	0.000(4)
C11	0.020(5)	0.035(6)	0.014(5)	-0.002(4)	-0.005(3)	0.003(4)
C12	0.032(5)	0.013(4)	0.016(4)	-0.004(3)	0.000(4)	0.007(4)
C13	0.027(5)	0.014(4)	0.016(4)	-0.001(4)	-0.002(4)	0.001(4)
C14	0.024(5)	0.021(5)	0.009(4)	-0.006(3)	-0.004(3)	0.002(4)
C15	0.033(6)	0.013(4)	0.027(5)	0.000(4)	0.004(4)	0.003(4)
C16	0.044(7)	0.010(4)	0.042(5)	-0.001(4)	0.002(5)	0.003(4)
C17	0.035(6)	0.013(5)	0.027(5)	0.004(4)	0.009(4)	-0.001(4)
C18	0.022(5)	0.014(4)	0.016(4)	0.000(3)	-0.005(3)	0.005(4)
C19	0.027(5)	0.013(4)	0.012(4)	0.000(3)	-0.004(4)	0.000(4)
C20	0.035(6)	0.018(5)	0.019(4)	0.005(4)	0.004(4)	-0.003(4)
C21	0.019(5)	0.030(5)	0.021(4)	0.003(4)	0.003(4)	-0.010(4)
C22	0.016(4)	0.015(4)	0.014(4)	0.001(3)	-0.003(3)	0.000(3)
C23	0.019(5)	0.022(5)	0.015(4)	-0.002(4)	-0.006(3)	-0.003(4)
C24	0.028(5)	0.031(6)	0.015(4)	0.005(4)	-0.002(4)	-0.010(4)
C25	0.030(6)	0.036(6)	0.023(5)	0.001(5)	-0.015(4)	-0.009(4)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
C26	0.037(6)	0.041(7)	0.024(5)	-0.003(5)	-0.014(4)	0.004(5)
C27	0.028(6)	0.035(6)	0.044(7)	0.016(5)	-0.009(5)	0.004(5)
C28	0.014(4)	0.023(5)	0.016(5)	0.010(4)	-0.004(3)	-0.001(4)
C29	0.007(4)	0.020(5)	0.030(5)	0.007(4)	0.002(3)	0.003(4)
C30	0.008(4)	0.023(5)	0.043(6)	0.010(4)	0.001(4)	-0.006(4)
C31	0.019(4)	0.007(4)	0.063(7)	0.006(5)	0.010(4)	-0.003(4)
C32	0.020(5)	0.014(5)	0.042(6)	-0.012(4)	0.010(4)	-0.005(4)
C33	0.011(4)	0.010(4)	0.030(5)	-0.012(4)	0.004(4)	0.001(3)
C34	0.014(5)	0.015(5)	0.025(5)	-0.006(4)	-0.002(4)	0.003(4)
C35	0.025(5)	0.028(5)	0.019(5)	-0.014(4)	-0.003(4)	0.000(4)
C36	0.030(6)	0.042(7)	0.024(5)	-0.016(5)	-0.008(4)	0.003(5)
C37	0.022(5)	0.052(7)	0.020(5)	0.002(5)	-0.005(4)	0.016(5)
C38	0.022(5)	0.018(5)	0.023(5)	-0.001(4)	-0.006(4)	0.006(4)
O90	0.032(4)	0.108(7)	0.046(5)	-0.027(4)	-0.003(5)	-0.012(5)
O91	0.034(4)	0.025(4)	0.036(4)	0.006(3)	0.005(3)	0.002(3)

Table S24. Hydrogen atomic coordinates and isotropic atomic displacement parameters ($Å^2$) for $[Co(terpy)_2](H_3O)[Mo_5O_{15}(1,4\text{-}O_3PC_8H_8PO_3)]\cdot H_2O$ (**3**·H_2O).

	x/a	y/b	z/c	U(eq)
H1A	0.7068	0.1101	0.4603	0.024
H1B	0.7707	0.1002	0.4168	0.024
H3	0.8016	0.1883	0.2197	0.025
H4	0.6852	0.2153	0.5122	0.025
H5	0.8162	0.2959	0.2044	0.024
H6	0.6961	0.3237	0.4984	0.022
H8A	0.7300	0.4124	0.3017	0.021
H8B	0.7938	0.4013	0.2599	0.021
H9	0.4037	0.1885	0.4990	0.02
H10	0.3237	0.2096	0.6301	0.021
H11	0.2986	0.3153	0.6748	0.027
H12	0.3548	0.3951	0.5865	0.024
H15	0.4172	0.4722	0.4682	0.029
H16	0.4862	0.5281	0.3484	0.038
H17	0.5615	0.4766	0.2387	0.03
H20	0.6312	0.4046	0.1332	0.028
H21	0.6916	0.3253	0.0425	0.028
H22	0.5949	0.1953	0.2294	0.018
H23	0.6737	0.2204	0.0919	0.022

	x/a	y/b	z/c	U(eq)
H24	0.5570	0.3483	0.5795	0.029
H25	0.6076	0.3273	0.7787	0.035
H26	0.6205	0.2230	0.8482	0.041
H27	0.5843	0.1420	0.7132	0.043
H30	0.5412	0.0662	0.5558	0.03
H31	0.5008	0.0086	0.3788	0.036
H32	0.4559	0.0584	0.1965	0.031
H35	0.4170	0.1290	0.0189	0.029
H36	0.3762	0.2055	-0.1202	0.038
H37	0.3883	0.3117	-0.0684	0.037
H38	0.4368	0.3406	0.1313	0.025
H90A	0.6269	0.0011	0.5519	0.093
H90B	0.6829	0.0131	0.5969	0.093

Table S25. Sample and crystal data for $[Co(2,2'\text{-bpy})_3](H_3O)[Mo_5O_{15}(1,4\text{-}O_3PC_8H_8O_3P)] \cdot 6H_2O$ (**4**·$6H_2O$).

Identification code	$[Co(2,2'\text{-bpy})_3] (H_3O)[Mo_5O_{15}(1,4\text{-}O_3PC_8H_8O_3P)] \cdot 6H_2O$	
Chemical formula	$C_{38}H_{47}CoMo_5N_6O_{28}P_2$	
Formula weight	1636.38	
Temperature	90(2) K	
Wavelength	0.71073 Å	
Crystal system	Orthorhombic	
Space group	P 21 21 21	
Unit cell dimensions	a = 14.574(3) Å	α = 90°
	b = 17.358(3) Å	β = 90°
	c = 20.413(4) Å	γ = 90°
Volume	5164.0(16) $Å^3$	
Z	4	
Density (calculated)	2.105 g/cm^3	
Absorption coefficient	1.652 mm^{-1}	
F(000)	3232	

Table S26. Data collection and structure refinement for $[Co(2,2'\text{-bpy})_3](H_3O)[Mo_5O_{15}(1,4\text{-}O_3PC_8H_8O_3P)] \cdot 6H_2O$ (**4**·$6H_2O$).

Theta range for data collection	1.72 to 25.68°	
Index ranges	-17<=h<=17, -21<=k<=21, -24<=l<=24	
Reflections collected	54251	
Independent reflections	9786 [R(int) = 0.0895]	
Refinement method	Full-matrix least-squares on F^2	
Refinement program	SHELXL-2013 (Sheldrick, 2013)	
Function minimized	$\Sigma\, w(F_o^2 - F_c^2)^2$	
Data / restraints / parameters	9786 / 273 / 757	
Goodness-of-fit on F^2	0.973	
Δ/σ_{max}	0.001	
Final R indices	8527 data; I>2σ(I)	R1 = 0.0377, wR2 = 0.0735
	all data	R1 = 0.0478, wR2 = 0.0766
Weighting scheme	$w=1/[\sigma^2(F_o^2)+(0.0390P)^2]$ where $P=(F_o^2+2F_c^2)/3$	
Absolute structure parameter	0.5(0)	
Largest diff. peak and hole	1.219 and -0.700 $eÅ^{-3}$	
R.M.S. deviation from mean	0.121 $eÅ^{-3}$	

Table S27. Atomic coordinates and equivalent isotropic atomic displacement parameters ($Å^2$) for $[Co(2,2'\text{-bpy})_3](H_3O)[Mo_5O_{15}(1,4\text{-}O_3PC_8H_8O_3P)] \cdot 6H_2O$ (**4**·$6H_2O$).

U(eq) is defined as one third of the trace of the orthogonalized U_{ij} tensor.

	x/a	**y/b**	**z/c**	**U(eq)**
Mo1	0.46730(5)	0.05527(4)	0.04360(3)	0.00945(17)
Mo2	0.41800(5)	0.20117(4)	0.14679(4)	0.01001(17)
Mo3	0.22436(5)	0.16188(4)	0.22870(4)	0.00943(17)
Mo4	0.12277(5)	0.99822(4)	0.15184(3)	0.00906(16)
Mo5	0.31402(5)	0.91297(4)	0.06053(4)	0.00916(17)
Co1	0.77506(8)	0.91804(7)	0.89269(5)	0.0082(2)
P1	0.36256(15)	0.01276(13)	0.19342(10)	0.0085(5)
P2	0.25662(15)	0.88578(12)	0.56068(11)	0.0085(5)
O1	0.4377(4)	0.9494(3)	0.0505(3)	0.0112(12)
O2	0.4729(4)	0.0668(3)	0.9604(3)	0.0146(13)
O3	0.5791(4)	0.0493(3)	0.0668(3)	0.0153(13)
O4	0.4447(4)	0.1638(3)	0.0616(3)	0.0095(12)
O5	0.4252(4)	0.0667(3)	0.1541(3)	0.0090(12)
O6	0.5239(4)	0.2070(4)	0.1821(3)	0.0158(14)
O7	0.3912(4)	0.2943(3)	0.1296(3)	0.0154(14)
O8	0.3485(4)	0.1861(3)	0.2280(3)	0.0116(13)
O9	0.1708(4)	0.2490(3)	0.2193(3)	0.0169(14)
O10	0.2135(4)	0.1435(3)	0.3095(3)	0.0161(14)

	x/a	y/b	z/c	U(eq)
O11	0.1088(4)	0.1042(3)	0.1960(3)	0.0097(12)
O12	0.2645(4)	0.0469(3)	0.1997(3)	0.0087(12)
O13	0.0149(4)	0.9870(3)	0.1215(3)	0.0151(14)
O14	0.1152(4)	0.9495(3)	0.2247(3)	0.0118(12)
O15	0.1936(4)	0.9305(3)	0.1018(3)	0.0104(12)
O16	0.3561(4)	0.9337(3)	0.1638(3)	0.0111(13)
O17	0.3348(4)	0.8179(3)	0.0735(3)	0.0145(14)
O18	0.2793(4)	0.9135(3)	0.9809(3)	0.0139(13)
O19	0.1854(4)	0.9502(3)	0.5503(3)	0.0092(12)
O20	0.2295(4)	0.8317(3)	0.6175(3)	0.0096(12)
O21	0.3497(4)	0.9216(3)	0.5725(3)	0.0085(12)
O90	0.4835(5)	0.8130(4)	0.1982(3)	0.0220(16)
O91	0.9524(6)	0.3041(5)	0.4367(4)	0.048(2)
O92	0.9587(6)	0.3605(4)	0.1703(3)	0.0353(19)
O93	0.9836(5)	0.1968(4)	0.1491(3)	0.0246(16)
O94	0.0421(7)	0.2577(5)	0.0356(4)	0.043(2)
O95A	0.1656(16)	0.3304(14)	0.1059(11)	0.024(8)
O95B	0.1340(9)	0.3612(8)	0.1265(6)	0.028(4)
O96	0.0390(10)	0.3169(7)	0.3104(5)	0.095(4)
N1	0.6430(5)	0.9270(4)	0.8920(3)	0.0082(14)
N2	0.7539(5)	0.8567(4)	0.9701(3)	0.0095(15)

	x/a	y/b	z/c	U(eq)
N3	0.8000(5)	0.0024(4)	0.9513(3)	0.0104(14)
N4	0.9051(5)	0.9031(4)	0.8980(3)	0.0113(15)
N5	0.7967(5)	0.9797(4)	0.8151(3)	0.0084(15)
N6	0.7574(5)	0.8375(4)	0.8295(3)	0.0090(15)
C1	0.5921(6)	0.9562(5)	0.8440(4)	0.0110(18)
C2	0.4983(6)	0.9451(5)	0.8413(4)	0.0160(19)
C3	0.4555(6)	0.9016(5)	0.8899(4)	0.0136(19)
C4	0.5073(6)	0.8721(5)	0.9399(4)	0.0128(19)
C5	0.6017(6)	0.8862(4)	0.9400(4)	0.0083(17)
C6	0.6644(6)	0.8539(5)	0.9897(4)	0.0108(18)
C7	0.6379(6)	0.8262(5)	0.0493(4)	0.0136(19)
C8	0.7050(7)	0.7973(5)	0.0910(4)	0.020(2)
C9	0.7959(6)	0.7953(5)	0.0704(4)	0.019(2)
C10	0.8164(6)	0.8259(5)	0.0102(4)	0.0125(18)
C11	0.7383(6)	0.0499(5)	0.9767(4)	0.0109(18)
C12	0.7621(7)	0.1094(5)	0.0176(4)	0.018(2)
C13	0.8537(6)	0.1224(5)	0.0312(4)	0.017(2)
C14	0.9189(7)	0.0726(5)	0.0053(4)	0.0144(19)
C15	0.8907(6)	0.0133(5)	0.9656(4)	0.0124(18)
C16	0.9505(6)	0.9546(5)	0.9369(4)	0.0106(17)
C17	0.0448(6)	0.9484(5)	0.9470(4)	0.0147(19)

	x/a	y/b	z/c	U(eq)
C18	0.0919(6)	0.8908(5)	0.9170(4)	0.018(2)
C19	0.0448(6)	0.8387(6)	0.8771(4)	0.018(2)
C20	0.9524(6)	0.8460(5)	0.8692(4)	0.0143(19)
C21	0.8124(6)	0.0556(5)	0.8127(4)	0.0126(18)
C22	0.8412(6)	0.0905(5)	0.7553(4)	0.016(2)
C23	0.8568(6)	0.0476(5)	0.7006(4)	0.015(2)
C24	0.8386(6)	0.9692(5)	0.7030(4)	0.016(2)
C25	0.8074(6)	0.9372(5)	0.7598(4)	0.0097(17)
C26	0.7806(6)	0.8561(5)	0.7678(4)	0.0097(17)
C27	0.7756(6)	0.8039(5)	0.7168(4)	0.0132(18)
C28	0.7445(6)	0.7309(5)	0.7302(5)	0.017(2)
C29	0.7166(6)	0.7123(5)	0.7919(4)	0.0147(19)
C30	0.7235(6)	0.7665(5)	0.8410(4)	0.0144(19)
C31	0.4139(6)	0.0063(5)	0.2731(4)	0.0115(17)
C32	0.3697(6)	0.9547(5)	0.3235(4)	0.0097(17)
C33	0.2821(6)	0.9696(5)	0.3470(4)	0.0109(17)
C34	0.4182(6)	0.8953(5)	0.3525(4)	0.0106(17)
C35	0.2460(6)	0.9281(5)	0.3988(4)	0.0121(19)
C36	0.3834(6)	0.8537(5)	0.4044(4)	0.0108(18)
C37	0.2963(6)	0.8703(5)	0.4291(4)	0.0120(18)
C38	0.2599(6)	0.8280(5)	0.4882(4)	0.0115(18)

Table S28. Bond lengths (Å) for $[Co(2,2'\text{-bpy})_3](H_3O)[Mo_5O_{15}(1,4\text{-}O_3PC_8H_8O_3P)] \cdot 6H_2O$ (**4**·$6H_2O$).

Mo1-O3	1.700(6)	Mo1-O2	1.712(6)
Mo1-O1	1.893(6)	Mo1-O4	1.947(6)
Mo1-O19	2.231(6)	Mo1-O5	2.346(5)
Mo2-O7	1.699(6)	Mo2-O6	1.706(6)
Mo2-O4	1.897(5)	Mo2-O8	1.961(6)
Mo2-O20	2.303(6)	Mo2-O5	2.341(5)
Mo3-O10	1.688(6)	Mo3-O9	1.712(6)
Mo3-O8	1.857(6)	Mo3-O11	2.070(6)
Mo3-O12	2.163(6)	Mo3-O20	2.370(5)
Mo4-O13	1.701(6)	Mo4-O14	1.715(6)
Mo4-O15	1.868(6)	Mo4-O11	2.059(5)
Mo4-O21	2.173(5)	Mo4-O12	2.436(6)
Mo5-O17	1.698(6)	Mo5-O18	1.702(6)
Mo5-O1	1.922(6)	Mo5-O15	1.970(6)
Mo5-O16	2.225(6)	Mo5-O19	2.384(5)
Co1-N4	1.916(7)	Co1-N6	1.921(7)
Co1-N3	1.926(7)	Co1-N2	1.931(7)
Co1-N1	1.931(7)	Co1-N5	1.937(7)
P1-O16	1.503(6)	P1-O5	1.535(6)

P1-O12	1.552(6)	P1-C31	1.793(8)
P2-O21	1.511(6)	P2-O19	1.540(6)
P2-O20	1.544(6)	P2-C38	1.788(8)
O19-Mo1	2.232(6)	O19-Mo5	2.384(5)
O20-Mo2	2.303(6)	O20-Mo3	2.370(5)
O21-Mo4	2.173(5)	O90-H90A	0.84(3)
O90-H90B	0.84(3)	O91-H91A	0.86(3)
O91-H91B	0.86(3)	O92-H92A	0.84(3)
O92-H92B	0.85(3)	O93-H93A	0.82(3)
O93-H93B	0.84(3)	O94-H94A	0.82(3)
O94-H94B	0.84(3)	O96-H96A	0.85(3)
O96-H96B	0.85(3)	N1-C1	1.330(11)
N1-C5	1.351(10)	N2-C10	1.335(11)
N2-C6	1.365(11)	N3-C11	1.325(11)
N3-C15	1.366(11)	N4-C20	1.343(11)
N4-C16	1.367(11)	N5-C21	1.339(10)
N5-C25	1.358(10)	N6-C26	1.343(11)
N6-C30	1.347(10)	C1-C2	1.381(12)
C1-H1	0.93	C2-C3	1.394(12)
C2-H2	0.93	C3-C4	1.369(12)
C3-H3	0.93	C4-C5	1.396(11)
C4-H4	0.93	C5-C6	1.475(11)

C6-C7	1.364(12)	C7-C8	1.391(12)
C7-H7	0.93	C8-C9	1.391(13)
C8-H8	0.93	C9-C10	1.372(12)
C9-H9	0.93	C10-H10	0.93
C11-C12	1.372(12)	C11-H11	0.93
C12-C13	1.383(13)	C12-H12	0.93
C13-C14	1.388(13)	C13-H13	0.93
C14-C15	1.374(12)	C14-H14	0.93
C15-C16	1.463(12)	C16-C17	1.393(12)
C17-C18	1.359(13)	C17-H17	0.93
C18-C19	1.397(13)	C18-H18	0.93
C19-C20	1.362(12)	C19-H19	0.93
C20-H20	0.93	C21-C22	1.383(12)
C21-H21	0.93	C22-C23	1.361(12)
C22-H22	0.93	C23-C24	1.387(12)
C23-H23	0.93	C24-C25	1.364(12)
C24-H24	0.93	C25-C26	1.470(11)
C26-C27	1.381(11)	C27-C28	1.373(12)
C27-H27	0.93	C28-C29	1.363(12)
C28-H28	0.93	C29-C30	1.378(12)
C29-H29	0.93	C30-H30	0.93
C31-C32	1.510(11)	C31-H31A	0.97

C31-H31B	0.97	C32-C34	1.382(12)
C32-C33	1.387(11)	C33-C35	1.384(12)
C33-H33	0.93	C34-C36	1.379(12)
C34-H34	0.93	C35-C37	1.387(12)
C35-H35	0.93	C36-C37	1.395(12)
C36-H36	0.93	C37-C38	1.510(11)
C38-H38A	0.97	C38-H38B	0.97

Table S29. Bond angles (°) for $[Co(2,2'\text{-bpy})_3](H_3O)[Mo_5O_{15}(1,4\text{-}O_3PC_8H_8O_3P)] \cdot 6H_2O$ (**4**·$6H_2O$).

O3-Mo1-O2	103.8(3)	O3-Mo1-O1	98.0(3)
O2-Mo1-O1	101.4(3)	O3-Mo1-O4	99.7(3)
O2-Mo1-O4	94.7(2)	O1-Mo1-O4	152.5(2)
O3-Mo1-O19	159.4(2)	O2-Mo1-O19	96.5(3)
O1-Mo1-O19	74.2(2)	O4-Mo1-O19	82.0(2)
O3-Mo1-O5	89.3(2)	O2-Mo1-O5	163.0(2)
O1-Mo1-O5	87.2(2)	O4-Mo1-O5	72.1(2)
O19-Mo1-O5	71.56(19)	O7-Mo2-O6	103.8(3)
O7-Mo2-O4	100.6(3)	O6-Mo2-O4	102.8(3)
O7-Mo2-O8	100.5(3)	O6-Mo2-O8	96.8(3)
O4-Mo2-O8	146.7(2)	O7-Mo2-O20	88.2(3)
O6-Mo2-O20	165.6(2)	O4-Mo2-O20	82.5(2)
O8-Mo2-O20	72.8(2)	O7-Mo2-O5	166.3(3)
O6-Mo2-O5	89.5(3)	O4-Mo2-O5	73.0(2)
O8-Mo2-O5	80.6(2)	O20-Mo2-O5	79.13(19)
O10-Mo3-O9	103.5(3)	O10-Mo3-O8	98.1(3)
O9-Mo3-O8	104.1(3)	O10-Mo3-O11	98.5(3)
O9-Mo3-O11	91.2(3)	O8-Mo3-O11	154.1(2)
O10-Mo3-O12	96.8(2)	O9-Mo3-O12	155.2(3)
O8-Mo3-O12	86.8(2)	O11-Mo3-O12	71.7(2)

O10-Mo3-O20	166.4(3)	O9-Mo3-O20	88.9(2)
O8-Mo3-O20	72.9(2)	O11-Mo3-O20	86.8(2)
O12-Mo3-O20	72.8(2)	O13-Mo4-O14	101.6(3)
O13-Mo4-O15	103.9(3)	O14-Mo4-O15	101.5(3)
O13-Mo4-O11	99.8(3)	O14-Mo4-O11	93.1(2)
O15-Mo4-O11	148.9(2)	O13-Mo4-O21	88.4(2)
O14-Mo4-O21	167.0(2)	O15-Mo4-O21	83.9(2)
O11-Mo4-O21	76.8(2)	O13-Mo4-O12	165.8(2)
O14-Mo4-O12	82.9(2)	O15-Mo4-O12	88.2(2)
O11-Mo4-O12	66.3(2)	O21-Mo4-O12	85.42(19)
O17-Mo5-O18	101.9(3)	O17-Mo5-O1	99.7(3)
O18-Mo5-O1	100.1(3)	O17-Mo5-O15	104.0(3)
O18-Mo5-O15	98.2(3)	O1-Mo5-O15	146.1(2)
O17-Mo5-O16	87.7(2)	O18-Mo5-O16	170.3(3)
O1-Mo5-O16	77.8(2)	O15-Mo5-O16	79.3(2)
O17-Mo5-O19	168.8(3)	O18-Mo5-O19	85.0(2)
O1-Mo5-O19	70.1(2)	O15-Mo5-O19	83.5(2)
O16-Mo5-O19	85.46(19)	N4-Co1-N6	94.1(3)
N4-Co1-N3	83.2(3)	N6-Co1-N3	175.3(3)
N4-Co1-N2	92.1(3)	N6-Co1-N2	97.3(3)
N3-Co1-N2	86.6(3)	N4-Co1-N1	175.7(3)
N6-Co1-N1	85.4(3)	N3-Co1-N1	97.6(3)

N2-Co1-N1	83.7(3)	N4-Co1-N5	87.7(3)
N6-Co1-N5	82.8(3)	N3-Co1-N5	93.3(3)
N2-Co1-N5	179.8(3)	N1-Co1-N5	96.5(3)
O16-P1-O5	112.6(3)	O16-P1-O12	108.9(3)
O5-P1-O12	111.0(3)	O16-P1-C31	109.5(4)
O5-P1-C31	105.3(4)	O12-P1-C31	109.5(4)
O21-P2-O19	109.2(3)	O21-P2-O20	111.1(3)
O19-P2-O20	111.8(3)	O21-P2-C38	109.8(4)
O19-P2-C38	108.1(4)	O20-P2-C38	106.7(4)
Mo1-O1-Mo5	122.8(3)	Mo2-O4-Mo1	122.6(3)
P1-O5-Mo2	128.0(3)	P1-O5-Mo1	127.4(3)
Mo2-O5-Mo1	92.01(19)	Mo3-O8-Mo2	122.7(3)
Mo4-O11-Mo3	119.5(3)	P1-O12-Mo3	128.6(3)
P1-O12-Mo4	128.0(3)	Mo3-O12-Mo4	101.6(2)
Mo4-O15-Mo5	145.4(3)	P1-O16-Mo5	123.1(3)
P2-O19-Mo1	130.5(3)	P2-O19-Mo5	135.1(3)
Mo1-O19-Mo5	92.9(2)	P2-O20-Mo2	125.8(3)
P2-O20-Mo3	128.2(3)	Mo2-O20-Mo3	91.60(19)
P2-O21-Mo4	123.2(3)	H90A-O90-H90B	105.(6)
H91A-O91-H91B	108.(7)	H92A-O92-H92B	105.(6)
H93A-O93-H93B	110.(6)	H94A-O94-H94B	117.(7)
H96A-O96-H96B	106.(7)	C1-N1-C5	119.1(7)

C1-N1-Co1	126.3(6)	C5-N1-Co1	113.4(5)
C10-N2-C6	117.4(7)	C10-N2-Co1	127.8(6)
C6-N2-Co1	114.3(6)	C11-N3-C15	119.1(7)
C11-N3-Co1	126.0(6)	C15-N3-Co1	114.9(6)
C20-N4-C16	119.2(7)	C20-N4-Co1	125.7(6)
C16-N4-Co1	115.1(6)	C21-N5-C25	119.0(7)
C21-N5-Co1	127.0(6)	C25-N5-Co1	113.5(5)
C26-N6-C30	118.5(7)	C26-N6-Co1	114.9(5)
C30-N6-Co1	126.6(6)	N1-C1-C2	121.8(8)
N1-C1-H1	119.1	C2-C1-H1	119.1
C1-C2-C3	119.4(8)	C1-C2-H2	120.3
C3-C2-H2	120.3	C4-C3-C2	119.1(8)
C4-C3-H3	120.5	C2-C3-H3	120.5
C3-C4-C5	118.7(8)	C3-C4-H4	120.7
C5-C4-H4	120.7	N1-C5-C4	121.9(8)
N1-C5-C6	114.9(7)	C4-C5-C6	123.0(8)
C7-C6-N2	122.9(8)	C7-C6-C5	124.9(8)
N2-C6-C5	112.2(7)	C6-C7-C8	118.4(8)
C6-C7-H7	120.8	C8-C7-H7	120.8
C9-C8-C7	119.5(8)	C9-C8-H8	120.2
C7-C8-H8	120.2	C10-C9-C8	118.0(9)
C10-C9-H9	121.0	C8-C9-H9	121.0

N2-C10-C9	123.7(9)	N2-C10-H10	118.2
C9-C10-H10	118.2	N3-C11-C12	122.4(8)
N3-C11-H11	118.8	C12-C11-H11	118.8
C11-C12-C13	119.3(9)	C11-C12-H12	120.4
C13-C12-H12	120.4	C12-C13-C14	118.8(8)
C12-C13-H13	120.6	C14-C13-H13	120.6
C15-C14-C13	119.1(9)	C15-C14-H14	120.4
C13-C14-H14	120.4	N3-C15-C14	121.3(8)
N3-C15-C16	113.3(7)	C14-C15-C16	125.4(8)
N4-C16-C17	120.9(8)	N4-C16-C15	113.5(7)
C17-C16-C15	125.6(8)	C18-C17-C16	119.2(8)
C18-C17-H17	120.4	C16-C17-H17	120.4
C17-C18-C19	119.4(9)	C17-C18-H18	120.3
C19-C18-H18	120.3	C20-C19-C18	119.6(9)
C20-C19-H19	120.2	C18-C19-H19	120.2
N4-C20-C19	121.6(9)	N4-C20-H20	119.2
C19-C20-H20	119.2	N5-C21-C22	120.9(8)
N5-C21-H21	119.5	C22-C21-H21	119.5
C23-C22-C21	120.4(9)	C23-C22-H22	119.8
C21-C22-H22	119.8	C22-C23-C24	118.5(9)
C22-C23-H23	120.8	C24-C23-H23	120.8
C25-C24-C23	119.5(9)	C25-C24-H24	120.3

C23-C24-H24	120.3	N5-C25-C24	121.6(8)
N5-C25-C26	113.4(7)	C24-C25-C26	125.0(8)
N6-C26-C27	122.3(8)	N6-C26-C25	113.7(7)
C27-C26-C25	123.9(8)	C28-C27-C26	118.2(8)
C28-C27-H27	120.9	C26-C27-H27	120.9
C29-C28-C27	120.0(8)	C29-C28-H28	120.0
C27-C28-H28	120.0	C28-C29-C30	119.3(8)
C28-C29-H29	120.4	C30-C29-H29	120.4
N6-C30-C29	121.5(8)	N6-C30-H30	119.2
C29-C30-H30	119.2	C32-C31-P1	118.5(6)
C32-C31-H31A	107.7	P1-C31-H31A	107.7
C32-C31-H31B	107.7	P1-C31-H31B	107.7
H31A-C31-H31B	107.1	C34-C32-C33	117.5(8)
C34-C32-C31	121.1(8)	C33-C32-C31	121.2(8)
C35-C33-C32	121.1(8)	C35-C33-H33	119.4
C32-C33-H33	119.4	C36-C34-C32	122.1(8)
C36-C34-H34	119.0	C32-C34-H34	119.0
C33-C35-C37	121.0(8)	C33-C35-H35	119.5
C37-C35-H35	119.5	C34-C36-C37	120.2(8)
C34-C36-H36	119.9	C37-C36-H36	119.9
C35-C37-C36	118.0(8)	C35-C37-C38	121.4(8)
C36-C37-C38	120.6(8)	C37-C38-P2	113.5(6)

C37-C38-H38A	108.9	P2-C38-H38A	108.9
C37-C38-H38B	108.9	P2-C38-H38B	108.9
H38A-C38-H38B	107.7		

Table S30. Torsion angles (°) for $[Co(2,2'\text{-bpy})_3](H_3O)[Mo_5O_{15}(1,4\text{-}O_3PC_8H_8O_3P)] \cdot 6H_2O$ (**4**·$6H_2O$).

O3-Mo1-O1-Mo5	158.4(3)	O2-Mo1-O1-Mo5	-95.7(4)
O4-Mo1-O1-Mo5	28.9(7)	O19-Mo1-O1-Mo5	-2.1(3)
O5-Mo1-O1-Mo5	69.5(3)	O7-Mo2-O4-Mo1	-172.6(3)
O6-Mo2-O4-Mo1	80.5(4)	O8-Mo2-O4-Mo1	-44.1(6)
O20-Mo2-O4-Mo1	-85.9(3)	O5-Mo2-O4-Mo1	-5.0(3)
O16-P1-O5-Mo2	152.5(3)	O12-P1-O5-Mo2	30.2(5)
C31-P1-O5-Mo2	-88.2(5)	O16-P1-O5-Mo1	21.9(5)
O12-P1-O5-Mo1	-100.5(4)	C31-P1-O5-Mo1	141.1(4)
O10-Mo3-O8-Mo2	171.4(4)	O9-Mo3-O8-Mo2	-82.4(4)
O11-Mo3-O8-Mo2	42.0(7)	O12-Mo3-O8-Mo2	75.0(3)
O20-Mo3-O8-Mo2	2.0(3)	O16-P1-O12-Mo3	-173.1(4)
O5-P1-O12-Mo3	-48.6(5)	C31-P1-O12-Mo3	67.2(5)
O16-P1-O12-Mo4	-11.6(5)	O5-P1-O12-Mo4	112.9(4)
C31-P1-O12-Mo4	-131.3(4)	O13-Mo4-O15-Mo5	145.1(5)
O14-Mo4-O15-Mo5	-109.8(5)	O11-Mo4-O15-Mo5	6.6(9)
O21-Mo4-O15-Mo5	58.2(5)	O12-Mo4-O15-Mo5	-27.3(5)
O5-P1-O16-Mo5	-55.0(5)	O12-P1-O16-Mo5	68.5(4)
C31-P1-O16-Mo5	-171.9(4)	O21-P2-O19-Mo1	-170.8(4)
O20-P2-O19-Mo1	-47.5(5)	C38-P2-O19-Mo1	69.7(5)

O21-P2-O19-Mo5	-8.3(5)	O20-P2-O19-Mo5	115.1(4)
C38-P2-O19-Mo5	-127.7(5)	O21-P2-O20-Mo2	155.3(3)
O19-P2-O20-Mo2	33.1(5)	C38-P2-O20-Mo2	-85.0(5)
O21-P2-O20-Mo3	27.7(5)	O19-P2-O20-Mo3	-94.6(4)
C38-P2-O20-Mo3	147.3(4)	O19-P2-O21-Mo4	58.0(4)
O20-P2-O21-Mo4	-65.8(4)	C38-P2-O21-Mo4	176.4(4)
C5-N1-C1-C2	-1.0(12)	Co1-N1-C1-C2	165.6(6)
N1-C1-C2-C3	-0.5(13)	C1-C2-C3-C4	1.3(13)
C2-C3-C4-C5	-0.6(13)	C1-N1-C5-C4	1.7(12)
Co1-N1-C5-C4	-166.5(6)	C1-N1-C5-C6	177.9(7)
Co1-N1-C5-C6	9.7(9)	C3-C4-C5-N1	-0.9(12)
C3-C4-C5-C6	-176.8(8)	C10-N2-C6-C7	4.6(12)
Co1-N2-C6-C7	-167.3(7)	C10-N2-C6-C5	-177.2(7)
Co1-N2-C6-C5	10.9(9)	N1-C5-C6-C7	164.6(8)
C4-C5-C6-C7	-19.3(13)	N1-C5-C6-N2	-13.6(10)
C4-C5-C6-N2	162.6(8)	N2-C6-C7-C8	-2.1(13)
C5-C6-C7-C8	179.9(8)	C6-C7-C8-C9	-1.9(13)
C7-C8-C9-C10	3.3(13)	C6-N2-C10-C9	-3.1(12)
Co1-N2-C10-C9	167.5(7)	C8-C9-C10-N2	-0.7(13)
C15-N3-C11-C12	-0.5(12)	Co1-N3-C11-C12	-179.1(6)
N3-C11-C12-C13	2.2(13)	C11-C12-C13-C14	-2.7(13)
C12-C13-C14-C15	1.7(13)	C11-N3-C15-C14	-0.5(12)

Co1-N3-C15-C14	178.2(6)	C11-N3-C15-C16	177.2(7)
Co1-N3-C15-C16	-4.1(9)	C13-C14-C15-N3	-0.1(13)
C13-C14-C15-C16	-177.5(8)	C20-N4-C16-C17	-0.1(12)
Co1-N4-C16-C17	177.5(6)	C20-N4-C16-C15	-179.4(7)
Co1-N4-C16-C15	-1.8(9)	N3-C15-C16-N4	3.8(10)
C14-C15-C16-N4	-178.6(8)	N3-C15-C16-C17	-175.4(8)
C14-C15-C16-C17	2.2(13)	N4-C16-C17-C18	0.8(12)
C15-C16-C17-C18	-180.0(8)	C16-C17-C18-C19	-0.5(13)
C17-C18-C19-C20	-0.5(13)	C16-N4-C20-C19	-0.9(12)
Co1-N4-C20-C19	-178.2(6)	C18-C19-C20-N4	1.2(13)
C25-N5-C21-C22	-1.5(12)	Co1-N5-C21-C22	169.7(6)
N5-C21-C22-C23	-2.0(13)	C21-C22-C23-C24	3.3(13)
C22-C23-C24-C25	-1.1(13)	C21-N5-C25-C24	3.8(12)
Co1-N5-C25-C24	-168.5(7)	C21-N5-C25-C26	-175.1(7)
Co1-N5-C25-C26	12.5(9)	C23-C24-C25-N5	-2.5(13)
C23-C24-C25-C26	176.3(8)	C30-N6-C26-C27	-3.6(12)
Co1-N6-C26-C27	177.3(7)	C30-N6-C26-C25	173.9(7)
Co1-N6-C26-C25	-5.2(9)	N5-C25-C26-N6	-4.9(10)
C24-C25-C26-N6	176.1(8)	N5-C25-C26-C27	172.5(8)
C24-C25-C26-C27	-6.4(14)	N6-C26-C27-C28	1.0(13)
C25-C26-C27-C28	-176.3(8)	C26-C27-C28-C29	2.1(13)
C27-C28-C29-C30	-2.5(14)	C26-N6-C30-C29	3.2(12)

Co1-N6-C30-C29	-177.9(6)	C28-C29-C30-N6	-0.2(13)
O16-P1-C31-C32	-56.8(7)	O5-P1-C31-C32	-178.1(6)
O12-P1-C31-C32	62.5(7)	P1-C31-C32-C34	122.2(8)
P1-C31-C32-C33	-64.0(10)	C34-C32-C33-C35	2.0(12)
C31-C32-C33-C35	-172.0(8)	C33-C32-C34-C36	-2.4(12)
C31-C32-C34-C36	171.6(8)	C32-C33-C35-C37	0.1(13)
C32-C34-C36-C37	0.7(13)	C33-C35-C37-C36	-1.8(12)
C33-C35-C37-C38	176.3(8)	C34-C36-C37-C35	1.5(12)
C34-C36-C37-C38	-176.7(8)	C35-C37-C38-P2	-70.7(10)
C36-C37-C38-P2	107.4(8)	O21-P2-C38-C37	-52.7(7)
O19-P2-C38-C37	66.4(7)	O20-P2-C38-C37	-173.1(6)

Table S31. Anisotropic atomic displacement parameters ($Å^2$) for $[Co(2,2'\text{-bpy})_3](H_3O)[Mo_5O_{15}(1,4\text{-}O_3PC_8H_8O_3P)] \cdot 6H_2O$ (**4**·$6H_2O$).

The anisotropic atomic displacement factor exponent takes the form: $-2\pi^2[\, h^2\, a^{*2}\, U_{11} + \ldots + 2\, h\, k\, a^*\, b^*\, U_{12}\,]$

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
Mo1	0.0082(4)	0.0110(4)	0.0091(4)	0.0002(3)	0.0007(3)	-0.0018(3)
Mo2	0.0105(4)	0.0093(4)	0.0102(4)	-0.0007(3)	0.0001(3)	-0.0038(3)
Mo3	0.0101(4)	0.0080(4)	0.0102(4)	-0.0004(3)	-0.0002(3)	-0.0005(3)
Mo4	0.0074(4)	0.0075(4)	0.0123(4)	0.0010(3)	0.0002(3)	-0.0006(3)
Mo5	0.0086(4)	0.0085(4)	0.0104(4)	-0.0014(3)	0.0004(3)	-0.0008(3)
Co1	0.0060(6)	0.0081(6)	0.0105(5)	0.0002(5)	0.0005(5)	-0.0011(5)
P1	0.0076(11)	0.0093(12)	0.0085(10)	0.0007(9)	0.0005(8)	-0.0010(9)
P2	0.0099(12)	0.0073(10)	0.0082(10)	0.0006(9)	0.0016(9)	-0.0009(9)
O1	0.011(3)	0.012(3)	0.011(3)	0.000(3)	0.001(2)	0.000(2)
O2	0.019(3)	0.011(3)	0.014(3)	-0.003(3)	0.003(3)	0.000(3)
O3	0.008(3)	0.023(3)	0.015(3)	-0.003(3)	0.002(2)	-0.002(3)
O4	0.012(3)	0.011(3)	0.006(3)	0.002(2)	0.000(2)	-0.003(2)
O5	0.009(3)	0.008(3)	0.010(3)	0.001(2)	0.001(2)	-0.003(2)
O6	0.010(3)	0.021(3)	0.016(3)	-0.004(3)	-0.005(3)	-0.006(3)
O7	0.019(4)	0.009(3)	0.018(3)	0.000(3)	0.003(3)	-0.003(3)
O8	0.012(3)	0.014(3)	0.009(3)	-0.002(3)	0.000(2)	-0.004(2)
O9	0.020(4)	0.012(3)	0.018(3)	-0.001(3)	-0.001(3)	0.003(3)
O10	0.016(3)	0.017(3)	0.016(3)	0.001(3)	-0.001(3)	-0.001(3)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
O11	0.010(3)	0.006(3)	0.012(3)	0.001(2)	0.002(2)	0.000(2)
O12	0.007(3)	0.006(3)	0.013(3)	0.000(2)	0.002(2)	0.001(2)
O13	0.008(3)	0.016(3)	0.021(3)	-0.002(3)	-0.003(2)	-0.002(3)
O14	0.012(3)	0.008(3)	0.015(3)	0.001(3)	0.006(3)	0.001(3)
O15	0.011(3)	0.008(3)	0.012(3)	-0.001(2)	0.002(2)	-0.002(2)
O16	0.012(3)	0.010(3)	0.012(3)	0.002(2)	0.000(2)	0.003(2)
O17	0.014(3)	0.012(3)	0.018(3)	-0.002(3)	0.002(3)	0.000(3)
O18	0.015(3)	0.014(3)	0.013(3)	-0.002(3)	0.001(2)	-0.006(3)
O19	0.009(3)	0.007(3)	0.012(3)	-0.002(2)	-0.002(2)	0.000(2)
O20	0.007(3)	0.011(3)	0.011(3)	0.001(2)	0.002(2)	-0.001(3)
O21	0.009(3)	0.005(3)	0.011(3)	-0.001(2)	0.002(2)	-0.002(2)
O90	0.025(4)	0.013(4)	0.028(4)	0.002(3)	-0.004(3)	0.004(3)
O91	0.054(6)	0.039(5)	0.052(6)	0.003(5)	0.005(5)	0.001(5)
O92	0.060(6)	0.025(4)	0.021(4)	0.002(3)	0.009(4)	0.007(4)
O93	0.023(4)	0.012(3)	0.039(4)	-0.004(3)	-0.014(4)	0.000(3)
O94	0.065(7)	0.040(5)	0.023(4)	-0.001(4)	-0.005(4)	0.037(5)
O96	0.095(9)	0.127(11)	0.063(7)	0.030(7)	0.027(7)	0.073(9)
N1	0.005(3)	0.008(4)	0.011(3)	-0.001(3)	0.000(3)	-0.001(3)
N2	0.009(4)	0.007(4)	0.013(3)	0.000(3)	-0.001(3)	-0.003(3)
N3	0.013(4)	0.011(3)	0.007(3)	0.001(3)	-0.002(3)	-0.006(3)
N4	0.007(3)	0.013(4)	0.014(4)	0.002(3)	-0.001(3)	-0.002(3)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
N5	0.004(4)	0.010(3)	0.011(3)	0.001(3)	0.001(3)	-0.005(3)
N6	0.006(4)	0.008(3)	0.012(3)	-0.004(3)	0.000(3)	0.003(3)
C1	0.007(4)	0.011(4)	0.015(4)	-0.001(4)	0.002(3)	0.002(4)
C2	0.006(4)	0.020(5)	0.022(5)	0.000(4)	-0.004(4)	0.007(4)
C3	0.005(4)	0.015(5)	0.021(5)	0.002(4)	0.000(3)	0.002(4)
C4	0.007(4)	0.015(5)	0.016(4)	-0.004(4)	0.006(3)	-0.005(4)
C5	0.008(4)	0.004(4)	0.013(4)	-0.006(3)	-0.001(3)	0.002(3)
C6	0.008(4)	0.010(5)	0.014(4)	-0.003(3)	0.000(3)	0.003(4)
C7	0.011(5)	0.018(5)	0.013(4)	-0.003(4)	-0.001(3)	-0.010(4)
C8	0.024(5)	0.016(5)	0.019(5)	0.005(4)	0.000(4)	-0.004(4)
C9	0.021(5)	0.015(5)	0.021(5)	0.004(4)	-0.008(4)	-0.008(4)
C10	0.009(4)	0.009(5)	0.020(4)	0.001(4)	-0.006(4)	0.000(4)
C11	0.011(4)	0.014(4)	0.008(4)	-0.002(3)	-0.001(3)	-0.005(4)
C12	0.019(5)	0.020(5)	0.014(5)	-0.001(4)	0.004(4)	0.005(4)
C13	0.022(5)	0.012(5)	0.018(5)	-0.009(4)	-0.001(4)	-0.009(4)
C14	0.018(5)	0.006(4)	0.019(5)	0.001(4)	-0.008(4)	-0.003(4)
C15	0.010(4)	0.013(4)	0.014(4)	0.005(3)	-0.003(3)	-0.002(4)
C16	0.008(4)	0.013(4)	0.010(4)	0.005(3)	0.002(3)	-0.003(3)
C17	0.005(4)	0.018(5)	0.021(5)	0.005(4)	-0.005(4)	-0.006(4)
C18	0.006(5)	0.025(5)	0.022(5)	0.010(4)	0.003(4)	0.003(4)
C19	0.012(5)	0.019(5)	0.022(5)	0.006(4)	0.006(4)	0.005(4)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
C20	0.014(5)	0.019(5)	0.010(4)	0.004(4)	-0.001(4)	0.000(4)
C21	0.011(5)	0.010(4)	0.017(4)	-0.002(4)	0.000(4)	0.002(4)
C22	0.017(5)	0.010(4)	0.021(5)	0.006(4)	-0.003(4)	0.000(4)
C23	0.014(5)	0.015(5)	0.016(4)	0.004(4)	0.001(4)	-0.002(4)
C24	0.017(5)	0.017(5)	0.014(4)	-0.004(4)	0.004(4)	0.001(4)
C25	0.003(4)	0.011(4)	0.015(4)	-0.001(3)	-0.002(3)	0.001(3)
C26	0.003(4)	0.009(4)	0.017(4)	-0.008(3)	-0.001(4)	-0.001(3)
C27	0.012(5)	0.015(4)	0.013(4)	0.001(4)	0.000(4)	0.001(4)
C28	0.023(6)	0.012(4)	0.017(4)	-0.004(4)	-0.005(4)	0.001(4)
C29	0.018(5)	0.004(4)	0.022(5)	-0.002(3)	0.002(4)	-0.002(4)
C30	0.009(5)	0.013(4)	0.021(5)	0.001(4)	0.005(4)	0.002(4)
C31	0.011(4)	0.013(4)	0.010(4)	0.000(4)	-0.002(3)	0.001(4)
C32	0.006(4)	0.015(5)	0.008(4)	-0.003(3)	0.001(3)	-0.005(4)
C33	0.005(4)	0.010(4)	0.018(4)	0.000(4)	-0.005(4)	-0.001(3)
C34	0.010(4)	0.010(4)	0.011(4)	-0.005(3)	0.003(4)	0.003(3)
C35	0.012(5)	0.012(5)	0.012(4)	-0.003(4)	0.000(3)	0.003(4)
C36	0.011(4)	0.006(4)	0.015(4)	-0.003(3)	-0.003(4)	0.004(4)
C37	0.015(5)	0.008(4)	0.013(4)	-0.002(3)	0.002(3)	-0.006(4)
C38	0.013(5)	0.005(4)	0.016(4)	-0.002(3)	0.005(3)	-0.001(4)

Table S32. Hydrogen atomic coordinates and isotropic atomic displacement parameters ($Å^2$) for $[Co(2,2'\text{-bpy})_3](H_3O)[Mo_5O_{15}(1,4\text{-}O_3PC_8H_8O_3P)] \cdot 6H_2O$ (**4**·$6H_2O$).

	x/a	y/b	z/c	U(eq)
H90A	0.453(6)	0.853(3)	0.203(4)	0.026
H90B	0.493(7)	0.797(5)	0.237(2)	0.026
H91A	0.987(7)	0.305(6)	0.403(4)	0.058
H91B	0.931(8)	0.349(3)	0.442(6)	0.058
H92A	0.926(7)	0.388(5)	0.194(4)	0.042
H92B	0.954(8)	0.380(5)	0.132(2)	0.042
H93A	0.961(7)	0.155(3)	0.158(4)	0.03
H93B	0.983(7)	0.225(4)	0.182(3)	0.03
H94A	0.005(6)	0.281(5)	0.012(5)	0.051
H94B	0.070(7)	0.283(5)	0.064(4)	0.051
H96A	0.030(13)	0.276(5)	0.333(6)	0.114
H96B	0.060(12)	0.302(8)	0.273(4)	0.114
H1	0.6204	0.9849	0.8113	0.013
H2	0.4641	0.9665	0.8075	0.019
H3	0.3926	0.8927	0.8884	0.016
H4	0.4803	0.8433	0.9731	0.015
H7	0.5765	0.8267	1.0617	0.016
H8	0.6892	0.7794	1.1325	0.023
H9	0.8414	0.7738	1.0966	0.023

	x/a	y/b	z/c	U(eq)
H10	0.8772	0.8251	0.9964	0.015
H11	0.6768	1.0425	0.9666	0.013
H12	0.7171	1.1407	1.0359	0.021
H13	0.8714	1.1638	1.0573	0.021
H14	0.9808	1.0794	1.0148	0.017
H17	1.0751	0.9833	0.9740	0.018
H18	1.1549	0.8861	0.9230	0.021
H19	1.0764	0.7993	0.8560	0.021
H20	0.9213	0.8105	0.8433	0.017
H21	0.8039	1.0854	0.8501	0.015
H22	0.8499	1.1436	0.7541	0.019
H23	0.8791	1.0704	0.6626	0.018
H24	0.8477	0.9387	0.6661	0.019
H27	0.7929	0.8179	0.6746	0.016
H28	0.7424	0.6942	0.6971	0.021
H29	0.6933	0.6636	0.8009	0.018
H30	0.7043	0.7539	0.8831	0.017
H31A	0.4768	0.9892	0.2673	0.014
H31B	0.4164	1.0579	0.2912	0.014
H33	0.2471	1.0081	0.3276	0.013
H34	0.4761	0.8830	0.3364	0.013

	x/a	y/b	z/c	U(eq)
H35	0.1871	0.9392	0.4136	0.015
H36	0.4181	0.8145	0.4230	0.013
H38A	0.2981	0.7832	0.4963	0.014
H38B	0.1983	0.8099	0.4787	0.014

Table S33. Sample and crystal data for $[\{Cu(2,2\text{-bpy})_2\}_2Mo_5O_{15}(1,4\text{-}O_3PC_8H_8PO_3)] \cdot 6H_2O$ (**5**·$6H_2O$).

Identification code	$[\{Cu(2,2\text{-bpy})_2\}_2Mo_5O_{15}(1,4\text{-}O_3PC_8H_8PO_3)] \cdot 6H_2O$	
Chemical formula	$C_{48}H_{52}Cu_2Mo_5N_8O_{27}P_2$	
Formula weight	1841.69	
Temperature	90(2) K	
Wavelength	0.71073 Å	
Crystal size	0.063 x 0.064 x 0.123 mm	
Crystal system	Triclinic	
Space group	P -1	
Unit cell dimensions	a = 13.0366(13) Å	α = 76.271(3)°
	b = 13.0703(13) Å	β = 85.514(3)°
	c = 20.989(2) Å	γ = 60.541(3)°
Volume	3021.9(5) $Å^3$	
Z	2	
Density (calculated)	2.024 g/cm^3	
Absorption coefficient	1.840 mm^{-1}	
F(000)	1820	

Table S34. Data collection and structure refinement for [{Cu(2,2-bpy)$_2$}$_2$Mo$_5$O$_{15}$(1,4-O$_3$PC$_8$H$_8$PO$_3$)] ·6H$_2$O (**5**·6H$_2$O).

Theta range for data collection	1.80 to 25.03°	
Index ranges	-14<=h<=15, -15<=k<=15, -24<=l<=24	
Reflections collected	33166	
Independent reflections	10663 [R(int) = 0.0530]	
Coverage of independent reflections	99.8%	
Absorption correction	multi-scan	
Max. and min. transmission	0.8930 and 0.8050	
Structure solution technique	direct methods	
Structure solution program	SHELXS-97 (Sheldrick, 2008)	
Refinement method	Full-matrix least-squares on F^2	
Refinement program	SHELXL-2013 (Sheldrick, 2013)	
Function minimized	$\Sigma\, w(F_o^2 - F_c^2)^2$	
Data / restraints / parameters	10663 / 6 / 821	
Goodness-of-fit on F^2	1.041	
Δ/σ_{max}	2.070	
Final R indices	8149 data; I>2σ(I)	R1 = 0.0450, wR2 = 0.1178
	all data	R1 = 0.0671, wR2 = 0.1292
Weighting scheme	$w=1/[\sigma^2(F_o^2)+(0.0606P)^2+18.5078P]$ where $P=(F_o^2+2F_c^2)/3$	
Largest diff. peak and hole	2.114 and -0.767 eÅ^{-3}	
R.M.S. deviation from mean	0.182 eÅ^{-3}	

Table S35. Atomic coordinates and equivalent isotropic atomic displacement parameters ($Å^2$) for $[\{Cu(2,2\text{-bpy})_2\}_2Mo_5O_{15}(1,4\text{-}O_3PC_8H_8PO_3)] \cdot 6H_2O$ (**5**·$6H_2O$).

U(eq) is defined as one third of the trace of the orthogonalized U_{ij} tensor.

	x/a	**y/b**	**z/c**	**U(eq)**
Mo1	0.91934(5)	0.89271(5)	0.27787(3)	0.01296(15)
Mo2	0.19444(5)	0.77422(5)	0.34394(3)	0.00851(14)
Mo3	0.38579(5)	0.56473(5)	0.26304(3)	0.00997(14)
Mo4	0.23353(5)	0.52310(5)	0.15856(3)	0.01144(15)
Mo5	0.94417(5)	0.67798(5)	0.18352(3)	0.01327(15)
Cu1	0.31923(7)	0.92749(7)	0.38978(4)	0.01181(19)
Cu2	0.38905(8)	0.23734(7)	0.12499(4)	0.01240(19)
P1	0.13555(16)	0.80838(15)	0.17694(9)	0.0113(4)
P2	0.12333(15)	0.57034(15)	0.31523(8)	0.0090(4)
O1	0.8620(4)	0.8507(4)	0.3492(3)	0.0184(11)
O2	0.8157(4)	0.0390(4)	0.2503(3)	0.0200(12)
O3	0.0421(4)	0.9078(4)	0.3154(2)	0.0110(10)
O4	0.0218(4)	0.9137(4)	0.1908(2)	0.0143(11)
O5	0.8887(4)	0.8090(4)	0.2252(2)	0.0158(11)
O6	0.1803(4)	0.7492(4)	0.4267(2)	0.0132(10)
O7	0.2715(4)	0.8557(4)	0.3301(2)	0.0123(10)
O8	0.3219(4)	0.6164(4)	0.3424(2)	0.0095(10)
O9	0.2305(4)	0.7600(4)	0.2320(2)	0.0118(10)

	x/a	y/b	z/c	U(eq)
O10	0.4810(4)	0.6216(4)	0.2455(2)	0.0160(11)
O11	0.4796(4)	0.4149(4)	0.2949(2)	0.0152(11)
O12	0.3500(4)	0.5639(4)	0.1765(2)	0.0136(10)
O13	0.2326(4)	0.5195(4)	0.2730(2)	0.0095(10)
O14	0.2388(5)	0.5542(4)	0.0754(2)	0.0190(11)
O15	0.3157(4)	0.3656(4)	0.1824(2)	0.0144(11)
O16	0.0828(4)	0.5341(4)	0.1686(2)	0.0131(10)
O17	0.1104(4)	0.7082(4)	0.1682(2)	0.0121(10)
O18	0.8992(5)	0.7557(4)	0.1038(2)	0.0220(12)
O19	0.8454(4)	0.6261(4)	0.2041(2)	0.0178(11)
O20	0.0183(4)	0.5735(4)	0.2882(2)	0.0121(10)
O21	0.0906(4)	0.6986(4)	0.3211(2)	0.0110(10)
O90	0.7727(7)	0.7085(7)	0.4158(5)	0.064(2)
O91	0.7208(7)	0.4175(8)	0.2929(5)	0.071(3)
O92	0.7841(9)	0.5691(7)	0.3318(5)	0.077(3)
N1	0.4225(5)	0.0079(5)	0.3690(3)	0.0136(13)
N2	0.1985(5)	0.0902(5)	0.3453(3)	0.0125(12)
N3	0.4362(5)	0.7638(5)	0.4374(3)	0.0137(13)
N4	0.2466(5)	0.9271(5)	0.4811(3)	0.0133(13)
N5	0.3816(6)	0.2879(6)	0.0257(3)	0.0202(14)
N6	0.5219(5)	0.2727(5)	0.1139(3)	0.0155(13)

	x/a	y/b	z/c	U(eq)
N7	0.4634(5)	0.0671(5)	0.1849(3)	0.0134(13)
N8	0.2476(5)	0.2148(5)	0.1357(3)	0.0147(13)
C1	0.5365(6)	0.9602(6)	0.3854(4)	0.0181(16)
C2	0.5947(7)	0.0263(7)	0.3731(4)	0.0198(17)
C3	0.5332(6)	0.1471(6)	0.3433(4)	0.0167(16)
C4	0.4141(6)	0.1985(6)	0.3259(3)	0.0137(15)
C5	0.3611(6)	0.1266(6)	0.3393(3)	0.0119(14)
C6	0.2355(6)	0.1713(6)	0.3240(3)	0.0096(14)
C7	0.1587(6)	0.2880(6)	0.2904(3)	0.0147(15)
C8	0.0401(6)	0.3240(6)	0.2819(4)	0.0149(15)
C9	0.0036(7)	0.2382(6)	0.3056(4)	0.0187(16)
C10	0.0853(6)	0.1230(6)	0.3353(4)	0.0176(16)
C11	0.5293(7)	0.6857(6)	0.4099(4)	0.0190(16)
C12	0.6101(6)	0.5739(6)	0.4459(4)	0.0170(16)
C13	0.5952(7)	0.5419(7)	0.5114(4)	0.0203(17)
C14	0.4988(6)	0.6229(6)	0.5400(4)	0.0164(16)
C15	0.4192(7)	0.7323(6)	0.5015(3)	0.0146(15)
C16	0.3089(6)	0.8224(6)	0.5257(4)	0.0140(15)
C17	0.2712(7)	0.8028(7)	0.5891(4)	0.0181(16)
C18	0.1717(7)	0.8932(7)	0.6077(4)	0.0180(16)
C19	0.1077(7)	0.0023(7)	0.5627(4)	0.0206(17)

	x/a	y/b	z/c	U(eq)
C20	0.1486(6)	0.0138(7)	0.4999(4)	0.0172(16)
C21	0.3089(8)	0.2866(9)	0.9842(4)	0.033(2)
C22	0.3140(9)	0.3171(9)	0.9175(4)	0.039(2)
C23	0.3984(9)	0.3488(9)	0.8919(4)	0.038(2)
C24	0.4750(7)	0.3478(7)	0.9342(4)	0.0262(19)
C25	0.4645(7)	0.3171(7)	0.0011(4)	0.0204(17)
C26	0.5413(7)	0.3130(6)	0.0505(4)	0.0179(16)
C27	0.6260(7)	0.3482(7)	0.0358(4)	0.028(2)
C28	0.6891(8)	0.3459(8)	0.0871(5)	0.035(2)
C29	0.6674(7)	0.3071(8)	0.1511(5)	0.032(2)
C30	0.5822(7)	0.2714(7)	0.1621(4)	0.0199(17)
C31	0.5767(6)	0.9961(6)	0.2058(4)	0.0169(16)
C32	0.6232(7)	0.8740(7)	0.2335(4)	0.0198(17)
C33	0.5511(7)	0.8233(7)	0.2375(4)	0.0199(17)
C34	0.4324(7)	0.8969(6)	0.2169(3)	0.0153(16)
C35	0.3912(6)	0.0180(6)	0.1919(3)	0.0133(15)
C36	0.2672(6)	0.1052(6)	0.1701(3)	0.0139(15)
C37	0.1761(7)	0.0787(7)	0.1840(4)	0.0201(17)
C38	0.0629(7)	0.1659(7)	0.1618(4)	0.0249(18)
C39	0.0423(7)	0.2795(7)	0.1270(4)	0.0208(17)
C40	0.1367(7)	0.3009(7)	0.1155(4)	0.0182(16)

	x/a	y/b	z/c	U(eq)
C41	0.1905(6)	0.8558(6)	0.1005(3)	0.0161(16)
C42	0.0933(7)	0.9307(6)	0.0470(3)	0.0170(16)
C43	0.0448(7)	0.8763(6)	0.0191(4)	0.0210(18)
C44	0.0477(7)	0.0539(7)	0.0269(3)	0.0197(17)
C45	0.1695(6)	0.4688(6)	0.3958(3)	0.0120(14)
C46	0.0799(6)	0.4881(6)	0.4483(3)	0.0120(14)
C47	0.0216(7)	0.4193(6)	0.4615(4)	0.0174(16)
C48	0.0554(6)	0.5695(6)	0.4877(4)	0.0172(16)

Table S36. Bond lengths (Å) for $[\{Cu(2,2\text{-bpy})_2\}_2Mo_5O_{15}(1,4\text{-}O_3PC_8H_8PO_3)]\cdot 6H_2O$ (**5**·$6H_2O$).

Mo1-O2	1.695(5)	Mo1-O1	1.716(5)
Mo1-O5	1.911(5)	Mo1-O3	1.950(5)
Mo1-O4	2.223(5)	Mo1-O21	2.429(5)
Mo2-O6	1.702(5)	Mo2-O7	1.757(5)
Mo2-O3	1.905(5)	Mo2-O8	1.916(4)
Mo2-O21	2.164(5)	Mo2-O9	2.393(5)
Mo3-O11	1.712(5)	Mo3-O10	1.713(5)
Mo3-O12	1.914(5)	Mo3-O8	1.920(4)
Mo3-O9	2.318(4)	Mo3-O13	2.328(4)
Mo4-O14	1.700(5)	Mo4-O15	1.750(5)
Mo4-O16	1.896(5)	Mo4-O12	1.926(5)
Mo4-O17	2.198(5)	Mo4-O13	2.390(4)
Mo5-O19	1.714(5)	Mo5-O18	1.714(5)
Mo5-O5	1.901(5)	Mo5-O16	1.937(5)
Mo5-O20	2.289(5)	Mo5-O17	2.373(5)
Cu1-N2	1.966(6)	Cu1-N3	1.979(6)
Cu1-O7	2.022(5)	Cu1-N1	2.048(6)
Cu1-N4	2.071(6)	Cu2-N6	1.985(6)
Cu2-N8	1.993(6)	Cu2-N5	2.027(6)
Cu2-N7	2.052(6)	Cu2-O15	2.093(5)
P1-O4	1.514(5)	P1-O9	1.541(5)

P1-O17	1.550(5)	P1-C41	1.805(7)
P2-O20	1.500(5)	P2-O21	1.547(5)
P2-O13	1.552(5)	P2-C45	1.814(7)
O90-H90A	0.86(2)	O90-H90B	0.85(2)
O91-H91A	0.84(2)	O91-H91B	0.84(2)
O92-H92A	0.96(4)	O92-H92B	0.85(2)
N1-C1	1.335(9)	N1-C5	1.354(9)
N2-C10	1.337(9)	N2-C6	1.342(9)
N3-C11	1.343(9)	N3-C15	1.344(9)
N4-C20	1.335(9)	N4-C16	1.352(9)
N5-C21	1.343(10)	N5-C25	1.344(9)
N6-C30	1.321(10)	N6-C26	1.364(9)
N7-C31	1.340(9)	N7-C35	1.359(9)
N8-C40	1.344(10)	N8-C36	1.348(9)
C1-C2	1.378(10)	C1-H1	0.95
C2-C3	1.375(10)	C2-H2	0.95
C3-C4	1.393(10)	C3-H3	0.95
C4-C5	1.386(10)	C4-H4	0.95
C5-C6	1.476(10)	C6-C7	1.387(9)
C7-C8	1.390(10)	C7-H7	0.95
C8-C9	1.397(10)	C8-H8	0.95
C9-C10	1.368(10)	C9-H9	0.95

C10-H10	0.95	C11-C12	1.379(10)
C11-H11	0.95	C12-C13	1.367(10)
C12-H12	0.95	C13-C14	1.392(11)
C13-H13	0.95	C14-C15	1.377(10)
C14-H14	0.95	C15-C16	1.490(10)
C16-C17	1.392(10)	C17-C18	1.364(11)
C17-H17	0.95	C18-C19	1.391(11)
C18-H18	0.95	C19-C20	1.385(10)
C19-H19	0.95	C20-H20	0.95
C21-C22	1.368(11)	C21-H21	0.95
C22-C23	1.386(13)	C22-H22	0.95
C23-C24	1.380(13)	C23-H23	0.95
C24-C25	1.382(11)	C24-H24	0.95
C25-C26	1.470(11)	C26-C27	1.380(11)
C27-C28	1.391(13)	C27-H27	0.95
C28-C29	1.379(12)	C28-H28	0.95
C29-C30	1.386(11)	C29-H29	0.95
C30-H30	0.95	C31-C32	1.383(10)
C31-H31	0.95	C32-C33	1.380(11)
C32-H32	0.95	C33-C34	1.397(11)
C33-H33	0.95	C34-C35	1.373(10)
C34-H34	0.95	C35-C36	1.475(10)

C36-C37	1.384(10)	C37-C38	1.376(11)
C37-H37	0.95	C38-C39	1.391(11)
C38-H38	0.95	C39-C40	1.382(10)
C39-H39	0.95	C40-H40	0.95
C41-C42	1.519(10)	C41-H41A	0.99
C41-H41B	0.99	C42-C44	1.378(10)
C42-C43	1.395(11)	C43-C44	1.379(11)
C43-H43	0.95	C44-C43	1.379(11)
C44-H44	0.95	C45-C46	1.511(9)
C45-H45A	0.99	C45-H45B	0.99
C46-C48	1.400(10)	C46-C47	1.408(10)
C47-C48	1.388(10)	C47-H47	0.95
C48-C47	1.388(10)	C48-H48	0.95

Table S37. Bond angles (°) for $[\{Cu(2,2\text{-bpy})_2\}_2Mo_5O_{15}(1,4\text{-}O_3PC_8H_8PO_3)]\cdot 6H_2O$ (**5**·$6H_2O$).

O2-Mo1-O1	102.2(2)	O2-Mo1-O5	103.9(2)
O1-Mo1-O5	98.3(2)	O2-Mo1-O3	102.1(2)
O1-Mo1-O3	99.0(2)	O5-Mo1-O3	144.8(2)
O2-Mo1-O4	88.2(2)	O1-Mo1-O4	169.5(2)
O5-Mo1-O4	79.75(19)	O3-Mo1-O4	77.77(19)
O2-Mo1-O21	169.3(2)	O1-Mo1-O21	81.7(2)
O5-Mo1-O21	85.23(18)	O3-Mo1-O21	67.31(17)
O4-Mo1-O21	87.91(16)	O6-Mo2-O7	103.4(2)
O6-Mo2-O3	100.3(2)	O7-Mo2-O3	97.1(2)
O6-Mo2-O8	96.9(2)	O7-Mo2-O8	99.8(2)
O3-Mo2-O8	152.4(2)	O6-Mo2-O21	97.8(2)
O7-Mo2-O21	158.2(2)	O3-Mo2-O21	74.07(18)
O8-Mo2-O21	82.33(18)	O6-Mo2-O9	166.60(18)
O7-Mo2-O9	85.44(19)	O3-Mo2-O9	88.37(17)
O8-Mo2-O9	71.44(17)	O21-Mo2-O9	74.64(16)
O11-Mo3-O10	102.1(2)	O11-Mo3-O12	101.9(2)
O10-Mo3-O12	100.1(2)	O11-Mo3-O8	100.0(2)
O10-Mo3-O8	100.8(2)	O12-Mo3-O8	145.6(2)
O11-Mo3-O9	168.39(19)	O10-Mo3-O9	88.6(2)
O12-Mo3-O9	80.42(18)	O8-Mo3-O9	73.16(17)
O11-Mo3-O13	87.9(2)	O10-Mo3-O13	169.4(2)

O12-Mo3-O13	73.87(18)	O8-Mo3-O13	80.80(17)
O9-Mo3-O13	81.82(16)	O14-Mo4-O15	104.3(2)
O14-Mo4-O16	100.7(2)	O15-Mo4-O16	96.5(2)
O14-Mo4-O12	97.1(2)	O15-Mo4-O12	99.7(2)
O16-Mo4-O12	152.1(2)	O14-Mo4-O17	97.4(2)
O15-Mo4-O17	157.62(19)	O16-Mo4-O17	73.81(18)
O12-Mo4-O17	82.77(18)	O14-Mo4-O13	166.5(2)
O15-Mo4-O13	85.95(19)	O16-Mo4-O13	86.49(18)
O12-Mo4-O13	72.22(17)	O17-Mo4-O13	73.55(16)
O19-Mo5-O18	101.4(2)	O19-Mo5-O5	103.7(2)
O18-Mo5-O5	100.1(2)	O19-Mo5-O16	101.3(2)
O18-Mo5-O16	99.5(2)	O5-Mo5-O16	144.2(2)
O19-Mo5-O20	82.8(2)	O18-Mo5-O20	175.5(2)
O5-Mo5-O20	80.24(19)	O16-Mo5-O20	77.95(18)
O19-Mo5-O17	167.7(2)	O18-Mo5-O17	88.1(2)
O5-Mo5-O17	82.00(18)	O16-Mo5-O17	69.11(18)
O20-Mo5-O17	87.48(16)	N2-Cu1-N3	177.6(2)
N2-Cu1-O7	89.9(2)	N3-Cu1-O7	90.4(2)
N2-Cu1-N1	81.1(2)	N3-Cu1-N1	100.5(2)
O7-Cu1-N1	128.0(2)	N2-Cu1-N4	97.0(2)
N3-Cu1-N4	80.8(2)	O7-Cu1-N4	115.4(2)
N1-Cu1-N4	116.6(2)	N6-Cu2-N8	175.8(2)

N6-Cu2-N5	81.2(3)	N8-Cu2-N5	98.4(2)
N6-Cu2-N7	102.8(2)	N8-Cu2-N7	80.8(2)
N5-Cu2-N7	129.3(2)	N6-Cu2-O15	86.3(2)
N8-Cu2-O15	90.4(2)	N5-Cu2-O15	121.2(2)
N7-Cu2-O15	109.5(2)	O4-P1-O9	111.2(3)
O4-P1-O17	109.3(3)	O9-P1-O17	111.2(3)
O4-P1-C41	109.7(3)	O9-P1-C41	108.5(3)
O17-P1-C41	106.8(3)	O20-P2-O21	109.4(3)
O20-P2-O13	112.3(3)	O21-P2-O13	110.6(2)
O20-P2-C45	110.3(3)	O21-P2-C45	109.0(3)
O13-P2-C45	105.0(3)	Mo2-O3-Mo1	123.0(2)
P1-O4-Mo1	121.7(3)	Mo5-O5-Mo1	149.8(3)
Mo2-O7-Cu1	132.2(3)	Mo2-O8-Mo3	123.6(2)
P1-O9-Mo3	126.8(3)	P1-O9-Mo2	125.8(3)
Mo3-O9-Mo2	91.71(16)	Mo3-O12-Mo4	122.7(2)
P2-O13-Mo3	126.4(3)	P2-O13-Mo4	127.3(3)
Mo3-O13-Mo4	91.15(15)	Mo4-O15-Cu2	129.8(3)
Mo4-O16-Mo5	122.4(2)	P1-O17-Mo4	129.9(3)
P1-O17-Mo5	134.3(3)	Mo4-O17-Mo5	94.53(17)
P2-O20-Mo5	120.8(3)	P2-O21-Mo2	132.6(3)
P2-O21-Mo1	130.5(3)	Mo2-O21-Mo1	94.91(16)
H90A-O90-H90B	118.(10)	H91A-O91-H91B	104.(10)

H92A-O92-H92B	135.(10)	C1-N1-C5	118.7(6)
C1-N1-Cu1	128.1(5)	C5-N1-Cu1	113.0(5)
C10-N2-C6	119.5(6)	C10-N2-Cu1	124.5(5)
C6-N2-Cu1	116.0(5)	C11-N3-C15	119.6(6)
C11-N3-Cu1	124.2(5)	C15-N3-Cu1	116.2(5)
C20-N4-C16	118.2(6)	C20-N4-Cu1	128.8(5)
C16-N4-Cu1	113.1(5)	C21-N5-C25	119.2(7)
C21-N5-Cu2	126.5(5)	C25-N5-Cu2	114.1(5)
C30-N6-C26	119.4(6)	C30-N6-Cu2	125.4(5)
C26-N6-Cu2	114.8(5)	C31-N7-C35	118.9(6)
C31-N7-Cu2	127.2(5)	C35-N7-Cu2	112.7(5)
C40-N8-C36	119.1(6)	C40-N8-Cu2	125.4(5)
C36-N8-Cu2	115.3(5)	N1-C1-C2	122.9(7)
N1-C1-H1	118.6	C2-C1-H1	118.6
C3-C2-C1	119.0(7)	C3-C2-H2	120.5
C1-C2-H2	120.5	C2-C3-C4	118.9(7)
C2-C3-H3	120.5	C4-C3-H3	120.5
C5-C4-C3	119.1(7)	C5-C4-H4	120.4
C3-C4-H4	120.4	N1-C5-C4	121.4(7)
N1-C5-C6	114.7(6)	C4-C5-C6	124.0(6)
N2-C6-C7	121.0(6)	N2-C6-C5	115.1(6)
C7-C6-C5	123.9(6)	C6-C7-C8	119.7(6)

C6-C7-H7	120.2	C8-C7-H7	120.2
C7-C8-C9	118.0(7)	C7-C8-H8	121.0
C9-C8-H8	121.0	C10-C9-C8	119.1(7)
C10-C9-H9	120.5	C8-C9-H9	120.5
N2-C10-C9	122.5(7)	N2-C10-H10	118.7
C9-C10-H10	118.7	N3-C11-C12	121.7(7)
N3-C11-H11	119.1	C12-C11-H11	119.1
C13-C12-C11	119.2(7)	C13-C12-H12	120.4
C11-C12-H12	120.4	C12-C13-C14	119.1(7)
C12-C13-H13	120.4	C14-C13-H13	120.4
C15-C14-C13	119.3(7)	C15-C14-H14	120.3
C13-C14-H14	120.3	N3-C15-C14	121.0(7)
N3-C15-C16	115.0(6)	C14-C15-C16	124.0(6)
N4-C16-C17	121.6(7)	N4-C16-C15	114.7(6)
C17-C16-C15	123.7(7)	C18-C17-C16	119.3(7)
C18-C17-H17	120.3	C16-C17-H17	120.3
C17-C18-C19	119.8(7)	C17-C18-H18	120.1
C19-C18-H18	120.1	C20-C19-C18	117.6(7)
C20-C19-H19	121.2	C18-C19-H19	121.2
N4-C20-C19	123.5(7)	N4-C20-H20	118.2
C19-C20-H20	118.2	N5-C21-C22	122.4(8)
N5-C21-H21	118.8	C22-C21-H21	118.8

C21-C22-C23	118.5(9)	C21-C22-H22	120.7
C23-C22-H22	120.7	C24-C23-C22	119.4(8)
C24-C23-H23	120.3	C22-C23-H23	120.3
C23-C24-C25	119.1(8)	C23-C24-H24	120.4
C25-C24-H24	120.4	N5-C25-C24	121.3(8)
N5-C25-C26	115.0(7)	C24-C25-C26	123.7(7)
N6-C26-C27	121.0(7)	N6-C26-C25	114.8(6)
C27-C26-C25	124.2(7)	C26-C27-C28	118.9(8)
C26-C27-H27	120.6	C28-C27-H27	120.6
C29-C28-C27	119.7(8)	C29-C28-H28	120.1
C27-C28-H28	120.1	C28-C29-C30	118.2(8)
C28-C29-H29	120.9	C30-C29-H29	120.9
N6-C30-C29	122.8(8)	N6-C30-H30	118.6
C29-C30-H30	118.6	N7-C31-C32	122.3(7)
N7-C31-H31	118.8	C32-C31-H31	118.8
C33-C32-C31	118.6(7)	C33-C32-H32	120.7
C31-C32-H32	120.7	C32-C33-C34	119.5(7)
C32-C33-H33	120.2	C34-C33-H33	120.2
C35-C34-C33	118.7(7)	C35-C34-H34	120.7
C33-C34-H34	120.7	N7-C35-C34	121.9(7)
N7-C35-C36	114.4(6)	C34-C35-C36	123.7(6)
N8-C36-C37	121.5(7)	N8-C36-C35	115.1(6)

C37-C36-C35	123.4(6)	C38-C37-C36	119.4(7)
C38-C37-H37	120.3	C36-C37-H37	120.3
C37-C38-C39	119.1(7)	C37-C38-H38	120.4
C39-C38-H38	120.4	C40-C39-C38	118.7(7)
C40-C39-H39	120.6	C38-C39-H39	120.6
N8-C40-C39	122.1(7)	N8-C40-H40	119.0
C39-C40-H40	119.0	C42-C41-P1	111.8(5)
C42-C41-H41A	109.3	P1-C41-H41A	109.3
C42-C41-H41B	109.3	P1-C41-H41B	109.3
H41A-C41-H41B	107.9	C44-C42-C43	118.6(7)
C44-C42-C41	121.0(7)	C43-C42-C41	120.3(6)
C44-C43-C42	119.8(7)	C44-C43-H43	120.1
C42-C43-H43	120.1	C42-C44-C43	121.6(7)
C42-C44-H44	119.2	C43-C44-H44	119.2
C46-C45-P2	118.7(5)	C46-C45-H45A	107.6
P2-C45-H45A	107.7	C46-C45-H45B	107.6
P2-C45-H45B	107.6	H45A-C45-H45B	107.1
C48-C46-C47	117.2(6)	C48-C46-C45	122.2(6)
C47-C46-C45	120.5(6)	C48-C47-C46	121.3(7)
C48-C47-H47	119.4	C46-C47-H47	119.4
C47-C48-C46	121.5(7)	C47-C48-H48	119.3
C46-C48-H48	119.3		

Table S38. Torsion angles (°) for $[\{Cu(2,2\text{-bpy})_2\}_2Mo_5O_{15}(1,4\text{-}O_3PC_8H_8PO_3)] \cdot 6H_2O$ (**5**·$6H_2O$).

O9-P1-O4-Mo1	-63.0(4)	O17-P1-O4-Mo1	60.2(4)
C41-P1-O4-Mo1	177.0(3)	O6-Mo2-O7-Cu1	-7.3(4)
O3-Mo2-O7-Cu1	95.1(3)	O8-Mo2-O7-Cu1	-106.9(3)
O21-Mo2-O7-Cu1	159.2(4)	O9-Mo2-O7-Cu1	-177.2(3)
O4-P1-O9-Mo3	152.6(3)	O17-P1-O9-Mo3	30.6(4)
C41-P1-O9-Mo3	-86.6(4)	O4-P1-O9-Mo2	26.8(4)
O17-P1-O9-Mo2	-95.2(3)	C41-P1-O9-Mo2	147.5(3)
O20-P2-O13-Mo3	152.6(3)	O21-P2-O13-Mo3	30.0(4)
C45-P2-O13-Mo3	-87.5(4)	O20-P2-O13-Mo4	26.2(4)
O21-P2-O13-Mo4	-96.4(3)	C45-P2-O13-Mo4	146.1(3)
O14-Mo4-O15-Cu2	-6.7(4)	O16-Mo4-O15-Cu2	96.1(3)
O12-Mo4-O15-Cu2	-106.7(3)	O17-Mo4-O15-Cu2	158.7(4)
O13-Mo4-O15-Cu2	-177.9(3)	O14-Mo4-O16-Mo5	-97.9(3)
O15-Mo4-O16-Mo5	156.1(3)	O12-Mo4-O16-Mo5	30.9(6)
O17-Mo4-O16-Mo5	-3.3(2)	O13-Mo4-O16-Mo5	70.6(3)
O4-P1-O17-Mo4	-172.0(3)	O9-P1-O17-Mo4	-48.9(4)
C41-P1-O17-Mo4	69.4(4)	O4-P1-O17-Mo5	-8.3(4)
O9-P1-O17-Mo5	114.8(4)	C41-P1-O17-Mo5	-126.9(4)
O21-P2-O20-Mo5	65.7(3)	O13-P2-O20-Mo5	-57.6(3)
C45-P2-O20-Mo5	-174.3(3)	O20-P2-O21-Mo2	-171.5(3)
O13-P2-O21-Mo2	-47.2(4)	C45-P2-O21-Mo2	67.8(4)

O20-P2-O21-Mo1	-11.8(4)	O13-P2-O21-Mo1	112.5(3)
C45-P2-O21-Mo1	-132.5(3)	C5-N1-C1-C2	-0.3(11)
Cu1-N1-C1-C2	-174.6(6)	N1-C1-C2-C3	0.5(12)
C1-C2-C3-C4	-0.4(11)	C2-C3-C4-C5	0.1(10)
C1-N1-C5-C4	-0.1(10)	Cu1-N1-C5-C4	175.1(5)
C1-N1-C5-C6	-179.2(6)	Cu1-N1-C5-C6	-4.0(7)
C3-C4-C5-N1	0.1(10)	C3-C4-C5-C6	179.2(6)
C10-N2-C6-C7	-1.4(10)	Cu1-N2-C6-C7	178.2(5)
C10-N2-C6-C5	178.3(6)	Cu1-N2-C6-C5	-2.0(7)
N1-C5-C6-N2	4.1(8)	C4-C5-C6-N2	-175.0(6)
N1-C5-C6-C7	-176.2(6)	C4-C5-C6-C7	4.7(10)
N2-C6-C7-C8	3.8(10)	C5-C6-C7-C8	-175.9(6)
C6-C7-C8-C9	-2.7(10)	C7-C8-C9-C10	-0.5(11)
C6-N2-C10-C9	-2.0(11)	Cu1-N2-C10-C9	178.3(6)
C8-C9-C10-N2	3.0(12)	C15-N3-C11-C12	-1.3(11)
Cu1-N3-C11-C12	178.0(6)	N3-C11-C12-C13	-0.1(12)
C11-C12-C13-C14	0.0(11)	C12-C13-C14-C15	1.5(11)
C11-N3-C15-C14	2.9(10)	Cu1-N3-C15-C14	-176.6(5)
C11-N3-C15-C16	-176.0(6)	Cu1-N3-C15-C16	4.6(8)
C13-C14-C15-N3	-2.9(11)	C13-C14-C15-C16	175.8(7)
C20-N4-C16-C17	2.0(10)	Cu1-N4-C16-C17	-177.1(5)
C20-N4-C16-C15	-177.8(6)	Cu1-N4-C16-C15	3.1(7)

N3-C15-C16-N4	-5.1(9)	C14-C15-C16-N4	176.2(6)
N3-C15-C16-C17	175.1(7)	C14-C15-C16-C17	-3.7(11)
N4-C16-C17-C18	-2.9(11)	C15-C16-C17-C18	176.9(7)
C16-C17-C18-C19	1.6(11)	C17-C18-C19-C20	0.4(10)
C16-N4-C20-C19	0.1(11)	Cu1-N4-C20-C19	179.0(5)
C18-C19-C20-N4	-1.3(11)	C25-N5-C21-C22	-1.7(14)
Cu2-N5-C21-C22	-176.8(7)	N5-C21-C22-C23	0.9(15)
C21-C22-C23-C24	0.5(15)	C22-C23-C24-C25	-1.1(14)
C21-N5-C25-C24	1.1(12)	Cu2-N5-C25-C24	176.8(6)
C21-N5-C25-C26	-178.6(7)	Cu2-N5-C25-C26	-2.9(8)
C23-C24-C25-N5	0.3(12)	C23-C24-C25-C26	179.9(8)
C30-N6-C26-C27	2.4(10)	Cu2-N6-C26-C27	175.9(6)
C30-N6-C26-C25	-176.6(6)	Cu2-N6-C26-C25	-3.0(8)
N5-C25-C26-N6	3.9(9)	C24-C25-C26-N6	-175.7(7)
N5-C25-C26-C27	-175.0(7)	C24-C25-C26-C27	5.4(12)
N6-C26-C27-C28	-2.1(12)	C25-C26-C27-C28	176.7(7)
C26-C27-C28-C29	0.9(13)	C27-C28-C29-C30	0.0(13)
C26-N6-C30-C29	-1.5(11)	Cu2-N6-C30-C29	-174.2(6)
C28-C29-C30-N6	0.3(12)	C35-N7-C31-C32	0.9(10)
Cu2-N7-C31-C32	-165.2(6)	N7-C31-C32-C33	2.4(11)
C31-C32-C33-C34	-3.4(11)	C32-C33-C34-C35	1.3(11)
C31-N7-C35-C34	-3.3(10)	Cu2-N7-C35-C34	164.7(5)

C31-N7-C35-C36	177.3(6)	Cu2-N7-C35-C36	-14.7(7)
C33-C34-C35-N7	2.2(10)	C33-C34-C35-C36	-178.4(6)
C40-N8-C36-C37	1.3(10)	Cu2-N8-C36-C37	177.0(5)
C40-N8-C36-C35	-178.2(6)	Cu2-N8-C36-C35	-2.5(8)
N7-C35-C36-N8	11.7(9)	C34-C35-C36-N8	-167.8(6)
N7-C35-C36-C37	-167.8(7)	C34-C35-C36-C37	12.8(11)
N8-C36-C37-C38	0.4(11)	C35-C36-C37-C38	179.8(7)
C36-C37-C38-C39	-1.2(12)	C37-C38-C39-C40	0.4(12)
C36-N8-C40-C39	-2.1(11)	Cu2-N8-C40-C39	-177.4(6)
C38-C39-C40-N8	1.3(12)	O4-P1-C41-C42	-45.2(6)
O9-P1-C41-C42	-166.8(5)	O17-P1-C41-C42	73.2(6)
P1-C41-C42-C44	105.7(7)	P1-C41-C42-C43	-71.5(8)
C44-C42-C43-C44	-0.7(12)	C41-C42-C43-C44	176.6(7)
C43-C42-C44-C43	0.7(12)	C41-C42-C44-C43	-176.6(7)
O20-P2-C45-C46	-54.7(6)	O21-P2-C45-C46	65.5(6)
O13-P2-C45-C46	-175.9(5)	P2-C45-C46-C48	-84.8(8)
P2-C45-C46-C47	98.3(7)	C48-C46-C47-C48	-1.1(12)
C45-C46-C47-C48	175.9(7)	C47-C46-C48-C47	1.1(12)
C45-C46-C48-C47	-175.9(7)		

Table S39. Anisotropic atomic displacement parameters ($Å^2$) for $[\{Cu(2,2\text{-bpy})_2\}_2Mo_5O_{15}(1,4\text{-}O_3PC_8H_8PO_3)] \cdot 6H_2O$ (**5**·$6H_2O$).

The anisotropic atomic displacement factor exponent takes the form: $-2\pi^2[h^2 a^{*2} U_{11} + ... + 2 h k a^* b^* U_{12}]$

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
Mo1	0.0088(3)	0.0105(3)	0.0170(3)	-0.0049(2)	-0.0020(3)	-0.0017(3)
Mo2	0.0085(3)	0.0075(3)	0.0083(3)	-0.0011(2)	0.0001(2)	-0.0033(2)
Mo3	0.0098(3)	0.0079(3)	0.0105(3)	-0.0018(2)	0.0028(2)	-0.0035(2)
Mo4	0.0156(3)	0.0091(3)	0.0091(3)	-0.0022(2)	0.0025(2)	-0.0058(3)
Mo5	0.0135(3)	0.0106(3)	0.0139(3)	-0.0026(2)	-0.0035(3)	-0.0040(3)
Cu1	0.0114(4)	0.0079(4)	0.0127(4)	0.0002(3)	-0.0010(3)	-0.0030(4)
Cu2	0.0140(5)	0.0143(4)	0.0111(4)	-0.0025(3)	0.0021(4)	-0.0090(4)
P1	0.0155(10)	0.0084(8)	0.0084(9)	0.0007(7)	-0.0012(7)	-0.0056(7)
P2	0.0090(9)	0.0084(8)	0.0083(9)	-0.0009(7)	0.0010(7)	-0.0038(7)
O1	0.014(3)	0.017(3)	0.025(3)	-0.011(2)	0.004(2)	-0.007(2)
O2	0.016(3)	0.011(2)	0.025(3)	-0.004(2)	-0.006(2)	0.000(2)
O3	0.013(3)	0.008(2)	0.010(2)	-0.0019(19)	-0.001(2)	-0.003(2)
O4	0.023(3)	0.009(2)	0.010(2)	-0.0003(19)	0.001(2)	-0.008(2)
O5	0.011(3)	0.015(2)	0.022(3)	-0.009(2)	-0.001(2)	-0.005(2)
O6	0.014(3)	0.013(2)	0.013(3)	-0.004(2)	0.002(2)	-0.006(2)
O7	0.012(3)	0.012(2)	0.014(3)	-0.004(2)	0.001(2)	-0.006(2)
O8	0.009(2)	0.009(2)	0.007(2)	0.0005(18)	-0.0008(19)	-0.003(2)
O9	0.011(3)	0.007(2)	0.012(2)	0.0025(19)	-0.004(2)	-0.002(2)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
O10	0.018(3)	0.017(3)	0.016(3)	-0.005(2)	0.006(2)	-0.010(2)
O11	0.013(3)	0.014(2)	0.017(3)	-0.006(2)	0.003(2)	-0.005(2)
O12	0.018(3)	0.013(2)	0.013(3)	-0.005(2)	0.004(2)	-0.009(2)
O13	0.011(2)	0.008(2)	0.009(2)	-0.0010(18)	0.0012(19)	-0.005(2)
O14	0.024(3)	0.020(3)	0.012(3)	-0.005(2)	0.003(2)	-0.011(2)
O15	0.015(3)	0.015(2)	0.015(3)	-0.007(2)	0.006(2)	-0.008(2)
O16	0.014(3)	0.012(2)	0.012(3)	-0.005(2)	0.001(2)	-0.005(2)
O17	0.016(3)	0.013(2)	0.008(2)	-0.0034(19)	0.001(2)	-0.007(2)
O18	0.034(3)	0.018(3)	0.014(3)	-0.001(2)	-0.010(2)	-0.012(3)
O19	0.013(3)	0.016(3)	0.023(3)	-0.002(2)	-0.006(2)	-0.007(2)
O20	0.012(3)	0.013(2)	0.012(2)	-0.002(2)	-0.001(2)	-0.007(2)
O21	0.011(2)	0.010(2)	0.012(2)	-0.0029(19)	0.002(2)	-0.005(2)
O90	0.038(5)	0.045(5)	0.085(6)	0.024(4)	-0.008(4)	-0.017(4)
O91	0.023(4)	0.064(6)	0.082(7)	0.010(5)	-0.014(4)	0.002(4)
O92	0.080(6)	0.049(5)	0.075(6)	0.003(5)	0.032(5)	-0.024(5)
N1	0.013(3)	0.008(3)	0.019(3)	-0.005(2)	0.003(3)	-0.004(3)
N2	0.011(3)	0.013(3)	0.015(3)	-0.003(2)	-0.001(2)	-0.007(3)
N3	0.013(3)	0.012(3)	0.013(3)	0.001(2)	-0.001(2)	-0.004(3)
N4	0.013(3)	0.012(3)	0.013(3)	-0.002(2)	-0.001(2)	-0.005(3)
N5	0.023(4)	0.029(4)	0.011(3)	-0.002(3)	0.003(3)	-0.016(3)
N6	0.015(3)	0.011(3)	0.020(3)	-0.003(3)	0.000(3)	-0.005(3)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
N7	0.017(3)	0.017(3)	0.010(3)	-0.005(2)	0.003(3)	-0.011(3)
N8	0.020(3)	0.022(3)	0.008(3)	-0.006(3)	0.003(3)	-0.014(3)
C1	0.012(4)	0.013(4)	0.021(4)	0.001(3)	-0.006(3)	-0.001(3)
C2	0.013(4)	0.020(4)	0.024(4)	-0.003(3)	-0.003(3)	-0.007(3)
C3	0.019(4)	0.017(4)	0.020(4)	-0.007(3)	0.004(3)	-0.011(3)
C4	0.018(4)	0.015(4)	0.011(4)	-0.002(3)	0.000(3)	-0.010(3)
C5	0.015(4)	0.015(4)	0.006(3)	-0.006(3)	0.003(3)	-0.006(3)
C6	0.013(4)	0.010(3)	0.005(3)	-0.003(3)	-0.001(3)	-0.004(3)
C7	0.022(4)	0.011(3)	0.012(4)	-0.003(3)	0.001(3)	-0.008(3)
C8	0.010(4)	0.009(3)	0.021(4)	-0.005(3)	0.002(3)	0.000(3)
C9	0.017(4)	0.015(4)	0.022(4)	-0.003(3)	-0.002(3)	-0.007(3)
C10	0.018(4)	0.017(4)	0.020(4)	0.002(3)	0.001(3)	-0.012(3)
C11	0.022(4)	0.015(4)	0.013(4)	-0.001(3)	0.001(3)	-0.005(3)
C12	0.013(4)	0.014(4)	0.017(4)	-0.002(3)	0.001(3)	-0.003(3)
C13	0.019(4)	0.014(4)	0.023(4)	0.005(3)	-0.010(3)	-0.006(3)
C14	0.020(4)	0.017(4)	0.014(4)	0.002(3)	-0.002(3)	-0.013(3)
C15	0.022(4)	0.012(3)	0.013(4)	-0.001(3)	0.000(3)	-0.012(3)
C16	0.015(4)	0.014(4)	0.018(4)	-0.004(3)	-0.002(3)	-0.010(3)
C17	0.022(4)	0.022(4)	0.017(4)	-0.003(3)	0.005(3)	-0.017(3)
C18	0.025(4)	0.033(4)	0.009(4)	-0.006(3)	0.003(3)	-0.024(4)
C19	0.019(4)	0.030(4)	0.022(4)	-0.015(4)	0.004(3)	-0.015(4)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
C20	0.014(4)	0.019(4)	0.019(4)	-0.004(3)	-0.001(3)	-0.008(3)
C21	0.043(6)	0.056(6)	0.016(4)	-0.010(4)	0.006(4)	-0.037(5)
C22	0.043(6)	0.071(7)	0.010(4)	-0.008(4)	0.004(4)	-0.034(6)
C23	0.052(6)	0.048(6)	0.016(4)	-0.002(4)	0.003(4)	-0.029(5)
C24	0.028(5)	0.028(4)	0.018(4)	-0.002(3)	0.008(4)	-0.012(4)
C25	0.018(4)	0.017(4)	0.024(4)	-0.004(3)	0.005(3)	-0.008(3)
C26	0.018(4)	0.009(3)	0.021(4)	0.000(3)	0.004(3)	-0.004(3)
C27	0.020(4)	0.028(4)	0.031(5)	0.001(4)	0.007(4)	-0.013(4)
C28	0.025(5)	0.037(5)	0.046(6)	0.005(4)	-0.002(4)	-0.023(4)
C29	0.022(5)	0.033(5)	0.043(6)	-0.002(4)	-0.007(4)	-0.016(4)
C30	0.015(4)	0.019(4)	0.025(4)	-0.003(3)	0.000(3)	-0.008(3)
C31	0.018(4)	0.020(4)	0.016(4)	-0.003(3)	0.001(3)	-0.012(3)
C32	0.014(4)	0.021(4)	0.023(4)	-0.009(3)	0.002(3)	-0.007(3)
C33	0.030(5)	0.016(4)	0.013(4)	-0.002(3)	0.002(3)	-0.012(4)
C34	0.023(4)	0.020(4)	0.011(4)	-0.006(3)	0.009(3)	-0.017(3)
C35	0.014(4)	0.020(4)	0.009(3)	-0.009(3)	0.003(3)	-0.008(3)
C36	0.018(4)	0.016(4)	0.012(4)	-0.006(3)	0.004(3)	-0.011(3)
C37	0.022(4)	0.021(4)	0.024(4)	-0.010(3)	0.002(3)	-0.013(4)
C38	0.024(5)	0.034(5)	0.025(4)	-0.009(4)	0.007(4)	-0.021(4)
C39	0.014(4)	0.024(4)	0.027(4)	-0.009(3)	-0.003(3)	-0.009(3)
C40	0.018(4)	0.017(4)	0.019(4)	-0.005(3)	0.001(3)	-0.008(3)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
C41	0.018(4)	0.015(4)	0.012(4)	0.001(3)	-0.001(3)	-0.006(3)
C42	0.022(4)	0.018(4)	0.005(3)	-0.001(3)	0.002(3)	-0.007(3)
C43	0.039(5)	0.010(3)	0.013(4)	-0.002(3)	-0.003(4)	-0.011(4)
C44	0.035(5)	0.019(4)	0.008(4)	-0.005(3)	0.003(3)	-0.015(4)
C45	0.013(4)	0.013(3)	0.007(3)	0.002(3)	0.000(3)	-0.006(3)
C46	0.010(4)	0.011(3)	0.011(3)	0.000(3)	-0.004(3)	-0.003(3)
C47	0.022(4)	0.017(4)	0.015(4)	-0.008(3)	0.003(3)	-0.008(3)
C48	0.020(4)	0.015(4)	0.023(4)	-0.005(3)	0.003(3)	-0.012(3)

Table S40. Hydrogen atomic coordinates and isotropic atomic displacement parameters ($Å^2$) for $[\{Cu(2,2\text{-bpy})_2\}_2Mo_5O_{15}(1,4\text{-}O_3PC_8H_8PO_3)] \cdot 6H_2O$ (**5**·$6H_2O$).

	x/a	y/b	z/c	U(eq)
H90A	0.789(11)	0.763(8)	0.395(5)	0.077
H90B	0.829(7)	0.640(6)	0.435(6)	0.077
H91A	0.743(12)	0.465(10)	0.271(6)	0.085
H91B	0.647(3)	0.456(11)	0.285(6)	0.085
H92A	0.800(12)	0.620(8)	0.351(7)	0.092
H92B	0.792(13)	0.499(5)	0.345(7)	0.092
H1	0.5794	0.8770	0.4063	0.022
H2	0.6761	0.9891	0.3851	0.024
H3	0.5713	1.1947	0.3346	0.02
H4	0.3699	1.2817	0.3051	0.016
H7	0.1870	1.3431	0.2732	0.018
H8	-0.0146	1.4044	0.2607	0.018
H9	-0.0769	1.2596	0.3010	0.022
H10	0.0608	1.0641	0.3493	0.021
H11	0.5398	0.7079	0.3643	0.023
H12	0.6754	0.5199	0.4254	0.02
H13	0.6499	0.4653	0.5370	0.024
H14	0.4879	0.6030	0.5857	0.02
H17	0.3143	0.7273	0.6192	0.022

	x/a	y/b	z/c	U(eq)
H18	0.1462	0.8816	0.6511	0.022
H19	0.0384	1.0667	0.5746	0.025
H20	0.1046	1.0874	0.4686	0.021
H21	0.2519	0.2638	0.0019	0.04
H22	0.2608	0.3165	-0.1107	0.047
H23	0.4034	0.3710	-0.1542	0.046
H24	0.5342	0.3680	-0.0826	0.031
H27	0.6409	0.3736	-0.0085	0.034
H28	0.7469	0.3710	0.0780	0.042
H29	0.7097	0.3049	0.1867	0.038
H30	0.5666	0.2450	0.2061	0.024
H31	0.6268	0.0307	0.2014	0.02
H32	0.7031	-0.1738	0.2495	0.024
H33	0.5821	-0.2611	0.2541	0.024
H34	0.3811	-0.1363	0.2201	0.018
H37	0.1916	0.0011	0.2085	0.024
H38	-0.0003	0.1487	0.1702	0.03
H39	-0.0351	0.3413	0.1114	0.025
H40	0.1228	0.3789	0.0925	0.022
H41A	0.2264	0.9040	0.1074	0.019
H41B	0.2528	0.7837	0.0862	0.019

	x/a	y/b	z/c	U(eq)
H43	0.0753	0.7914	0.0319	0.025
H44	0.0808	1.0916	0.0453	0.024
H45A	0.2006	0.3857	0.3900	0.014
H45B	0.2360	0.4726	0.4127	0.014
H47	0.0352	0.3642	0.4352	0.021
H48	0.0922	0.6183	0.4794	0.021

Table S41. Sample and crystal data for $[\{Cu(o\text{-}phen)(H_2O)_2\}_2Mo_5O_{15}(1,4\text{-}O_3PC_8H_8PO_3)]\cdot 4H_2O$ (**6**·$4H_2O$).

Identification code	$[\{Cu(o\text{-}phen)(H_2O)_2\}_2Mo_5O_{15}(1,4\text{-}O_3PC_8H_8PO_3)]\cdot 4H_2O$	
Chemical formula	$C_{32}H_{40}Cu_2Mo_5N_4O_{29}P_2$	
Formula weight	1613.40	
Temperature	90(2) K	
Wavelength	0.71073 Å	
Crystal size	0.127 x 0.147 x 0.332 mm	
Crystal system	Monoclinic	
Space group	P21/c	
Unit cell dimensions	a = 13.4404(14) Å	α = 90°
	b = 21.237(2) Å	β = 118.357(6)°
	c = 18.1894(14) Å	γ = 90°
Volume	4568.9(8) $Å^3$	
Z	4	
Density (calculated)	2.346 g/cm^3	
Absorption coefficient	2.416 mm^{-1}	
F(000)	3160	

Table S42. Data collection and structure refinement for [{Cu(o-phen)(H_2O)$_2$}$_2$$Mo_5O_{15}$(1,4-$O_3PC_8H_8PO_3$)]·4$H_2O$ (**6**·4H_2O).

Theta range for data collection	1.72 to 34.16°	
Index ranges	-21<=h<=21, -29<=k<=32, -28<=l<=28	
Reflections collected	86173	
Independent reflections	17963 [R(int) = 0.0355]	
Coverage of independent reflections	95.1%	
Max. and min. transmission	0.7490 and 0.5010	
Structure solution technique	direct methods	
Structure solution program	SHELXS-97 (Sheldrick, 2008)	
Refinement method	Full-matrix least-squares on F^2	
Refinement program	SHELXL-2013 (Sheldrick, 2013)	
Function minimized	$\Sigma\, w(F_o^2 - F_c^2)^2$	
Data / restraints / parameters	17963 / 14 / 715	
Goodness-of-fit on F^2	1.108	
Δ/σ_{max}	0.004	
Final R indices	14688 data; I>2σ(I)	R1 = 0.0350, wR2 = 0.0743
	all data	R1 = 0.0505, wR2 = 0.0822
Weighting scheme	$w=1/[\sigma^2(F_o^2)+(0.0198P)^2+21.3187P]$ where $P=(F_o^2+2F_c^2)/3$	
Largest diff. peak and hole	1.875 and -1.210 eÅ^{-3}	
R.M.S. deviation from mean	0.178 eÅ^{-3}	

Table S43. Atomic coordinates and equivalent isotropic atomic displacement parameters ($Å^2$) for $[\{Cu(o\text{-}phen)(H_2O)_2\}_2Mo_5O_{15}(1,4\text{-}O_3PC_8H_8PO_3)]\cdot 4H_2O$ (**6**·$4H_2O$).

U(eq) is defined as one third of the trace of the orthogonalized U_{ij} tensor.

	x/a	**y/b**	**z/c**	**U(eq)**
Mo1	0.25011(2)	0.71780(2)	0.88020(2)	0.00818(4)
Mo2	0.32390(2)	0.63808(2)	0.75412(2)	0.00868(4)
Mo3	0.10166(2)	0.55259(2)	0.61561(2)	0.00798(4)
Mo4	0.87314(2)	0.59728(2)	0.63040(2)	0.00813(4)
Mo5	0.98604(2)	0.67466(2)	0.83768(2)	0.00787(4)
Cu1	0.82358(3)	0.81329(2)	0.82056(2)	0.01037(6)
Cu2	0.73862(3)	0.45654(2)	0.64505(2)	0.01020(6)
P1	0.05749(6)	0.70785(3)	0.67279(4)	0.00787(11)
P2	0.85937(6)	0.06728(3)	0.68598(4)	0.00746(11)
O1	0.13779(16)	0.69560(10)	0.91600(12)	0.0089(3)
O2	0.23915(18)	0.79772(10)	0.88380(14)	0.0127(4)
O3	0.36770(17)	0.70083(10)	0.97281(13)	0.0122(4)
O4	0.30727(17)	0.71478(10)	0.80218(13)	0.0106(4)
O5	0.08426(16)	0.70744(10)	0.76547(12)	0.0097(3)
O6	0.45059(17)	0.61062(11)	0.83144(14)	0.0141(4)
O7	0.35760(18)	0.66741(11)	0.68094(14)	0.0152(4)
O8	0.25296(16)	0.56045(10)	0.70544(12)	0.0098(3)
O9	0.13679(16)	0.66277(10)	0.65799(12)	0.0092(3)

	x/a	y/b	z/c	U(eq)
O10	0.12548(17)	0.56748(10)	0.53275(13)	0.0118(4)
O11	0.08975(17)	0.47286(10)	0.61600(13)	0.0123(4)
O12	0.94230(16)	0.57670(10)	0.56471(12)	0.0097(3)
O13	0.84219(17)	0.52125(10)	0.65185(13)	0.0123(4)
O14	0.74263(17)	0.62150(11)	0.55812(13)	0.0135(4)
O15	0.93416(16)	0.69050(10)	0.61860(12)	0.0097(3)
O16	0.88748(16)	0.63637(10)	0.73122(12)	0.0095(3)
O17	0.93417(18)	0.64604(11)	0.89938(13)	0.0131(4)
O18	0.92557(17)	0.75035(10)	0.81644(13)	0.0111(4)
O19	0.9422(2)	0.84512(13)	0.92713(16)	0.0225(5)
O20	0.7712(2)	0.75436(14)	0.90043(19)	0.0259(5)
O21	0.92830(16)	0.08313(10)	0.64258(12)	0.0092(3)
O22	0.93884(16)	0.06640(10)	0.78116(12)	0.0087(3)
O23	0.76055(16)	0.11392(10)	0.66258(13)	0.0095(3)
O24	0.74350(19)	0.41630(11)	0.54727(14)	0.0145(4)
O25	0.5922(2)	0.52392(15)	0.5789(2)	0.0396(8)
O90	0.6724(2)	0.86250(17)	0.3564(2)	0.0344(7)
O91	0.6545(2)	0.87676(15)	0.19520(19)	0.0270(6)
O92	0.5231(2)	0.30168(14)	0.40767(17)	0.0228(5)
O93	0.1520(2)	0.69726(12)	0.50521(15)	0.0172(4)
N1	0.7126(2)	0.87998(12)	0.81167(16)	0.0119(4)

	x/a	y/b	z/c	U(eq)
N2	0.7327(2)	0.81422(12)	0.69718(16)	0.0122(4)
N3	0.6314(2)	0.39292(12)	0.64591(15)	0.0110(4)
N4	0.7799(2)	0.46410(12)	0.76555(15)	0.0112(4)
C1	0.7042(2)	0.91058(15)	0.87198(19)	0.0149(5)
C2	0.6239(3)	0.95887(15)	0.8544(2)	0.0166(5)
C3	0.5541(3)	0.97617(15)	0.7735(2)	0.0178(6)
C4	0.6413(2)	0.89567(14)	0.73121(18)	0.0113(5)
C5	0.5617(2)	0.94485(15)	0.7080(2)	0.0145(5)
C6	0.4951(2)	0.95893(15)	0.6207(2)	0.0169(6)
C7	0.5062(3)	0.92533(16)	0.5611(2)	0.0175(6)
C8	0.5866(2)	0.87490(15)	0.58402(18)	0.0135(5)
C9	0.6527(2)	0.86044(14)	0.66888(18)	0.0117(5)
C10	0.7514(2)	0.78235(15)	0.64178(19)	0.0142(5)
C11	0.6903(3)	0.79457(16)	0.55578(19)	0.0164(5)
C12	0.6070(3)	0.84003(16)	0.52659(19)	0.0164(6)
C13	0.5498(2)	0.36202(15)	0.58267(19)	0.0141(5)
C14	0.4832(2)	0.31698(15)	0.5948(2)	0.0171(6)
C15	0.5032(2)	0.30180(15)	0.6738(2)	0.0170(6)
C16	0.5893(2)	0.33385(14)	0.7427(2)	0.0138(5)
C17	0.6486(2)	0.38027(14)	0.72420(18)	0.0113(5)
C18	0.7301(2)	0.41882(14)	0.78984(18)	0.0112(5)

	x/a	y/b	z/c	U(eq)
C19	0.6174(3)	0.32380(15)	0.8283(2)	0.0162(5)
C20	0.6967(3)	0.35931(15)	0.8906(2)	0.0161(5)
C21	0.7539(2)	0.40943(15)	0.87249(18)	0.0133(5)
C22	0.8303(3)	0.45163(15)	0.93252(18)	0.0148(5)
C23	0.8768(3)	0.49905(15)	0.90704(19)	0.0156(5)
C24	0.8514(2)	0.50342(15)	0.82283(18)	0.0135(5)
C25	0.0844(2)	0.78609(13)	0.64805(18)	0.0106(5)
C26	0.0096(2)	0.83855(13)	0.65122(17)	0.0096(4)
C27	0.9309(2)	0.86740(14)	0.57722(18)	0.0118(5)
C28	0.0186(2)	0.86041(14)	0.72660(18)	0.0117(5)
C29	0.8629(2)	0.91633(14)	0.57856(18)	0.0118(5)
C30	0.9518(2)	0.91007(14)	0.72811(18)	0.0121(5)
C31	0.8728(2)	0.93855(13)	0.65403(18)	0.0099(4)
C32	0.7958(2)	0.99082(13)	0.65348(18)	0.0102(5)

Table S44. Bond lengths (Å) for [{Cu(o-phen)(H_2O)$_2$}$_2$$Mo_5O_{15}$(1,4-$O_3PC_8H_8PO_3$)]·4$H_2O$ (**6**·4H_2O).

Mo1-O2	1.708(2)	Mo1-O3	1.714(2)
Mo1-O4	1.909(2)	Mo1-O1	1.964(2)
Mo1-O5	2.226(2)	Mo1-O23	2.321(2)
Mo2-O7	1.714(2)	Mo2-O6	1.715(2)
Mo2-O8	1.900(2)	Mo2-O4	1.911(2)
Mo2-O9	2.341(2)	Mo2-O23	2.341(2)
Mo3-O11	1.701(2)	Mo3-O10	1.713(2)
Mo3-O8	1.917(2)	Mo3-O12	1.955(2)
Mo3-O22	2.2088(19)	Mo3-O9	2.439(2)
Mo4-O14	1.697(2)	Mo4-O13	1.757(2)
Mo4-O12	1.881(2)	Mo4-O16	1.937(2)
Mo4-O15	2.192(2)	Mo4-O22	2.3542(19)
Mo5-O17	1.691(2)	Mo5-O18	1.759(2)
Mo5-O1	1.904(2)	Mo5-O16	1.931(2)
Mo5-O21	2.200(2)	Mo5-O5	2.366(2)
Cu1-O18	1.941(2)	Cu1-O19	1.954(2)
Cu1-N2	1.981(3)	Cu1-N1	2.007(3)
Cu1-O20	2.268(3)	Cu2-O13	1.918(2)
Cu2-N3	1.981(2)	Cu2-N4	1.996(2)
Cu2-O24	2.002(2)	Cu2-O25	2.262(3)

P1-O15	1.518(2)	P1-O5	1.548(2)
P1-O9	1.548(2)	P1-C25	1.802(3)
P2-O21	1.513(2)	P2-O22	1.543(2)
P2-O23	1.546(2)	P2-C32	1.799(3)
O19-H19C	0.807(19)	O19-H19D	0.837(19)
O20-H20C	0.844(19)	O20-H20D	0.844(19)
O21-Mo5	2.200(2)	O22-Mo3	2.2088(19)
O22-Mo4	2.3542(19)	O23-Mo1	2.321(2)
O23-Mo2	2.341(2)	O24-H24C	0.829(19)
O24-H24D	0.820(19)	O25-H25C	0.829(19)
O25-H25D	0.818(19)	O90-H90A	0.852(19)
O90-H90B	0.854(19)	O91-H91A	0.855(19)
O91-H91B	0.861(19)	O92-H92A	0.77(5)
O92-H92B	0.80(5)	O93-H93A	0.837(19)
O93-H93B	0.837(19)	N1-C1	1.326(4)
N1-C4	1.356(4)	N2-C10	1.333(4)
N2-C9	1.363(4)	N3-C13	1.326(4)
N3-C17	1.357(4)	N4-C24	1.325(4)
N4-C18	1.360(4)	C1-C2	1.411(4)
C1-H1	0.93	C2-C3	1.366(5)
C2-H2	0.93	C3-C5	1.410(5)
C3-H3	0.93	C4-C5	1.409(4)

C4-C9	1.426(4)	C5-C6	1.436(4)
C6-C7	1.364(5)	C6-H6	0.93
C7-C8	1.436(4)	C7-H7	0.93
C8-C9	1.401(4)	C8-C12	1.410(5)
C10-C11	1.403(4)	C10-H10	0.93
C11-C12	1.380(5)	C11-H11	0.93
C12-H12	0.93	C13-C14	1.397(4)
C13-H13	0.93	C14-C15	1.370(5)
C14-H14	0.93	C15-C16	1.412(4)
C15-H15	0.93	C16-C17	1.406(4)
C16-C19	1.433(4)	C17-C18	1.433(4)
C18-C21	1.395(4)	C19-C20	1.357(5)
C19-H19A	0.93	C20-C21	1.440(4)
C20-H20A	0.93	C21-C22	1.409(4)
C22-C23	1.377(5)	C22-H22	0.93
C23-C24	1.405(4)	C23-H23	0.93
C24-H24	0.93	C25-C26	1.519(4)
C25-H25A	0.97	C25-H25B	0.97
C26-C28	1.397(4)	C26-C27	1.398(4)
C27-C29	1.392(4)	C27-H27	0.93
C28-C30	1.394(4)	C28-H28	0.93
C29-C31	1.397(4)	C29-H29	0.93

C30-C31	1.396(4)	C30-H30	0.93
C31-C32	1.515(4)	C32-H32A	0.97
C32-H32B	0.97		

Table S45. Bond angles (°) for [{Cu(o-phen)(H_2O)$_2$}$_2$$Mo_5O_{15}$(1,4-$O_3PC_8H_8PO_3$)]·4$H_2O$ (**6**·4H_2O).

O2-Mo1-O3	103.14(11)	O2-Mo1-O4	97.73(10)
O3-Mo1-O4	102.40(9)	O2-Mo1-O1	97.58(9)
O3-Mo1-O1	96.82(9)	O4-Mo1-O1	151.93(8)
O2-Mo1-O5	93.90(9)	O3-Mo1-O5	161.14(9)
O4-Mo1-O5	82.94(8)	O1-Mo1-O5	72.66(8)
O2-Mo1-O23	164.78(9)	O3-Mo1-O23	90.63(9)
O4-Mo1-O23	72.62(8)	O1-Mo1-O23	87.03(8)
O5-Mo1-O23	73.55(7)	O7-Mo2-O6	103.57(11)
O7-Mo2-O8	102.27(10)	O6-Mo2-O8	99.80(10)
O7-Mo2-O4	100.19(10)	O6-Mo2-O4	101.59(10)
O8-Mo2-O4	144.19(8)	O7-Mo2-O9	85.69(9)
O6-Mo2-O9	169.89(9)	O8-Mo2-O9	73.91(8)
O4-Mo2-O9	80.40(8)	O7-Mo2-O23	166.04(9)
O6-Mo2-O23	89.58(9)	O8-Mo2-O23	79.70(8)
O4-Mo2-O23	72.11(8)	O9-Mo2-O23	81.57(7)
O11-Mo3-O10	104.17(10)	O11-Mo3-O8	98.45(9)
O10-Mo3-O8	99.66(9)	O11-Mo3-O12	100.29(9)
O10-Mo3-O12	98.95(9)	O8-Mo3-O12	149.38(8)
O11-Mo3-O22	93.85(9)	O10-Mo3-O22	161.48(9)
O8-Mo3-O22	81.60(8)	O12-Mo3-O22	73.20(8)

O11-Mo3-O9	163.39(9)	O10-Mo3-O9	90.68(9)
O8-Mo3-O9	71.29(8)	O12-Mo3-O9	84.41(8)
O22-Mo3-O9	72.11(7)	O14-Mo4-O13	100.60(10)
O14-Mo4-O12	102.66(10)	O13-Mo4-O12	99.67(10)
O14-Mo4-O16	101.83(10)	O13-Mo4-O16	96.38(9)
O12-Mo4-O16	147.56(8)	O14-Mo4-O15	87.00(9)
O13-Mo4-O15	172.23(9)	O12-Mo4-O15	80.04(8)
O16-Mo4-O15	80.33(8)	O14-Mo4-O22	173.22(9)
O13-Mo4-O22	83.26(8)	O12-Mo4-O22	71.06(8)
O16-Mo4-O22	83.14(8)	O15-Mo4-O22	89.34(7)
O17-Mo5-O18	100.44(10)	O17-Mo5-O1	102.72(9)
O18-Mo5-O1	99.78(9)	O17-Mo5-O16	102.21(10)
O18-Mo5-O16	98.00(9)	O1-Mo5-O16	146.02(8)
O17-Mo5-O21	85.57(9)	O18-Mo5-O21	173.95(8)
O1-Mo5-O21	78.04(8)	O16-Mo5-O21	81.26(8)
O17-Mo5-O5	171.45(9)	O18-Mo5-O5	86.03(8)
O1-Mo5-O5	70.42(8)	O16-Mo5-O5	82.25(8)
O21-Mo5-O5	87.93(7)	O18-Cu1-O19	91.05(10)
O18-Cu1-N2	91.71(10)	O19-Cu1-N2	153.33(12)
O18-Cu1-N1	173.81(10)	O19-Cu1-N1	93.47(10)
N2-Cu1-N1	82.36(10)	O18-Cu1-O20	94.85(10)
O19-Cu1-O20	84.87(12)	N2-Cu1-O20	121.28(11)

N1-Cu1-O20	89.77(10)	O13-Cu2-N3	175.35(10)
O13-Cu2-N4	93.26(10)	N3-Cu2-N4	82.52(10)
O13-Cu2-O24	91.96(9)	N3-Cu2-O24	92.69(10)
N4-Cu2-O24	154.25(10)	O13-Cu2-O25	90.13(11)
N3-Cu2-O25	89.05(12)	N4-Cu2-O25	104.54(12)
O24-Cu2-O25	100.65(12)	O15-P1-O5	108.86(11)
O15-P1-O9	111.51(12)	O5-P1-O9	111.27(11)
O15-P1-C25	110.38(12)	O5-P1-C25	107.86(13)
O9-P1-C25	106.89(12)	O21-P2-O22	108.60(11)
O21-P2-O23	111.89(11)	O22-P2-O23	111.28(11)
O21-P2-C32	110.01(12)	O22-P2-C32	108.99(13)
O23-P2-C32	106.02(12)	Mo5-O1-Mo1	121.78(10)
Mo1-O4-Mo2	122.99(11)	P1-O5-Mo1	129.19(11)
P1-O5-Mo5	134.74(11)	Mo1-O5-Mo5	94.76(7)
Mo2-O8-Mo3	124.55(11)	P1-O9-Mo2	126.34(11)
P1-O9-Mo3	127.04(11)	Mo2-O9-Mo3	89.96(7)
Mo4-O12-Mo3	121.38(10)	Mo4-O13-Cu2	152.30(13)
P1-O15-Mo4	119.98(11)	Mo5-O16-Mo4	147.36(11)
Mo5-O18-Cu1	151.84(13)	Cu1-O19-H19C	91.(4)
Cu1-O19-H19D	119.(3)	H19C-O19-H19D	99.(5)
Cu1-O20-H20C	81.(4)	Cu1-O20-H20D	134.(4)
H20C-O20-H20D	64.(4)	P2-O21-Mo5	120.93(11)

P2-O22-Mo3	129.53(11)	P2-O22-Mo4	135.20(11)
Mo3-O22-Mo4	94.25(7)	P2-O23-Mo1	127.56(11)
P2-O23-Mo2	125.42(11)	Mo1-O23-Mo2	92.12(7)
Cu2-O24-H24C	129.(3)	Cu2-O24-H24D	113.(3)
H24C-O24-H24D	101.(4)	Cu2-O25-H25C	112.(4)
Cu2-O25-H25D	102.(4)	H25C-O25-H25D	132.(6)
H90A-O90-H90B	100.(5)	H91A-O91-H91B	75.(4)
H92A-O92-H92B	116.(5)	H93A-O93-H93B	104.(4)
C1-N1-C4	118.6(3)	C1-N1-Cu1	129.2(2)
C4-N1-Cu1	112.18(19)	C10-N2-C9	118.5(3)
C10-N2-Cu1	128.0(2)	C9-N2-Cu1	112.7(2)
C13-N3-C17	118.1(3)	C13-N3-Cu2	129.5(2)
C17-N3-Cu2	112.41(19)	C24-N4-C18	118.4(3)
C24-N4-Cu2	129.3(2)	C18-N4-Cu2	112.15(19)
N1-C1-C2	121.7(3)	N1-C1-H1	119.2
C2-C1-H1	119.2	C3-C2-C1	120.0(3)
C3-C2-H2	120.0	C1-C2-H2	120.0
C2-C3-C5	119.6(3)	C2-C3-H3	120.2
C5-C3-H3	120.2	N1-C4-C5	123.5(3)
N1-C4-C9	116.2(3)	C5-C4-C9	120.3(3)
C4-C5-C3	116.5(3)	C4-C5-C6	118.1(3)
C3-C5-C6	125.4(3)	C7-C6-C5	121.6(3)

C7-C6-H6	119.2	C5-C6-H6	119.2
C6-C7-C8	120.7(3)	C6-C7-H7	119.6
C8-C7-H7	119.6	C9-C8-C12	117.2(3)
C9-C8-C7	118.6(3)	C12-C8-C7	124.2(3)
N2-C9-C8	123.2(3)	N2-C9-C4	116.2(3)
C8-C9-C4	120.6(3)	N2-C10-C11	121.9(3)
N2-C10-H10	119.0	C11-C10-H10	119.0
C12-C11-C10	119.8(3)	C12-C11-H11	120.1
C10-C11-H11	120.1	C11-C12-C8	119.4(3)
C11-C12-H12	120.3	C8-C12-H12	120.3
N3-C13-C14	122.0(3)	N3-C13-H13	119.0
C14-C13-H13	119.0	C15-C14-C13	120.3(3)
C15-C14-H14	119.8	C13-C14-H14	119.8
C14-C15-C16	119.2(3)	C14-C15-H15	120.4
C16-C15-H15	120.4	C17-C16-C15	116.4(3)
C17-C16-C19	118.5(3)	C15-C16-C19	125.1(3)
N3-C17-C16	123.8(3)	N3-C17-C18	116.3(2)
C16-C17-C18	119.8(3)	N4-C18-C21	123.7(3)
N4-C18-C17	115.6(3)	C21-C18-C17	120.7(3)
C20-C19-C16	121.5(3)	C20-C19-H19A	119.3
C16-C19-H19A	119.3	C19-C20-C21	120.8(3)
C19-C20-H20A	119.6	C21-C20-H20A	119.6

C18-C21-C22	117.0(3)	C18-C21-C20	118.6(3)
C22-C21-C20	124.4(3)	C23-C22-C21	119.0(3)
C23-C22-H22	120.5	C21-C22-H22	120.5
C22-C23-C24	120.1(3)	C22-C23-H23	120.0
C24-C23-H23	120.0	N4-C24-C23	121.7(3)
N4-C24-H24	119.2	C23-C24-H24	119.2
C26-C25-P1	116.88(19)	C26-C25-H25A	108.1
P1-C25-H25A	108.1	C26-C25-H25B	108.1
P1-C25-H25B	108.1	H25A-C25-H25B	107.3
C28-C26-C27	118.2(3)	C28-C26-C25	122.0(2)
C27-C26-C25	119.8(2)	C29-C27-C26	120.9(3)
C29-C27-H27	119.6	C26-C27-H27	119.6
C30-C28-C26	121.0(3)	C30-C28-H28	119.5
C26-C28-H28	119.5	C27-C29-C31	120.8(3)
C27-C29-H29	119.6	C31-C29-H29	119.6
C28-C30-C31	120.7(3)	C28-C30-H30	119.7
C31-C30-H30	119.7	C30-C31-C29	118.4(3)
C30-C31-C32	121.9(3)	C29-C31-C32	119.7(2)
C31-C32-P2	116.29(19)	C31-C32-H32A	108.2
P2-C32-H32A	108.2	C31-C32-H32B	108.2
P2-C32-H32B	108.2	H32A-C32-H32B	107.4

Table S46. Torsion angles (°) for [{Cu(o-phen)(H_2O)$_2$}$_2$$Mo_5O_{15}$(1,4-$O_3PC_8H_8PO_3$)]·4$H_2O$ (**6**·4H_2O).

O15-P1-O5-Mo1	172.62(14)	O9-P1-O5-Mo1	49.34(18)
C25-P1-O5-Mo1	-67.60(17)	O15-P1-O5-Mo5	9.1(2)
O9-P1-O5-Mo5	-114.15(16)	C25-P1-O5-Mo5	128.91(16)
O7-Mo2-O8-Mo3	-76.63(14)	O6-Mo2-O8-Mo3	177.04(13)
O4-Mo2-O8-Mo3	51.0(2)	O9-Mo2-O8-Mo3	5.16(11)
O23-Mo2-O8-Mo3	89.27(12)	O15-P1-O9-Mo2	-153.05(12)
O5-P1-O9-Mo2	-31.30(17)	C25-P1-O9-Mo2	86.23(16)
O15-P1-O9-Mo3	-29.42(17)	O5-P1-O9-Mo3	92.33(15)
C25-P1-O9-Mo3	-150.14(14)	O14-Mo4-O12-Mo3	174.35(12)
O13-Mo4-O12-Mo3	-82.38(13)	O16-Mo4-O12-Mo3	36.2(2)
O15-Mo4-O12-Mo3	89.75(12)	O22-Mo4-O12-Mo3	-3.00(10)
O14-Mo4-O13-Cu2	-11.8(3)	O12-Mo4-O13-Cu2	-116.7(3)
O16-Mo4-O13-Cu2	91.6(3)	O22-Mo4-O13-Cu2	173.8(3)
O5-P1-O15-Mo4	-63.24(15)	O9-P1-O15-Mo4	59.89(15)
C25-P1-O15-Mo4	178.55(12)	O17-Mo5-O18-Cu1	-8.5(3)
O1-Mo5-O18-Cu1	-113.5(3)	O16-Mo5-O18-Cu1	95.6(3)
O5-Mo5-O18-Cu1	177.1(3)	O22-P2-O21-Mo5	-63.25(15)
O23-P2-O21-Mo5	59.98(16)	C32-P2-O21-Mo5	177.55(12)
O21-P2-O22-Mo3	176.00(13)	O23-P2-O22-Mo3	52.41(18)
C32-P2-O22-Mo3	-64.16(18)	O21-P2-O22-Mo4	10.7(2)

O23-P2-O22-Mo4	-112.87(16)	C32-P2-O22-Mo4	130.56(16)
O21-P2-O23-Mo1	-26.57(18)	O22-P2-O23-Mo1	95.12(15)
C32-P2-O23-Mo1	-146.51(14)	O21-P2-O23-Mo2	-153.73(12)
O22-P2-O23-Mo2	-32.04(17)	C32-P2-O23-Mo2	86.33(16)
C4-N1-C1-C2	-0.4(4)	Cu1-N1-C1-C2	176.6(2)
N1-C1-C2-C3	-1.6(5)	C1-C2-C3-C5	1.0(5)
C1-N1-C4-C5	2.9(4)	Cu1-N1-C4-C5	-174.6(2)
C1-N1-C4-C9	-178.9(3)	Cu1-N1-C4-C9	3.6(3)
N1-C4-C5-C3	-3.4(4)	C9-C4-C5-C3	178.5(3)
N1-C4-C5-C6	176.8(3)	C9-C4-C5-C6	-1.3(4)
C2-C3-C5-C4	1.3(4)	C2-C3-C5-C6	-178.9(3)
C4-C5-C6-C7	1.2(4)	C3-C5-C6-C7	-178.6(3)
C5-C6-C7-C8	-0.9(5)	C6-C7-C8-C9	0.7(4)
C6-C7-C8-C12	-176.8(3)	C10-N2-C9-C8	2.0(4)
Cu1-N2-C9-C8	172.8(2)	C10-N2-C9-C4	-176.2(3)
Cu1-N2-C9-C4	-5.3(3)	C12-C8-C9-N2	-1.2(4)
C7-C8-C9-N2	-178.9(3)	C12-C8-C9-C4	176.8(3)
C7-C8-C9-C4	-0.8(4)	N1-C4-C9-N2	1.1(4)
C5-C4-C9-N2	179.4(3)	N1-C4-C9-C8	-177.1(3)
C5-C4-C9-C8	1.2(4)	C9-N2-C10-C11	-0.8(4)
Cu1-N2-C10-C11	-170.0(2)	N2-C10-C11-C12	-1.1(5)
C10-C11-C12-C8	1.8(5)	C9-C8-C12-C11	-0.7(4)

C7-C8-C12-C11	176.8(3)	C17-N3-C13-C14	-0.7(4)
Cu2-N3-C13-C14	178.2(2)	N3-C13-C14-C15	-2.2(5)
C13-C14-C15-C16	2.3(5)	C14-C15-C16-C17	0.4(4)
C14-C15-C16-C19	178.4(3)	C13-N3-C17-C16	3.7(4)
Cu2-N3-C17-C16	-175.5(2)	C13-N3-C17-C18	-174.2(3)
Cu2-N3-C17-C18	6.6(3)	C15-C16-C17-N3	-3.5(4)
C19-C16-C17-N3	178.3(3)	C15-C16-C17-C18	174.4(3)
C19-C16-C17-C18	-3.8(4)	C24-N4-C18-C21	-3.1(4)
Cu2-N4-C18-C21	173.1(2)	C24-N4-C18-C17	176.4(3)
Cu2-N4-C18-C17	-7.4(3)	N3-C17-C18-N4	0.6(4)
C16-C17-C18-N4	-177.4(3)	N3-C17-C18-C21	-179.9(3)
C16-C17-C18-C21	2.1(4)	C17-C16-C19-C20	2.2(4)
C15-C16-C19-C20	-175.8(3)	C16-C19-C20-C21	1.3(5)
N4-C18-C21-C22	3.0(4)	C17-C18-C21-C22	-176.5(3)
N4-C18-C21-C20	-179.2(3)	C17-C18-C21-C20	1.3(4)
C19-C20-C21-C18	-3.0(4)	C19-C20-C21-C22	174.7(3)
C18-C21-C22-C23	-0.1(4)	C20-C21-C22-C23	-177.8(3)
C21-C22-C23-C24	-2.5(4)	C18-N4-C24-C23	0.3(4)
Cu2-N4-C24-C23	-175.1(2)	C22-C23-C24-N4	2.5(5)
O15-P1-C25-C26	53.9(2)	O5-P1-C25-C26	-64.9(2)
O9-P1-C25-C26	175.3(2)	P1-C25-C26-C28	71.2(3)
P1-C25-C26-C27	-110.4(3)	C28-C26-C27-C29	-0.4(4)

C25-C26-C27-C29	-178.8(3)	C27-C26-C28-C30	-0.7(4)
C25-C26-C28-C30	177.8(3)	C26-C27-C29-C31	1.0(4)
C26-C28-C30-C31	1.0(4)	C28-C30-C31-C29	-0.4(4)
C28-C30-C31-C32	177.5(3)	C27-C29-C31-C30	-0.6(4)
C27-C29-C31-C32	-178.6(3)	C30-C31-C32-P2	76.0(3)
C29-C31-C32-P2	-106.2(3)	O21-P2-C32-C31	47.3(2)
O22-P2-C32-C31	-71.6(2)	O23-P2-C32-C31	168.5(2)

Table S47. Anisotropic atomic displacement parameters ($Å^2$) for [{Cu(o-phen)$(H_2O)_2$}$_2$$Mo_5O_{15}$(1,4-$O_3PC_8H_8PO_3$)]·4$H_2O$ (**6**·4H_2O).

The anisotropic atomic displacement factor exponent takes the form: $-2\pi^2[h^2 a^{*2} U_{11} + ... + 2 h k a^* b^* U_{12}]$

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
Mo1	0.00814(9)	0.00641(10)	0.00938(9)	-0.00083(7)	0.00366(7)	-0.00091(7)
Mo2	0.00807(9)	0.00767(10)	0.01062(10)	-0.00084(7)	0.00470(8)	-0.00077(7)
Mo3	0.00862(9)	0.00669(10)	0.00866(9)	-0.00041(7)	0.00414(7)	-0.00021(7)
Mo4	0.00756(9)	0.00778(10)	0.00831(9)	-0.00017(7)	0.00317(7)	-0.00079(7)
Mo5	0.00803(9)	0.00705(10)	0.00875(9)	-0.00036(7)	0.00415(7)	0.00022(7)
Cu1	0.00940(14)	0.00918(16)	0.01110(15)	-0.00034(11)	0.00369(12)	0.00169(11)
Cu2	0.01098(14)	0.00901(16)	0.01068(15)	-0.00078(11)	0.00519(12)	-0.00264(11)
P1	0.0085(3)	0.0059(3)	0.0092(3)	0.0002(2)	0.0042(2)	0.0002(2)
P2	0.0073(3)	0.0058(3)	0.0087(3)	-0.0001(2)	0.0033(2)	0.0003(2)
O1	0.0106(8)	0.0070(9)	0.0091(8)	-0.0007(6)	0.0045(7)	0.0004(6)
O2	0.0154(9)	0.0076(9)	0.0166(10)	-0.0009(7)	0.0089(8)	-0.0011(7)
O3	0.0114(8)	0.0123(10)	0.0112(9)	0.0008(7)	0.0040(7)	0.0000(7)
O4	0.0126(8)	0.0070(9)	0.0133(9)	-0.0027(7)	0.0070(7)	-0.0024(7)
O5	0.0094(8)	0.0097(9)	0.0090(8)	-0.0004(7)	0.0037(7)	-0.0004(6)
O6	0.0105(8)	0.0135(11)	0.0165(10)	-0.0018(8)	0.0050(8)	-0.0002(7)
O7	0.0161(9)	0.0143(11)	0.0177(10)	-0.0007(8)	0.0102(8)	-0.0004(8)
O8	0.0100(8)	0.0082(9)	0.0109(9)	-0.0011(7)	0.0047(7)	0.0012(6)
O9	0.0092(8)	0.0069(9)	0.0108(8)	-0.0002(6)	0.0042(7)	0.0008(6)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
O10	0.0123(8)	0.0126(10)	0.0114(9)	-0.0001(7)	0.0064(7)	0.0004(7)
O11	0.0132(9)	0.0077(10)	0.0157(10)	-0.0007(7)	0.0066(8)	-0.0006(7)
O12	0.0101(8)	0.0100(9)	0.0088(8)	-0.0004(7)	0.0043(7)	0.0003(7)
O13	0.0132(9)	0.0103(10)	0.0127(9)	-0.0017(7)	0.0056(7)	-0.0033(7)
O14	0.0093(8)	0.0160(11)	0.0130(9)	0.0016(7)	0.0035(7)	0.0010(7)
O15	0.0095(8)	0.0067(9)	0.0118(9)	0.0008(7)	0.0041(7)	0.0004(6)
O16	0.0099(8)	0.0094(9)	0.0095(8)	-0.0008(7)	0.0048(7)	-0.0002(6)
O17	0.0142(9)	0.0142(11)	0.0127(9)	0.0005(7)	0.0078(8)	0.0000(7)
O18	0.0107(8)	0.0097(10)	0.0125(9)	0.0001(7)	0.0051(7)	0.0021(7)
O19	0.0160(10)	0.0211(13)	0.0197(12)	-0.0068(9)	-0.0003(9)	0.0055(9)
O20	0.0279(13)	0.0224(14)	0.0349(15)	0.0100(11)	0.0211(12)	0.0052(10)
O21	0.0103(8)	0.0072(9)	0.0110(8)	-0.0003(6)	0.0059(7)	0.0009(6)
O22	0.0088(8)	0.0088(9)	0.0079(8)	0.0001(6)	0.0035(7)	0.0003(6)
O23	0.0096(8)	0.0072(9)	0.0119(9)	0.0013(7)	0.0050(7)	0.0019(6)
O24	0.0202(10)	0.0089(10)	0.0181(10)	-0.0012(8)	0.0122(8)	-0.0018(8)
O25	0.0184(12)	0.0205(15)	0.052(2)	-0.0118(14)	-0.0058(13)	0.0058(10)
O90	0.0229(13)	0.048(2)	0.0364(16)	0.0036(14)	0.0172(12)	0.0085(13)
O91	0.0212(12)	0.0286(15)	0.0326(15)	0.0019(11)	0.0138(11)	-0.0026(10)
O92	0.0239(12)	0.0255(14)	0.0264(13)	0.0077(10)	0.0179(11)	0.0076(10)
O93	0.0207(10)	0.0140(11)	0.0154(10)	0.0040(8)	0.0075(9)	-0.0028(8)
N1	0.0112(10)	0.0104(11)	0.0138(11)	-0.0002(8)	0.0057(8)	-0.0001(8)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
N2	0.0110(10)	0.0112(12)	0.0139(11)	-0.0006(8)	0.0055(8)	0.0002(8)
N3	0.0100(9)	0.0093(11)	0.0128(10)	-0.0021(8)	0.0048(8)	-0.0011(8)
N4	0.0106(9)	0.0096(11)	0.0131(10)	0.0002(8)	0.0055(8)	-0.0011(8)
C1	0.0149(12)	0.0144(14)	0.0169(13)	-0.0031(10)	0.0089(10)	-0.0008(10)
C2	0.0161(12)	0.0134(14)	0.0226(15)	-0.0035(11)	0.0112(11)	-0.0015(10)
C3	0.0153(12)	0.0113(14)	0.0300(17)	-0.0013(11)	0.0134(12)	-0.0004(10)
C4	0.0090(10)	0.0094(12)	0.0146(12)	0.0000(9)	0.0051(9)	-0.0007(8)
C5	0.0095(11)	0.0105(13)	0.0231(14)	0.0036(10)	0.0075(10)	0.0017(9)
C6	0.0119(11)	0.0130(14)	0.0234(15)	0.0046(11)	0.0065(11)	0.0013(10)
C7	0.0123(12)	0.0167(15)	0.0188(14)	0.0054(11)	0.0035(11)	-0.0018(10)
C8	0.0115(11)	0.0131(14)	0.0135(12)	0.0022(10)	0.0039(10)	-0.0031(9)
C9	0.0097(10)	0.0099(13)	0.0141(12)	0.0010(9)	0.0045(9)	-0.0012(9)
C10	0.0136(12)	0.0135(14)	0.0158(13)	-0.0023(10)	0.0071(10)	-0.0017(10)
C11	0.0161(12)	0.0184(15)	0.0133(13)	-0.0037(10)	0.0057(10)	-0.0058(10)
C12	0.0137(12)	0.0199(16)	0.0122(12)	0.0003(10)	0.0034(10)	-0.0058(10)
C13	0.0115(11)	0.0105(13)	0.0177(13)	-0.0044(10)	0.0048(10)	-0.0015(9)
C14	0.0108(11)	0.0110(14)	0.0257(15)	-0.0030(11)	0.0056(11)	-0.0029(9)
C15	0.0109(11)	0.0091(13)	0.0310(16)	-0.0005(11)	0.0100(11)	-0.0018(9)
C16	0.0129(11)	0.0070(12)	0.0247(15)	0.0018(10)	0.0114(11)	0.0005(9)
C17	0.0120(11)	0.0075(12)	0.0160(12)	0.0000(9)	0.0079(10)	-0.0004(9)
C18	0.0116(11)	0.0090(12)	0.0140(12)	0.0001(9)	0.0069(9)	0.0003(9)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
C19	0.0165(12)	0.0118(14)	0.0251(15)	0.0066(11)	0.0138(12)	0.0035(10)
C20	0.0188(13)	0.0147(14)	0.0183(13)	0.0062(11)	0.0118(11)	0.0056(10)
C21	0.0145(12)	0.0122(13)	0.0136(12)	0.0036(9)	0.0070(10)	0.0034(9)
C22	0.0169(12)	0.0153(14)	0.0115(12)	0.0013(10)	0.0061(10)	0.0038(10)
C23	0.0169(12)	0.0148(14)	0.0109(12)	-0.0020(10)	0.0032(10)	-0.0004(10)
C24	0.0131(11)	0.0118(13)	0.0140(12)	-0.0018(9)	0.0052(10)	-0.0005(9)
C25	0.0129(11)	0.0059(12)	0.0159(12)	0.0018(9)	0.0093(10)	0.0001(8)
C26	0.0100(10)	0.0064(12)	0.0131(11)	0.0011(9)	0.0061(9)	0.0008(8)
C27	0.0156(12)	0.0092(13)	0.0113(11)	0.0011(9)	0.0068(10)	0.0012(9)
C28	0.0123(11)	0.0087(12)	0.0129(12)	0.0005(9)	0.0048(9)	0.0011(9)
C29	0.0127(11)	0.0099(13)	0.0115(12)	0.0002(9)	0.0047(9)	0.0022(9)
C30	0.0153(12)	0.0077(12)	0.0126(12)	-0.0013(9)	0.0060(10)	0.0007(9)
C31	0.0109(10)	0.0050(11)	0.0143(12)	0.0001(9)	0.0063(9)	0.0000(8)
C32	0.0092(10)	0.0058(12)	0.0157(12)	-0.0015(9)	0.0059(9)	-0.0010(8)

Table S48. Hydrogen atomic coordinates and isotropic atomic displacement parameters ($Å^2$) for $[\{Cu(o\text{-}phen)(H_2O)_2\}_2Mo_5O_{15}(1,4\text{-}O_3PC_8H_8PO_3)]\cdot 4H_2O$ (**6**·$4H_2O$).

	x/a	y/b	z/c	U(eq)
H19C	0.919(4)	0.824(2)	0.953(3)	0.027
H19D	1.008(2)	0.830(2)	0.947(3)	0.027
H20C	0.732(4)	0.737(2)	0.8539(19)	0.031
H20D	0.759(4)	0.7156(10)	0.903(3)	0.031
H24C	0.783(3)	0.425(2)	0.525(2)	0.017
H24D	0.752(4)	0.3780(9)	0.552(3)	0.017
H25C	0.602(5)	0.5572(17)	0.605(3)	0.047
H25D	0.573(5)	0.517(3)	0.5297(16)	0.047
H90A	0.606(2)	0.872(3)	0.348(3)	0.041
H90B	0.666(5)	0.871(3)	0.3085(19)	0.041
H91A	0.679(4)	0.8439(16)	0.226(3)	0.032
H91B	0.608(4)	0.8474(19)	0.167(3)	0.032
H92A	0.549(4)	0.276(3)	0.442(3)	0.027
H92B	0.561(4)	0.313(2)	0.387(3)	0.027
H93A	0.178(3)	0.696(2)	0.5571(12)	0.021
H93B	0.160(4)	0.7351(11)	0.496(3)	0.021
H1	0.7524	0.8999	0.9273	0.018
H2	0.6184	0.9789	0.8978	0.02
H3	0.5019	1.0084	0.7618	0.021

	x/a	y/b	z/c	U(eq)
H6	0.4432	0.9918	0.6044	0.02
H7	0.4611	0.9352	0.5050	0.021
H10	0.8065	0.7511	0.6607	0.017
H11	0.7060	0.7721	0.5186	0.02
H12	0.5647	0.8476	0.4696	0.02
H13	0.5365	0.3706	0.5286	0.017
H14	0.4249	0.2972	0.5489	0.02
H15	0.4607	0.2708	0.6820	0.02
H19A	0.5805	0.2922	0.8415	0.019
H20A	0.7143	0.3512	0.9457	0.019
H22	0.8490	0.4475	0.9885	0.018
H23	0.9251	0.5282	0.9456	0.019
H24	0.8857	0.5347	0.8069	0.016
H25A	1.1623	0.7967	0.6864	0.013
H25B	1.0768	0.7853	0.5923	0.013
H27	0.9239	0.8537	0.5264	0.014
H28	1.0700	0.8416	0.7765	0.014
H29	0.8102	0.9345	0.5286	0.014
H30	0.9599	0.9244	0.7790	0.015
H32A	0.7645	0.9785	0.6896	0.012
H32B	0.7333	0.9945	0.5973	0.012

Table S49. Sample and crystal data for $[Cu_2(o\text{-}phen)_2(H_2O)_2(1,4\text{-}HO_3PC_8H_8PO_3H)][Cu(o\text{-}phen)(H_2O)Mo_5O_{14}(OH)(1,4\text{-}O_3PC_8H_8O_3P)]_2$ (**7**).

Identification code	$[Cu_2(o\text{-}phen)_2(H_2O)_2(1,4\text{-}HO_3PC_8H_8PO_3H)][Cu(o\text{-}phen)(H_2O)Mo_5O_{14}(OH)(1,4\text{-}O_3PC_8H_8O_3P)]_2$	
Chemical formula	$C_{36}H_{34}Cu_2Mo_5N_4O_{26}P_3$	
Formula weight	1638.36	
Temperature	90(2) K	
Wavelength	0.71073 Å	
Crystal size	0.076 x 0.121 x 0.121 mm	
Crystal system	Monoclinic	
Space group	C2/c	
Unit cell dimensions	a = 53.884(3) Å	α = 90°
	b = 9.5839(5) Å	β = 91.5890(10)°
	c = 18.7832(9) Å	γ = 90°
Volume	9696.3(8) $Å^3$	
Z	8	
Density (calculated)	2.245 g/cm^3	
Absorption coefficient	2.306 mm^{-1}	
F(000)	6392	

Table S50. Data collection and structure refinement for $[Cu_2(o\text{-phen})_2(H_2O)_2(1,4\text{-}HO_3PC_8H_8PO_3H)][Cu(o\text{-phen})(H_2O)Mo_5O_{14}(OH)(1,4\text{-}O_3PC_8H_8O_3P)]_2$ (**7**).

Theta range for data collection	2.16 to 26.37°	
Index ranges	-67<=h<=67, -11<=k<=11, -20<=l<=23	
Reflections collected	55032	
Independent reflections	9899 [R(int) = 0.0619]	
Coverage of independent reflections	99.9%	
Absorption correction	multi-scan	
Max. and min. transmission	0.8440 and 0.7680	
Refinement method	Full-matrix least-squares on F^2	
Refinement program	SHELXL-2013 (Sheldrick, 2013)	
Function minimized	$\Sigma\, w(F_o^2 - F_c^2)^2$	
Data / restraints / parameters	9899 / 252 / 703	
Goodness-of-fit on F^2	1.033	
Δ/σ_{max}	0.005	
Final R indices	7966 data; I>2σ(I)	R1 = 0.0317, wR2 = 0.0620
	all data	R1 = 0.0476, wR2 = 0.0661
Weighting scheme	$w=1/[\sigma^2(F_o^2)+(0.0284P)^2+10.5994P]$ where $P=(F_o^2+2F_c^2)/3$	
Largest diff. peak and hole	1.627 and -0.480 eÅ^{-3}	
R.M.S. deviation from mean	0.122 eÅ^{-3}	

Table S51. Atomic coordinates and equivalent isotropic atomic displacement parameters ($Å^2$) for $[Cu_2(o\text{-phen})_2(H_2O)_2(1,4\text{-}HO_3PC_8H_8PO_3H)][Cu(o\text{-phen})(H_2O)Mo_5O_{14}(OH)(1,4\text{-}O_3PC_8H_8O_3P)]_2$ (**7**).

U(eq) is defined as one third of the trace of the orthogonalized U_{ij} tensor.

	x/a	**y/b**	**z/c**	**U(eq)**
Mo1	0.40593(2)	0.57178(4)	0.27422(2)	0.00991(8)
Mo2	0.39149(2)	0.31642(4)	0.38596(2)	0.01020(8)
Mo3	0.32635(2)	0.30677(4)	0.40175(2)	0.00982(8)
Mo4	0.30417(2)	0.50314(4)	0.24442(2)	0.00934(8)
Mo5	0.35344(2)	0.69458(3)	0.19184(2)	0.00916(8)
Cu1	0.46926(2)	0.36339(5)	0.07308(3)	0.01019(11)
Cu2	0.31561(2)	0.26293(5)	0.11493(2)	0.01055(11)
P1	0.49304(2)	0.38306(10)	0.91299(5)	0.0098(2)
P2	0.35899(2)	0.32977(10)	0.23506(5)	0.0081(2)
P3	0.35277(2)	0.38662(10)	0.86870(5)	0.0087(2)
O1	0.43789(5)	0.5078(3)	0.06614(16)	0.0191(7)
O2	0.47609(5)	0.4023(3)	0.97430(14)	0.0125(6)
O3	0.47758(5)	0.3183(3)	0.84802(15)	0.0143(6)
O4	0.50511(5)	0.5148(3)	0.88682(14)	0.0121(6)
O5	0.42225(5)	0.5542(3)	0.19842(15)	0.0179(6)
O6	0.42635(5)	0.6548(3)	0.33165(15)	0.0165(6)
O7	0.40868(5)	0.3809(3)	0.30746(14)	0.0116(6)
O8	0.37157(5)	0.4664(3)	0.21442(14)	0.0105(6)

	x/a	y/b	z/c	U(eq)
O9	0.38380(5)	0.7167(3)	0.24822(14)	0.0111(6)
O10	0.36377(5)	0.6921(3)	0.10625(14)	0.0156(6)
O11	0.34250(5)	0.8582(3)	0.20396(14)	0.0139(6)
O12	0.32267(5)	0.5953(3)	0.17189(14)	0.0101(6)
O13	0.28554(5)	0.6296(3)	0.27626(15)	0.0162(6)
O14	0.28460(5)	0.4086(3)	0.18927(14)	0.0128(6)
O15	0.30700(4)	0.3862(3)	0.32778(14)	0.0107(6)
O16	0.35784(5)	0.3118(3)	0.31745(13)	0.0093(5)
O17	0.32352(5)	0.1337(3)	0.38299(14)	0.0161(6)
O18	0.30967(5)	0.3238(3)	0.47614(15)	0.0166(6)
O19	0.36092(5)	0.2962(3)	0.44992(14)	0.0121(6)
O20	0.40977(5)	0.3637(3)	0.45756(14)	0.0159(6)
O21	0.39645(5)	0.1434(3)	0.37491(15)	0.0161(6)
O22	0.33221(5)	0.3252(3)	0.20531(13)	0.0095(5)
O23	0.32865(6)	0.4205(3)	0.06470(15)	0.0161(6)
O24	0.33697(4)	0.4640(3)	0.92167(13)	0.0092(6)
O25	0.33916(5)	0.3868(3)	0.79512(13)	0.0093(6)
O26	0.37876(5)	0.4550(3)	0.86526(13)	0.0092(6)
N1	0.44889(6)	0.1946(3)	0.04769(17)	0.0118(7)
N2	0.46315(6)	0.2934(3)	0.17253(17)	0.0111(7)
N3	0.29337(6)	0.2040(3)	0.03308(17)	0.0112(7)

	x/a	y/b	z/c	U(eq)
N4	0.29925(6)	0.1072(3)	0.16397(17)	0.0106(7)
C1	0.44043(7)	0.1554(4)	0.9841(2)	0.0181(9)
C2	0.42593(8)	0.0349(5)	0.9741(2)	0.0213(10)
C3	0.42033(8)	0.9542(5)	0.0317(2)	0.0199(10)
C4	0.42883(7)	0.9940(4)	0.1002(2)	0.0141(8)
C5	0.44271(7)	0.1164(4)	0.1051(2)	0.0114(8)
C6	0.42363(8)	0.9209(4)	0.1647(2)	0.0197(9)
C7	0.43147(7)	0.9687(4)	0.2288(2)	0.0186(9)
C8	0.44507(7)	0.0981(4)	0.2352(2)	0.0146(9)
C9	0.45063(7)	0.1699(4)	0.1728(2)	0.0120(8)
C10	0.45273(7)	0.1596(4)	0.3002(2)	0.0170(9)
C11	0.46468(7)	0.2862(4)	0.2996(2)	0.0149(9)
C12	0.46956(7)	0.3507(4)	0.2346(2)	0.0134(8)
C13	0.29028(7)	0.2592(4)	0.9687(2)	0.0134(8)
C14	0.27270(7)	0.2077(4)	0.9193(2)	0.0153(9)
C15	0.25816(8)	0.0966(5)	0.9361(2)	0.0190(9)
C16	0.26098(7)	0.0351(4)	0.0041(2)	0.0148(9)
C17	0.24564(8)	0.9251(5)	0.0296(2)	0.0209(10)
C18	0.24782(7)	0.8799(5)	0.0977(2)	0.0198(9)
C19	0.26602(7)	0.9395(4)	0.1467(2)	0.0140(8)
C20	0.27872(7)	0.0946(4)	0.0508(2)	0.0124(8)

	x/a	y/b	z/c	U(eq)
C21	0.28144(7)	0.0442(4)	0.1224(2)	0.0113(8)
C22	0.26894(7)	0.9018(4)	0.2193(2)	0.0173(9)
C23	0.28696(8)	0.9652(5)	0.2603(2)	0.0198(10)
C24	0.30178(7)	0.0678(4)	0.2310(2)	0.0137(9)
C25	0.51671(7)	0.2544(4)	0.9310(2)	0.0171(9)
C26	0.50821(7)	0.1224(4)	0.9658(2)	0.0132(8)
C27	0.49859(7)	0.0113(4)	0.9262(2)	0.0160(9)
C28	0.49070(7)	0.8913(4)	0.9605(2)	0.0154(9)
C29	0.37569(7)	0.1836(4)	0.2013(2)	0.0116(8)
C30	0.37312(7)	0.1781(4)	0.1208(2)	0.0102(8)
C31	0.38628(7)	0.2719(4)	0.0798(2)	0.0123(8)
C32	0.38207(7)	0.2783(4)	0.0068(2)	0.0124(8)
C33	0.35648(7)	0.0875(4)	0.0872(2)	0.0111(8)
C34	0.35198(7)	0.0949(4)	0.0137(2)	0.0116(8)
C35	0.36434(7)	0.1927(4)	0.9733(2)	0.0103(8)
C36	0.35765(7)	0.2078(4)	0.8945(2)	0.0113(8)

Table S52. Bond lengths (Å) for $[Cu_2(o\text{-phen})_2(H_2O)_2(1,4\text{-}HO_3PC_8H_8PO_3H)][Cu(o\text{-phen})(H_2O)Mo_5O_{14}(OH)(1,4\text{-}O_3PC_8H_8O_3P)]_2$ (**7**).

Mo1-O5	1.703(3)	Mo1-O6	1.715(3)
Mo1-O9	1.886(3)	Mo1-O7	1.938(3)
Mo1-O26	2.296(3)	Mo1-O8	2.365(2)
Mo2-O21	1.693(3)	Mo2-O20	1.706(3)
Mo2-O7	1.868(3)	Mo2-O19	2.075(3)
Mo2-O16	2.195(2)	Mo2-O26	2.326(3)
Mo3-O18	1.690(3)	Mo3-O17	1.702(3)
Mo3-O15	1.875(2)	Mo3-O19	2.051(3)
Mo3-O24	2.298(3)	Mo3-O16	2.353(3)
Mo4-O13	1.693(3)	Mo4-O14	1.716(3)
Mo4-O12	1.925(3)	Mo4-O15	1.928(3)
Mo4-O25	2.340(2)	Mo4-O22	2.407(3)
Mo5-O11	1.693(3)	Mo5-O10	1.716(3)
Mo5-O9	1.935(3)	Mo5-O12	1.939(3)
Mo5-O25	2.246(3)	Mo5-O8	2.427(3)
Cu1-O2	1.938(3)	Cu1-O4	1.945(3)
Cu1-N1	2.005(3)	Cu1-N2	2.021(3)
Cu1-O1	2.186(3)	Cu2-O23	1.924(3)
Cu2-N4	1.975(3)	Cu2-O22	1.988(3)
Cu2-N3	2.004(3)	P1-O2	1.501(3)

P1-O4	1.508(3)	P1-O3	1.585(3)
P1-C25	1.799(4)	P2-O8	1.530(3)
P2-O22	1.533(3)	P2-O16	1.560(3)
P2-C29	1.791(4)	P3-O24	1.520(3)
P3-O25	1.546(3)	P3-O26	1.549(3)
P3-C36	1.798(4)	O1-H1A	0.816(19)
O1-H1B	0.821(19)	O3-H3'	0.815(19)
O4-Cu1	1.945(3)	O19-H19'	0.820(19)
O23-H23A	0.821(19)	O23-H23B	0.818(19)
O24-Mo3	2.298(3)	O25-Mo5	2.246(3)
O25-Mo4	2.340(2)	O26-Mo1	2.296(3)
O26-Mo2	2.325(3)	N1-C1	1.322(5)
N1-C5	1.362(5)	N2-C12	1.325(5)
N2-C9	1.362(5)	N3-C13	1.327(5)
N3-C20	1.360(5)	N4-C24	1.317(5)
N4-C21	1.362(5)	C1-C2	1.404(6)
C1-H1	0.95	C2-C3	1.370(6)
C2-H2	0.95	C3-C4	1.406(6)
C3-H3	0.95	C4-C5	1.393(5)
C4-C6	1.434(6)	C5-C9	1.427(6)
C6-C7	1.345(6)	C6-H6	0.95
C7-C8	1.444(6)	C7-H7	0.95

C8-C9	1.398(6)	C8-C10	1.408(6)
C10-C11	1.374(6)	C10-H10	0.95
C11-C12	1.400(6)	C11-H11	0.95
C12-H12	0.95	C13-C14	1.398(5)
C13-H13	0.95	C14-C15	1.364(6)
C14-H14	0.95	C15-C16	1.412(6)
C15-H15	0.95	C16-C20	1.400(5)
C16-C17	1.431(6)	C17-C18	1.351(6)
C17-H17	0.95	C18-C19	1.444(6)
C18-H18	0.95	C19-C21	1.388(5)
C19-C22	1.416(6)	C20-C21	1.432(5)
C22-C23	1.365(6)	C22-H22	0.95
C23-C24	1.391(6)	C23-H23	0.95
C24-H24	0.95	C25-C26	1.501(6)
C25-H25A	0.99	C25-H25B	0.99
C26-C28	1.391(6)	C26-C27	1.391(6)
C27-C28	1.391(6)	C27-H27	0.95
C28-C26	1.391(6)	C28-H28	0.95
C29-C30	1.517(5)	C29-H29A	0.99
C29-H29B	0.99	C30-C33	1.387(5)
C30-C31	1.390(5)	C31-C32	1.384(5)
C31-H31	0.95	C32-C35	1.396(5)

C32-H32	0.95	C33-C34	1.398(5)
C33-H33	0.95	C34-C35	1.387(5)
C34-H34	0.95	C35-C36	1.520(5)
C36-H36A	0.99	C36-H36B	0.99

Table S53. Bond angles (°) for $[Cu_2(o\text{-phen})_2(H_2O)_2(1,4\text{-}HO_3PC_8H_8PO_3H)][Cu(o\text{-phen})(H_2O)Mo_5O_{14}(OH)(1,4\text{-}O_3PC_8H_8O_3P)]_2$ (**7**).

O5-Mo1-O6	103.55(13)	O5-Mo1-O9	101.19(12)
O6-Mo1-O9	102.20(12)	O5-Mo1-O7	97.99(12)
O6-Mo1-O7	101.15(12)	O9-Mo1-O7	145.19(11)
O5-Mo1-O26	165.15(12)	O6-Mo1-O26	89.70(11)
O9-Mo1-O26	82.15(10)	O7-Mo1-O26	72.50(10)
O5-Mo1-O8	88.64(11)	O6-Mo1-O8	167.67(12)
O9-Mo1-O8	73.16(10)	O7-Mo1-O8	78.57(10)
O26-Mo1-O8	78.42(9)	O21-Mo2-O20	105.50(13)
O21-Mo2-O7	98.26(13)	O20-Mo2-O7	104.25(12)
O21-Mo2-O19	96.31(12)	O20-Mo2-O19	91.07(12)
O7-Mo2-O19	155.11(11)	O21-Mo2-O16	92.23(11)
O20-Mo2-O16	156.51(12)	O7-Mo2-O16	87.91(10)
O19-Mo2-O16	71.42(10)	O21-Mo2-O26	161.63(11)
O20-Mo2-O26	92.44(11)	O7-Mo2-O26	72.95(10)
O19-Mo2-O26	87.12(10)	O16-Mo2-O26	71.70(9)
O18-Mo3-O17	102.63(14)	O18-Mo3-O15	106.02(12)
O17-Mo3-O15	101.41(12)	O18-Mo3-O19	97.89(12)
O17-Mo3-O19	96.79(12)	O15-Mo3-O19	145.74(11)
O18-Mo3-O24	84.77(12)	O17-Mo3-O24	170.38(11)
O15-Mo3-O24	82.14(10)	O19-Mo3-O24	75.90(10)

O18-Mo3-O16	164.62(11)	O17-Mo3-O16	86.72(11)
O15-Mo3-O16	83.66(10)	O19-Mo3-O16	68.58(10)
O24-Mo3-O16	84.78(9)	O13-Mo4-O14	103.34(13)
O13-Mo4-O12	104.20(12)	O14-Mo4-O12	97.86(12)
O13-Mo4-O15	99.41(12)	O14-Mo4-O15	102.53(12)
O12-Mo4-O15	144.14(10)	O13-Mo4-O25	90.72(11)
O14-Mo4-O25	163.40(11)	O12-Mo4-O25	69.74(10)
O15-Mo4-O25	83.49(10)	O13-Mo4-O22	176.62(11)
O14-Mo4-O22	79.74(11)	O12-Mo4-O22	76.55(10)
O15-Mo4-O22	78.45(10)	O25-Mo4-O22	86.46(9)
O11-Mo5-O10	105.19(13)	O11-Mo5-O9	96.73(12)
O10-Mo5-O9	102.89(12)	O11-Mo5-O12	100.45(12)
O10-Mo5-O12	96.31(12)	O9-Mo5-O12	149.84(11)
O11-Mo5-O25	94.38(11)	O10-Mo5-O25	158.76(12)
O9-Mo5-O25	82.53(10)	O12-Mo5-O25	71.65(10)
O11-Mo5-O8	161.96(11)	O10-Mo5-O8	90.68(11)
O9-Mo5-O8	70.91(10)	O12-Mo5-O8	85.97(9)
O25-Mo5-O8	71.48(9)	O2-Cu1-O4	95.94(11)
O2-Cu1-N1	92.58(12)	O4-Cu1-N1	162.74(12)
O2-Cu1-N2	171.62(12)	O4-Cu1-N2	88.48(12)
N1-Cu1-N2	81.36(13)	O2-Cu1-O1	89.32(11)
O4-Cu1-O1	100.52(12)	N1-Cu1-O1	94.55(13)

N2-Cu1-O1	96.89(12)	O23-Cu2-N4	174.90(13)
O23-Cu2-O22	91.23(11)	N4-Cu2-O22	91.40(12)
O23-Cu2-N3	93.65(13)	N4-Cu2-N3	83.09(13)
O22-Cu2-N3	169.97(12)	O2-P1-O4	115.09(16)
O2-P1-O3	108.72(15)	O4-P1-O3	107.39(15)
O2-P1-C25	112.55(18)	O4-P1-C25	108.97(17)
O3-P1-C25	103.36(18)	O8-P2-O22	110.58(15)
O8-P2-O16	112.08(15)	O22-P2-O16	107.19(15)
O8-P2-C29	110.47(17)	O22-P2-C29	109.02(17)
O16-P2-C29	107.36(16)	O24-P3-O25	108.91(14)
O24-P3-O26	110.14(15)	O25-P3-O26	111.68(14)
O24-P3-C36	111.62(17)	O25-P3-C36	107.71(16)
O26-P3-C36	106.78(16)	Cu1-O1-H1A	132.(3)
Cu1-O1-H1B	122.(3)	H1A-O1-H1B	105.(5)
P1-O2-Cu1	147.86(17)	P1-O3-H3'	120.(3)
P1-O4-Cu1	133.18(16)	Mo2-O7-Mo1	122.10(14)
P2-O8-Mo1	126.29(14)	P2-O8-Mo5	129.52(14)
Mo1-O8-Mo5	90.25(9)	Mo1-O9-Mo5	125.44(14)
Mo4-O12-Mo5	123.04(13)	Mo3-O15-Mo4	150.19(14)
P2-O16-Mo2	121.65(14)	P2-O16-Mo3	135.96(14)
Mo2-O16-Mo3	101.85(10)	Mo3-O19-Mo2	117.77(13)
Mo3-O19-H19'	121.(3)	Mo2-O19-H19'	116.(3)

P2-O22-Cu2	135.74(16)	P2-O22-Mo4	117.43(14)
Cu2-O22-Mo4	101.65(10)	Cu2-O23-H23A	109.(3)
Cu2-O23-H23B	136.(3)	H23A-O23-H23B	112.(5)
P3-O24-Mo3	120.07(15)	P3-O25-Mo5	127.31(14)
P3-O25-Mo4	136.13(15)	Mo5-O25-Mo4	95.53(9)
P3-O26-Mo1	125.44(14)	P3-O26-Mo2	130.87(15)
Mo1-O26-Mo2	92.20(9)	C1-N1-C5	118.2(3)
C1-N1-Cu1	128.1(3)	C5-N1-Cu1	113.6(3)
C12-N2-C9	118.3(3)	C12-N2-Cu1	129.0(3)
C9-N2-Cu1	112.7(3)	C13-N3-C20	118.2(3)
C13-N3-Cu2	130.1(3)	C20-N3-Cu2	111.6(2)
C24-N4-C21	118.3(3)	C24-N4-Cu2	128.8(3)
C21-N4-Cu2	112.6(3)	N1-C1-C2	122.1(4)
N1-C1-H1	119.0	C2-C1-H1	119.0
C3-C2-C1	119.5(4)	C3-C2-H2	120.2
C1-C2-H2	120.2	C2-C3-C4	119.7(4)
C2-C3-H3	120.1	C4-C3-H3	120.1
C5-C4-C3	116.7(4)	C5-C4-C6	118.2(4)
C3-C4-C6	125.1(4)	N1-C5-C4	123.7(4)
N1-C5-C9	115.8(3)	C4-C5-C9	120.6(4)
C7-C6-C4	121.7(4)	C7-C6-H6	119.1
C4-C6-H6	119.1	C6-C7-C8	120.9(4)

C6-C7-H7	119.5	C8-C7-H7	119.5
C9-C8-C10	117.2(4)	C9-C8-C7	118.2(4)
C10-C8-C7	124.6(4)	N2-C9-C8	123.3(4)
N2-C9-C5	116.4(3)	C8-C9-C5	120.3(4)
C11-C10-C8	119.2(4)	C11-C10-H10	120.4
C8-C10-H10	120.4	C10-C11-C12	119.8(4)
C10-C11-H11	120.1	C12-C11-H11	120.1
N2-C12-C11	122.2(4)	N2-C12-H12	118.9
C11-C12-H12	118.9	N3-C13-C14	122.0(4)
N3-C13-H13	119.0	C14-C13-H13	119.0
C15-C14-C13	120.3(4)	C15-C14-H14	119.8
C13-C14-H14	119.8	C14-C15-C16	119.2(4)
C14-C15-H15	120.4	C16-C15-H15	120.4
C20-C16-C15	116.8(4)	C20-C16-C17	118.7(4)
C15-C16-C17	124.2(4)	C18-C17-C16	121.1(4)
C18-C17-H17	119.4	C16-C17-H17	119.4
C17-C18-C19	121.1(4)	C17-C18-H18	119.5
C19-C18-H18	119.5	C21-C19-C22	116.7(4)
C21-C19-C18	118.6(4)	C22-C19-C18	124.7(4)
N3-C20-C16	123.5(4)	N3-C20-C21	116.4(3)
C16-C20-C21	120.1(4)	N4-C21-C19	123.4(4)
N4-C21-C20	116.2(3)	C19-C21-C20	120.4(4)

C23-C22-C19	119.2(4)	C23-C22-H22	120.4
C19-C22-H22	120.4	C22-C23-C24	119.9(4)
C22-C23-H23	120.1	C24-C23-H23	120.1
N4-C24-C23	122.4(4)	N4-C24-H24	118.8
C23-C24-H24	118.8	C26-C25-P1	115.8(3)
C26-C25-H25A	108.3	P1-C25-H25A	108.3
C26-C25-H25B	108.3	P1-C25-H25B	108.3
H25A-C25-H25B	107.4	C28-C26-C27	117.7(4)
C28-C26-C25	120.6(4)	C27-C26-C25	121.7(4)
C28-C27-C26	120.0(4)	C28-C27-H27	120.0
C26-C27-H27	120.0	C26-C28-C27	122.3(4)
C26-C28-H28	118.9	C27-C28-H28	118.9
C30-C29-P2	110.3(3)	C30-C29-H29A	109.6
P2-C29-H29A	109.6	C30-C29-H29B	109.6
P2-C29-H29B	109.6	H29A-C29-H29B	108.1
C33-C30-C31	119.1(4)	C33-C30-C29	121.0(3)
C31-C30-C29	119.7(3)	C32-C31-C30	120.3(4)
C32-C31-H31	119.8	C30-C31-H31	119.8
C31-C32-C35	120.8(4)	C31-C32-H32	119.6
C35-C32-H32	119.6	C30-C33-C34	120.6(4)
C30-C33-H33	119.7	C34-C33-H33	119.7
C35-C34-C33	120.1(4)	C35-C34-H34	119.9

C33-C34-H34	119.9	C34-C35-C32	118.9(3)
C34-C35-C36	119.5(3)	C32-C35-C36	121.6(3)
C35-C36-P3	112.5(3)	C35-C36-H36A	109.1
P3-C36-H36A	109.1	C35-C36-H36B	109.1
P3-C36-H36B	109.1	H36A-C36-H36B	107.8

Table S54. Anisotropic atomic displacement parameters ($Å^2$) for $[Cu_2(o\text{-phen})_2(H_2O)_2(1,4\text{-}HO_3PC_8H_8PO_3H)][Cu(o\text{-phen})(H_2O)Mo_5O_{14}(OH)(1,4\text{-}O_3PC_8H_8O_3P)]_2$ (**7**).

The anisotropic atomic displacement factor exponent takes the form: $-2\pi^2[\ h^2\ a^{*2}\ U_{11} + ... + 2\ h\ k\ a^*\ b^*\ U_{12}\ ]$

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
Mo1	0.00969(16)	0.01294(18)	0.00717(17)	0.00018(14)	0.00111(13)	-0.00001(14)
Mo2	0.01079(16)	0.01254(18)	0.00715(17)	0.00091(14)	-0.00224(13)	0.00190(14)
Mo3	0.01121(16)	0.01161(18)	0.00662(17)	0.00198(14)	-0.00015(13)	-0.00051(14)
Mo4	0.00977(16)	0.01099(17)	0.00722(17)	0.00130(14)	-0.00070(13)	0.00114(13)
Mo5	0.01187(16)	0.00983(17)	0.00580(17)	0.00177(14)	0.00062(13)	0.00011(13)
Cu1	0.0116(2)	0.0098(2)	0.0091(3)	0.00094(19)	-0.00066(19)	-0.00272(19)
Cu2	0.0133(2)	0.0117(3)	0.0066(2)	0.00136(19)	-0.00186(19)	-0.00342(19)
P1	0.0125(5)	0.0084(5)	0.0083(5)	0.0005(4)	-0.0006(4)	-0.0027(4)
P2	0.0097(5)	0.0088(5)	0.0057(5)	-0.0003(4)	-0.0008(4)	0.0011(4)
P3	0.0113(5)	0.0097(5)	0.0050(5)	0.0004(4)	0.0007(4)	-0.0006(4)
O1	0.0187(16)	0.0302(18)	0.0083(16)	-0.0005(14)	0.0006(12)	0.0104(14)
O2	0.0146(14)	0.0150(15)	0.0079(14)	-0.0014(11)	0.0002(11)	-0.0018(11)
O3	0.0140(14)	0.0158(15)	0.0130(15)	-0.0047(12)	-0.0008(12)	-0.0034(12)
O4	0.0136(13)	0.0112(14)	0.0115(15)	0.0033(11)	-0.0015(11)	-0.0051(11)
O5	0.0167(14)	0.0211(16)	0.0162(16)	0.0017(13)	0.0052(12)	0.0040(12)
O6	0.0150(14)	0.0215(16)	0.0131(15)	-0.0011(12)	-0.0001(12)	-0.0019(12)
O7	0.0109(13)	0.0139(14)	0.0099(14)	0.0005(11)	0.0003(11)	0.0037(11)
O8	0.0115(13)	0.0120(14)	0.0079(14)	0.0008(11)	-0.0002(11)	0.0009(11)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
O9	0.0145(13)	0.0107(14)	0.0083(14)	0.0001(11)	0.0033(11)	-0.0006(11)
O10	0.0171(14)	0.0200(16)	0.0099(15)	0.0033(12)	0.0003(11)	0.0006(12)
O11	0.0174(14)	0.0111(14)	0.0130(15)	0.0022(12)	-0.0003(12)	0.0005(12)
O12	0.0125(13)	0.0098(14)	0.0082(14)	0.0011(11)	-0.0001(11)	0.0025(11)
O13	0.0167(14)	0.0173(15)	0.0148(16)	0.0014(12)	0.0032(12)	0.0038(12)
O14	0.0121(13)	0.0170(15)	0.0091(14)	0.0029(12)	-0.0018(11)	-0.0006(11)
O15	0.0090(13)	0.0149(15)	0.0082(14)	0.0018(11)	0.0001(11)	0.0000(11)
O16	0.0120(13)	0.0100(14)	0.0058(13)	0.0001(11)	-0.0003(10)	0.0004(11)
O17	0.0235(15)	0.0147(15)	0.0101(15)	0.0037(12)	-0.0020(12)	-0.0020(12)
O18	0.0162(14)	0.0209(16)	0.0128(15)	0.0046(12)	0.0019(12)	0.0000(12)
O19	0.0171(14)	0.0144(15)	0.0047(14)	0.0008(12)	-0.0023(11)	-0.0005(12)
O20	0.0171(14)	0.0219(16)	0.0084(15)	0.0024(12)	-0.0039(12)	0.0003(12)
O21	0.0170(14)	0.0159(15)	0.0153(16)	0.0017(12)	-0.0033(12)	0.0013(12)
O22	0.0131(13)	0.0096(14)	0.0058(13)	0.0006(11)	0.0001(10)	0.0008(11)
O23	0.0300(17)	0.0107(15)	0.0077(15)	0.0003(12)	0.0017(13)	-0.0046(13)
O24	0.0103(13)	0.0114(14)	0.0059(14)	0.0003(11)	-0.0002(10)	0.0023(11)
O25	0.0125(13)	0.0090(14)	0.0064(14)	-0.0005(11)	0.0019(11)	-0.0005(11)
O26	0.0107(13)	0.0111(14)	0.0059(14)	0.0006(11)	0.0010(11)	-0.0004(11)
N1	0.0108(16)	0.0121(17)	0.0124(17)	-0.0040(14)	0.0012(13)	-0.0034(13)
N2	0.0086(15)	0.0130(17)	0.0117(17)	0.0019(14)	-0.0021(13)	-0.0017(13)
N3	0.0103(16)	0.0141(18)	0.0090(17)	-0.0005(14)	-0.0010(13)	-0.0002(13)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
N4	0.0120(16)	0.0105(17)	0.0092(17)	0.0001(13)	0.0004(13)	-0.0003(13)
C1	0.017(2)	0.020(2)	0.017(2)	-0.0070(18)	0.0022(18)	-0.0020(17)
C2	0.024(2)	0.023(2)	0.017(2)	-0.0112(19)	0.0026(19)	-0.0071(19)
C3	0.018(2)	0.017(2)	0.024(2)	-0.0096(19)	0.0054(18)	-0.0046(18)
C4	0.0096(19)	0.009(2)	0.024(2)	-0.0038(17)	0.0067(17)	0.0007(16)
C5	0.0101(18)	0.0070(19)	0.017(2)	0.0011(16)	0.0048(16)	0.0017(15)
C6	0.019(2)	0.010(2)	0.030(3)	0.0023(18)	0.0023(19)	-0.0042(17)
C7	0.017(2)	0.015(2)	0.024(2)	0.0065(18)	0.0057(18)	0.0003(17)
C8	0.0093(19)	0.016(2)	0.019(2)	0.0051(17)	0.0000(17)	0.0012(16)
C9	0.0074(18)	0.013(2)	0.016(2)	0.0027(16)	0.0020(16)	0.0005(15)
C10	0.014(2)	0.020(2)	0.018(2)	0.0089(18)	0.0006(17)	-0.0006(17)
C11	0.0118(19)	0.023(2)	0.010(2)	0.0013(17)	-0.0024(16)	-0.0009(17)
C12	0.0103(19)	0.014(2)	0.016(2)	0.0034(17)	-0.0013(16)	0.0005(16)
C13	0.0132(19)	0.016(2)	0.011(2)	-0.0020(17)	0.0003(16)	-0.0001(16)
C14	0.017(2)	0.024(2)	0.005(2)	-0.0007(17)	0.0015(16)	0.0036(18)
C15	0.016(2)	0.028(3)	0.013(2)	-0.0083(18)	-0.0025(17)	0.0009(18)
C16	0.0094(19)	0.019(2)	0.016(2)	-0.0068(17)	0.0019(16)	0.0006(16)
C17	0.016(2)	0.020(2)	0.027(3)	-0.0055(19)	-0.0047(19)	-0.0031(18)
C18	0.014(2)	0.015(2)	0.031(3)	-0.0029(19)	0.0023(18)	-0.0047(17)
C19	0.0093(19)	0.011(2)	0.021(2)	-0.0023(17)	0.0021(16)	-0.0003(16)
C20	0.0090(18)	0.015(2)	0.013(2)	-0.0028(16)	-0.0005(15)	0.0014(15)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
C21	0.0097(18)	0.010(2)	0.014(2)	-0.0025(16)	0.0022(16)	-0.0006(15)
C22	0.015(2)	0.014(2)	0.024(2)	0.0087(18)	0.0047(17)	-0.0019(17)
C23	0.024(2)	0.022(2)	0.014(2)	0.0034(18)	0.0020(18)	0.0025(18)
C24	0.016(2)	0.014(2)	0.011(2)	0.0009(17)	-0.0014(16)	0.0009(16)
C25	0.016(2)	0.012(2)	0.023(2)	0.0032(18)	0.0013(18)	0.0019(17)
C26	0.0098(19)	0.0096(19)	0.020(2)	0.0011(17)	0.0021(17)	0.0047(16)
C27	0.020(2)	0.015(2)	0.012(2)	-0.0008(17)	-0.0008(17)	0.0051(17)
C28	0.017(2)	0.008(2)	0.021(2)	-0.0045(17)	-0.0031(18)	0.0009(16)
C29	0.016(2)	0.0105(19)	0.0081(19)	0.0030(16)	0.0011(16)	0.0014(16)
C30	0.0108(18)	0.010(2)	0.0095(19)	0.0009(16)	0.0007(15)	0.0058(15)
C31	0.0128(19)	0.013(2)	0.011(2)	-0.0005(16)	-0.0015(16)	-0.0012(16)
C32	0.015(2)	0.013(2)	0.009(2)	0.0031(16)	0.0037(16)	-0.0014(16)
C33	0.0145(19)	0.009(2)	0.010(2)	0.0006(16)	0.0056(16)	0.0013(15)
C34	0.0121(19)	0.010(2)	0.012(2)	-0.0018(16)	0.0003(16)	-0.0013(15)
C35	0.0149(19)	0.010(2)	0.0064(19)	0.0004(15)	0.0006(15)	0.0038(16)
C36	0.016(2)	0.010(2)	0.008(2)	0.0009(16)	0.0020(16)	-0.0004(16)

Table S55. Hydrogen atomic coordinates and isotropic atomic displacement parameters ($Å^2$) for $[Cu_2(o\text{-phen})_2(H_2O)_2(1,4\text{-}HO_3PC_8H_8PO_3H)][Cu(o\text{-phen})(H_2O)Mo_5O_{14}(OH)(1,4\text{-}O_3PC_8H_8O_3P)]_2$ (**7**).

	x/a	y/b	z/c	U(eq)
H1A	0.4302(8)	0.542(5)	0.0324(18)	0.023
H1B	0.4318(8)	0.542(5)	0.1016(17)	0.023
H3'	0.4628(4)	0.335(5)	-0.156(2)	0.017
H19'	0.3631(8)	0.318(4)	0.4918(12)	0.015
H23A	0.3261(8)	0.492(3)	0.088(2)	0.019
H23B	0.3318(8)	0.438(5)	0.0233(12)	0.019
H1	0.4443	0.2104	-0.0562	0.022
H2	0.4200	0.0095	-0.0722	0.026
H3	0.4107	-0.1283	0.0254	0.024
H6	0.4144	-0.1634	0.1621	0.024
H7	0.4280	-0.0834	0.2703	0.022
H10	0.4497	0.1140	0.3440	0.02
H11	0.4696	0.3298	0.3431	0.018
H12	0.4777	0.4385	0.2349	0.016
H13	0.3003	0.3362	-0.0443	0.016
H14	0.2709	0.2502	-0.1263	0.018
H15	0.2463	0.0611	-0.0976	0.023
H17	0.2337	-0.1170	-0.0018	0.025
H18	0.2372	-0.1921	0.1135	0.024

	x/a	y/b	z/c	U(eq)
H22	0.2585	-0.1669	0.2393	0.021
H23	0.2894	-0.0608	0.3088	0.024
H24	0.3142	0.1112	0.2603	0.016
H25A	0.5244	0.2295	-0.1146	0.021
H25B	0.5298	0.2973	-0.0381	0.021
H27	0.4974	0.0175	-0.1243	0.019
H28	0.4844	-0.1840	-0.0675	0.018
H29A	0.3935	0.1915	0.2157	0.014
H29B	0.3691	0.0963	0.2219	0.014
H31	0.3982	0.3318	0.1020	0.015
H32	0.3914	0.3417	-0.0207	0.015
H33	0.3481	0.0198	0.1145	0.013
H34	0.3404	0.0329	-0.0087	0.014
H36A	0.3424	0.1538	-0.1165	0.014
H36B	0.3712	0.1680	-0.1337	0.014

Table S56. Sample and crystal data for (H-4,4'bpy)[{Ni(H-4,4'-bpy)(H_2O)$_4$}Mo_5O_{15}(1,2-$O_3PC_8H_8PO_3$)]·4H_2O (**8**·4H_2O).

Identification code	(H-4,4'bpy)[{Ni(H-4,4'-bpy)(H_2O)$_4$}Mo_5O_{15}(1,2-$O_3PC_8H_8PO_3$)]·4H_2O	
Chemical formula	$C_{28}H_{42}Mo_5N_4NiO_{29}P_2$	
Formula weight	1499.00	
Temperature	90(2) K	
Wavelength	0.71073 Å	
Crystal size	0.086 x 0.170 x 0.400 mm	
Crystal system	Monoclinic	
Space group	P21/c	
Unit cell dimensions	a = 15.6642(11) Å	α = 90°
	b = 17.0366(13) Å	β = 110.3660(10)°
	c = 18.1045(13) Å	γ = 90°
Volume	4529.4(6) Å^3	
Z	4	
Density (calculated)	2.198 g/cm^3	
Absorption coefficient	1.922 mm^{-1}	
F(000)	2952	

Table S57. Data collection and structure refinement for (H-4,4'bpy)[{Ni(H-4,4'-bpy)(H_2O)$_4$}Mo_5O_{15}(1,2-$O_3PC_8H_8PO_3$)]·4H_2O (**8**·4H_2O).

Theta range for data collection	1.83 to 25.68°	
Index ranges	-19<=h<=18, -20<=k<=20, -21<=l<=22	
Reflections collected	31509	
Independent reflections	8602 [R(int) = 0.0217]	
Coverage of independent reflections	100.0%	
Max. and min. transmission	0.8520 and 0.5140	
Refinement method	Full-matrix least-squares on F^2	
Refinement program	SHELXL-2013 (Sheldrick, 2013)	
Function minimized	$\Sigma\, w(F_o^2 - F_c^2)^2$	
Data / restraints / parameters	8602 / 10 / 677	
Goodness-of-fit on F^2	1.031	
Δ/σ_{max}	0.001	
Final R indices	7675 data; I>2σ(I)	R1 = 0.0183, wR2 = 0.0428
	all data	R1 = 0.0229, wR2 = 0.0448
Weighting scheme	$w=1/[\sigma^2(F_o^2)+(0.0187P)^2+5.9249P]$ where $P=(F_o^2+2F_c^2)/3$	
Largest diff. peak and hole	0.699 and -0.598 eÅ^{-3}	
R.M.S. deviation from mean	0.077 eÅ^{-3}	

Table S58. Atomic coordinates and equivalent isotropic atomic displacement parameters ($Å^2$) for (H-4,4'bpy)[{Ni(H-4,4'-bpy)$(H_2O)_4$}Mo_5O_{15}(1,2-$O_3PC_8H_8PO_3$)]·$4H_2O$ (**8**·$4H_2O$).

U(eq) is defined as one third of the trace of the orthogonalized U_{ij} tensor.

	x/a	**y/b**	**z/c**	**U(eq)**
Mo1	0.11372(2)	0.13930(2)	0.92861(2)	0.00886(5)
Mo2	0.04462(2)	0.31458(2)	0.97417(2)	0.00877(5)
Mo3	0.23245(2)	0.42683(2)	0.05549(2)	0.00785(5)
Mo4	0.42252(2)	0.32077(2)	0.07984(2)	0.00891(5)
Mo5	0.36168(2)	0.13227(2)	0.03320(2)	0.00884(5)
Ni1	0.33465(2)	0.49020(2)	0.26712(2)	0.00902(7)
P1	0.23556(4)	0.29390(3)	0.91415(3)	0.00821(12)
P2	0.23390(4)	0.23197(3)	0.11265(3)	0.00771(12)
O1	0.23896(11)	0.10683(10)	0.97377(9)	0.0114(3)
O2	0.08138(11)	0.09345(10)	0.83935(9)	0.0121(3)
O3	0.17666(11)	0.22721(9)	0.86926(9)	0.0113(3)
O4	0.02864(11)	0.22633(9)	0.90377(9)	0.0111(3)
O5	0.06657(11)	0.08112(10)	0.98151(10)	0.0132(4)
O6	0.15129(11)	0.22691(9)	0.03412(9)	0.0090(3)
O7	0.18075(11)	0.34930(9)	0.94900(9)	0.0092(3)
O8	0.98867(11)	0.38351(10)	0.90502(10)	0.0143(4)
O9	0.96831(11)	0.28808(10)	0.01730(10)	0.0149(4)
O10	0.12084(11)	0.38176(9)	0.05773(9)	0.0094(3)

	x/a	y/b	z/c	U(eq)
O11	0.19580(12)	0.50927(10)	0.00068(10)	0.0133(4)
O12	0.29080(11)	0.30612(9)	0.11437(9)	0.0088(3)
O13	0.34502(11)	0.41092(9)	0.03807(9)	0.0102(3)
O14	0.27036(11)	0.46198(9)	0.15147(9)	0.0104(3)
O15	0.47690(11)	0.35184(10)	0.17561(10)	0.0139(4)
O16	0.49897(12)	0.33961(10)	0.03445(11)	0.0172(4)
O17	0.43989(11)	0.20974(9)	0.10038(9)	0.0106(3)
O18	0.32012(11)	0.26109(9)	0.98018(9)	0.0100(3)
O19	0.40756(11)	0.12457(10)	0.95929(10)	0.0136(4)
O20	0.40210(11)	0.04848(10)	0.08589(10)	0.0136(4)
O21	0.28973(11)	0.15720(9)	0.12097(9)	0.0097(3)
O22	0.35097(13)	0.37138(11)	0.29573(12)	0.0173(4)
O23	0.45700(12)	0.48569(12)	0.25107(11)	0.0154(4)
O24	0.41047(13)	0.51558(11)	0.38475(10)	0.0129(4)
O25	0.21502(13)	0.48835(11)	0.29081(12)	0.0165(4)
O90	0.39587(15)	0.51311(12)	0.93275(12)	0.0270(5)
O91	0.06509(13)	0.46705(11)	0.16265(11)	0.0172(4)
O92	0.43625(14)	0.22607(13)	0.27870(13)	0.0285(5)
O93	0.53195(15)	0.56976(13)	0.16777(14)	0.0305(5)
N1	0.30976(13)	0.60891(12)	0.23993(11)	0.0109(4)
N2	0.22089(15)	0.00332(12)	0.12687(13)	0.0163(5)

	x/a	y/b	z/c	U(eq)
N3	0.58696(17)	0.27022(17)	0.40881(16)	0.0321(6)
N4	0.9834(2)	0.24759(17)	0.74567(15)	0.0428(8)
C1	0.27312(16)	0.15064(14)	0.34672(13)	0.0100(5)
C2	0.19230(16)	0.12057(14)	0.27944(14)	0.0101(5)
C3	0.15336(17)	0.04939(14)	0.29097(14)	0.0139(5)
C4	0.07563(18)	0.02012(15)	0.23449(15)	0.0164(5)
C5	0.03468(17)	0.06133(15)	0.16463(15)	0.0155(5)
C6	0.07223(17)	0.13154(15)	0.15224(14)	0.0141(5)
C7	0.15084(16)	0.16209(14)	0.20858(14)	0.0105(5)
C8	0.18865(16)	0.23927(13)	0.19152(13)	0.0099(5)
C9	0.24408(17)	0.63005(14)	0.17198(14)	0.0130(5)
C10	0.35263(17)	0.66648(14)	0.28967(15)	0.0137(5)
C11	0.22130(17)	0.70757(14)	0.15153(15)	0.0135(5)
C12	0.33320(17)	0.74575(14)	0.27345(15)	0.0147(5)
C13	0.26671(16)	0.76749(14)	0.20207(14)	0.0122(5)
C14	0.24778(17)	0.85133(14)	0.17884(14)	0.0129(5)
C15	0.16607(17)	0.87396(15)	0.12082(15)	0.0156(5)
C16	0.31353(17)	0.90883(15)	0.21252(15)	0.0155(5)
C17	0.15397(17)	0.95064(15)	0.09524(15)	0.0172(5)
C18	0.29827(18)	0.98482(15)	0.18520(15)	0.0169(5)
C19	0.6234(2)	0.20092(19)	0.43976(18)	0.0282(7)

	x/a	y/b	z/c	U(eq)
C20	0.6257(2)	0.3349(2)	0.4478(2)	0.0358(8)
C21	0.69943(19)	0.19431(18)	0.50772(17)	0.0239(6)
C22	0.7018(2)	0.33379(19)	0.5154(2)	0.0336(7)
C23	0.74078(19)	0.26194(17)	0.54662(17)	0.0236(6)
C24	0.82488(19)	0.25719(17)	0.61745(16)	0.0214(6)
C25	0.8521(2)	0.31874(18)	0.67234(19)	0.0320(8)
C26	0.88061(19)	0.19111(16)	0.63095(15)	0.0201(6)
C27	0.9319(3)	0.3114(2)	0.73626(17)	0.0412(10)
C28	0.9599(2)	0.18775(18)	0.69533(17)	0.0307(7)

Table S59. Bond lengths (Å) for (H-4,4'bpy)[{Ni(H-4,4'-bpy)(H_2O)$_4$}Mo_5O_{15}(1,2-$O_3PC_8H_8PO_3$)]·4H_2O (**8**·4H_2O).

Mo1-O2	1.7055(16)	Mo1-O5	1.7140(17)
Mo1-O1	1.9253(16)	Mo1-O4	1.9389(16)
Mo1-O3	2.2597(16)	Mo1-O6	2.3325(16)
Mo2-O9	1.7005(17)	Mo2-O8	1.7177(16)
Mo2-O4	1.9301(16)	Mo2-O10	1.9408(16)
Mo2-O6	2.2223(16)	Mo2-O7	2.4044(16)
Mo3-O11	1.7001(16)	Mo3-O14	1.7356(16)
Mo3-O13	1.9155(16)	Mo3-O10	1.9228(16)
Mo3-O7	2.2429(16)	Mo3-O12	2.3501(16)
Mo4-O16	1.7021(17)	Mo4-O15	1.7275(17)
Mo4-O17	1.9287(16)	Mo4-O13	1.9405(16)
Mo4-O18	2.2016(16)	Mo4-O12	2.3681(16)
Mo5-O20	1.7112(16)	Mo5-O19	1.7301(17)
Mo5-O1	1.8977(16)	Mo5-O17	1.9197(16)
Mo5-O21	2.2851(16)	Mo5-O18	2.3930(16)
Ni1-O23	2.0374(19)	Ni1-O14	2.0401(16)
Ni1-O25	2.0630(19)	Ni1-O22	2.0831(19)
Ni1-N1	2.086(2)	Ni1-O24	2.0911(18)
P1-O3	1.5103(17)	P1-O18	1.5473(17)
P1-O7	1.5502(16)	P1-C1	1.797(2)

P2-O21	1.5226(17)	P2-O12	1.5400(16)
P2-O6	1.5566(16)	P2-C8	1.806(2)
O22-H22C	0.78(3)	O22-H22D	0.75(3)
O23-H23C	0.79(3)	O23-H23D	0.84(3)
O24-H24C	0.80(3)	O24-H24D	0.80(3)
O25-H25C	0.73(3)	O25-H25D	0.84(3)
O90-H90A	0.841(18)	O90-H90B	0.839(18)
O91-H91A	0.806(17)	O91-H91B	0.817(17)
O92-H92A	0.863(18)	O92-H92B	0.842(18)
O93-H93A	0.876(18)	O93-H93B	0.849(18)
N1-C10	1.343(3)	N1-C9	1.349(3)
N2-C18	1.339(3)	N2-C17	1.347(3)
N2-H2'	0.884(17)	N3-C20	1.335(4)
N3-C19	1.346(4)	N4-C27	1.329(5)
N4-C28	1.331(4)	N4-H4'	0.889(19)
C1-C2	1.509(3)	C1-P1	1.797(2)
C1-H1A	0.99	C1-H1B	0.99
C2-C3	1.405(3)	C2-C7	1.410(3)
C3-C4	1.382(4)	C3-H3	0.95
C4-C5	1.393(4)	C4-H4	0.95
C5-C6	1.385(3)	C5-H5	0.95
C6-C7	1.398(3)	C6-H6	0.95

C7-C8	1.517(3)	C8-H8A	0.99
C8-H8B	0.99	C9-C11	1.384(3)
C9-H9	0.95	C10-C12	1.393(3)
C10-H10	0.95	C11-C13	1.390(3)
C11-H11	0.95	C12-C13	1.398(3)
C12-H12	0.95	C13-C14	1.489(3)
C14-C15	1.398(3)	C14-C16	1.398(4)
C15-C17	1.377(4)	C15-H15	0.95
C16-C18	1.377(4)	C16-H16	0.95
C17-H17	0.95	C18-H18	0.95
C19-C21	1.387(4)	C19-H19	0.95
C20-C22	1.379(5)	C20-H20	0.95
C21-C23	1.388(4)	C21-H21	0.95
C22-C23	1.397(4)	C22-H22A	0.95
C23-C24	1.487(4)	C24-C26	1.393(4)
C24-C25	1.405(4)	C25-C27	1.383(5)
C25-H25A	0.95	C26-C28	1.377(4)
C26-H26	0.95	C27-H27	0.95
C28-H28	0.95		

Table S60. Bond angles (°) for (H-4,4'bpy)[{Ni(H-4,4'-bpy)(H_2O)$_4$}Mo_5O_{15}(1,2-$O_3PC_8H_8PO_3$)]·4H_2O (**8**·4H_2O).

O2-Mo1-O5	102.77(8)	O2-Mo1-O1	101.16(7)
O5-Mo1-O1	99.79(7)	O2-Mo1-O4	100.20(7)
O5-Mo1-O4	100.18(7)	O1-Mo1-O4	146.59(7)
O2-Mo1-O3	83.56(7)	O5-Mo1-O3	173.27(7)
O1-Mo1-O3	81.01(6)	O4-Mo1-O3	76.20(6)
O2-Mo1-O6	167.47(7)	O5-Mo1-O6	86.98(7)
O1-Mo1-O6	84.64(6)	O4-Mo1-O6	69.99(6)
O3-Mo1-O6	86.44(6)	O9-Mo2-O8	104.73(8)
O9-Mo2-O4	98.39(8)	O8-Mo2-O4	97.77(7)
O9-Mo2-O10	97.97(7)	O8-Mo2-O10	100.63(7)
O4-Mo2-O10	151.31(7)	O9-Mo2-O6	97.26(7)
O8-Mo2-O6	157.16(7)	O4-Mo2-O6	72.64(6)
O10-Mo2-O6	82.04(6)	O9-Mo2-O7	164.54(7)
O8-Mo2-O7	87.42(7)	O4-Mo2-O7	89.21(6)
O10-Mo2-O7	69.94(6)	O6-Mo2-O7	72.07(6)
O11-Mo3-O14	103.16(8)	O11-Mo3-O13	99.44(7)
O14-Mo3-O13	101.53(7)	O11-Mo3-O10	102.55(8)
O14-Mo3-O10	96.51(7)	O13-Mo3-O10	147.44(7)
O11-Mo3-O7	92.13(7)	O14-Mo3-O7	163.56(7)
O13-Mo3-O7	81.54(6)	O10-Mo3-O7	73.98(6)

O11-Mo3-O12	170.65(7)	O14-Mo3-O12	84.50(7)
O13-Mo3-O12	73.49(6)	O10-Mo3-O12	81.60(6)
O7-Mo3-O12	80.89(6)	O16-Mo4-O15	104.05(8)
O16-Mo4-O17	101.67(8)	O15-Mo4-O17	96.88(7)
O16-Mo4-O13	96.46(8)	O15-Mo4-O13	99.89(7)
O17-Mo4-O13	151.47(7)	O16-Mo4-O18	97.23(7)
O15-Mo4-O18	158.15(7)	O17-Mo4-O18	73.67(6)
O13-Mo4-O18	82.39(6)	O16-Mo4-O12	166.25(7)
O15-Mo4-O12	86.44(7)	O17-Mo4-O12	85.59(6)
O13-Mo4-O12	72.66(6)	O18-Mo4-O12	73.41(6)
O20-Mo5-O19	101.26(8)	O20-Mo5-O1	103.34(8)
O19-Mo5-O1	98.96(7)	O20-Mo5-O17	100.93(7)
O19-Mo5-O17	101.35(7)	O1-Mo5-O17	144.43(7)
O20-Mo5-O21	86.78(7)	O19-Mo5-O21	171.88(7)
O1-Mo5-O21	77.83(6)	O17-Mo5-O21	78.00(6)
O20-Mo5-O18	170.02(7)	O19-Mo5-O18	83.54(7)
O1-Mo5-O18	84.35(6)	O17-Mo5-O18	69.40(6)
O21-Mo5-O18	88.70(5)	O23-Ni1-O14	89.86(7)
O23-Ni1-O25	175.33(8)	O14-Ni1-O25	92.87(7)
O23-Ni1-O22	87.63(8)	O14-Ni1-O22	90.01(7)
O25-Ni1-O22	88.58(8)	O23-Ni1-N1	96.36(8)
O14-Ni1-N1	90.08(7)	O25-Ni1-N1	87.43(8)

O22-Ni1-N1	176.01(8)	O23-Ni1-O24	85.64(7)
O14-Ni1-O24	175.26(7)	O25-Ni1-O24	91.52(8)
O22-Ni1-O24	88.32(7)	N1-Ni1-O24	91.90(7)
O3-P1-O18	110.02(9)	O3-P1-O7	110.86(9)
O18-P1-O7	110.60(9)	O3-P1-C1	108.25(10)
O18-P1-C1	108.67(10)	O7-P1-C1	108.36(10)
O21-P2-O12	112.07(9)	O21-P2-O6	108.08(9)
O12-P2-O6	110.80(9)	O21-P2-C8	110.46(10)
O12-P2-C8	108.13(10)	O6-P2-C8	107.20(10)
Mo5-O1-Mo1	148.94(9)	P1-O3-Mo1	121.91(9)
Mo2-O4-Mo1	121.45(8)	P2-O6-Mo2	130.82(9)
P2-O6-Mo1	132.54(9)	Mo2-O6-Mo1	95.60(6)
P1-O7-Mo3	128.54(9)	P1-O7-Mo2	125.49(9)
Mo3-O7-Mo2	93.48(6)	Mo3-O10-Mo2	122.43(8)
P2-O12-Mo3	125.29(9)	P2-O12-Mo4	129.23(9)
Mo3-O12-Mo4	91.43(5)	Mo3-O13-Mo4	122.32(8)
Mo3-O14-Ni1	169.59(10)	Mo5-O17-Mo4	122.41(8)
P1-O18-Mo4	130.54(9)	P1-O18-Mo5	132.96(9)
Mo4-O18-Mo5	94.36(6)	P2-O21-Mo5	119.48(9)
Ni1-O22-H22C	126.(2)	Ni1-O22-H22D	117.(2)
H22C-O22-H22D	105.(3)	Ni1-O23-H23C	121.(2)
Ni1-O23-H23D	108.(2)	H23C-O23-H23D	107.(3)

Ni1-O24-H24C	110.(2)	Ni1-O24-H24D	110.(2)
H24C-O24-H24D	98.(3)	Ni1-O25-H25C	119.(2)
Ni1-O25-H25D	114.(2)	H25C-O25-H25D	106.(3)
H90A-O90-H90B	105.(3)	H91A-O91-H91B	106.(3)
H92A-O92-H92B	107.(3)	H93A-O93-H93B	119.(3)
C10-N1-C9	117.5(2)	C10-N1-Ni1	122.77(16)
C9-N1-Ni1	119.64(16)	C18-N2-C17	122.4(2)
C18-N2-H2'	117.6(19)	C17-N2-H2'	119.9(19)
C20-N3-C19	117.1(3)	C27-N4-C28	121.8(3)
C27-N4-H4'	116.(3)	C28-N4-H4'	122.(3)
C2-C1-P1	110.24(16)	C2-C1-H1A	109.6
P1-C1-H1A	109.6	C2-C1-H1B	109.6
P1-C1-H1B	109.6	H1A-C1-H1B	108.1
C3-C2-C7	118.8(2)	C3-C2-C1	117.3(2)
C7-C2-C1	123.8(2)	C4-C3-C2	121.2(2)
C4-C3-H3	119.4	C2-C3-H3	119.4
C3-C4-C5	119.9(2)	C3-C4-H4	120.0
C5-C4-H4	120.0	C6-C5-C4	119.7(2)
C6-C5-H5	120.2	C4-C5-H5	120.2
C5-C6-C7	121.3(2)	C5-C6-H6	119.4
C7-C6-H6	119.4	C6-C7-C2	119.1(2)
C6-C7-C8	118.8(2)	C2-C7-C8	122.1(2)

C7-C8-P2	112.93(16)	C7-C8-H8A	109.0
P2-C8-H8A	109.0	C7-C8-H8B	109.0
P2-C8-H8B	109.0	H8A-C8-H8B	107.8
N1-C9-C11	122.8(2)	N1-C9-H9	118.6
C11-C9-H9	118.6	N1-C10-C12	123.0(2)
N1-C10-H10	118.5	C12-C10-H10	118.5
C9-C11-C13	119.9(2)	C9-C11-H11	120.0
C13-C11-H11	120.0	C10-C12-C13	119.3(2)
C10-C12-H12	120.3	C13-C12-H12	120.3
C11-C13-C12	117.4(2)	C11-C13-C14	120.8(2)
C12-C13-C14	121.7(2)	C15-C14-C16	118.5(2)
C15-C14-C13	121.1(2)	C16-C14-C13	120.3(2)
C17-C15-C14	119.8(2)	C17-C15-H15	120.1
C14-C15-H15	120.1	C18-C16-C14	119.5(2)
C18-C16-H16	120.2	C14-C16-H16	120.2
N2-C17-C15	119.6(2)	N2-C17-H17	120.2
C15-C17-H17	120.2	N2-C18-C16	120.1(2)
N2-C18-H18	120.0	C16-C18-H18	120.0
N3-C19-C21	123.3(3)	N3-C19-H19	118.3
C21-C19-H19	118.3	N3-C20-C22	123.4(3)
N3-C20-H20	118.3	C22-C20-H20	118.3
C19-C21-C23	119.2(3)	C19-C21-H21	120.4

C23-C21-H21	120.4	C20-C22-C23	119.5(3)
C20-C22-H22A	120.2	C23-C22-H22A	120.2
C21-C23-C22	117.4(3)	C21-C23-C24	120.6(3)
C22-C23-C24	121.9(3)	C26-C24-C25	117.5(3)
C26-C24-C23	120.6(2)	C25-C24-C23	121.9(3)
C27-C25-C24	119.2(3)	C27-C25-H25A	120.4
C24-C25-H25A	120.4	C28-C26-C24	120.5(3)
C28-C26-H26	119.7	C24-C26-H26	119.7
N4-C27-C25	120.9(3)	N4-C27-H27	119.6
C25-C27-H27	119.6	N4-C28-C26	120.1(3)
N4-C28-H28	120.0	C26-C28-H28	120.0

Table S61. Torsion angles (°) for (H-4,4'bpy)[{Ni(H-4,4'-bpy)(H_2O)$_4$}Mo_5O_{15}(1,2-$O_3PC_8H_8PO_3$)]·4H_2O (**8**·4H_2O).

O20-Mo5-O1-Mo1	143.47(18)	O19-Mo5-O1-Mo1	-112.62(18)
O17-Mo5-O1-Mo1	11.7(3)	O21-Mo5-O1-Mo1	59.80(17)
O18-Mo5-O1-Mo1	-30.08(18)	O18-P1-O3-Mo1	60.73(13)
O7-P1-O3-Mo1	-61.93(13)	C1-P1-O3-Mo1	179.35(10)
O21-P2-O6-Mo2	-174.47(11)	O12-P2-O6-Mo2	-51.32(14)
C8-P2-O6-Mo2	66.46(14)	O21-P2-O6-Mo1	-9.11(15)
O12-P2-O6-Mo1	114.04(12)	C8-P2-O6-Mo1	-128.18(12)
O3-P1-O7-Mo3	153.47(10)	O18-P1-O7-Mo3	31.15(14)
C1-P1-O7-Mo3	-87.87(13)	O3-P1-O7-Mo2	21.92(13)
O18-P1-O7-Mo2	-100.40(11)	C1-P1-O7-Mo2	140.57(11)
O21-P2-O12-Mo3	153.87(9)	O6-P2-O12-Mo3	33.06(13)
C8-P2-O12-Mo3	-84.15(12)	O21-P2-O12-Mo4	25.75(14)
O6-P2-O12-Mo4	-95.06(12)	C8-P2-O12-Mo4	147.73(11)
O11-Mo3-O14-Ni1	-135.8(5)	O13-Mo3-O14-Ni1	-33.2(6)
O10-Mo3-O14-Ni1	119.6(5)	O7-Mo3-O14-Ni1	66.1(7)
O12-Mo3-O14-Ni1	38.7(5)	O3-P1-O18-Mo4	-168.74(11)
O7-P1-O18-Mo4	-45.93(14)	C1-P1-O18-Mo4	72.91(14)
O3-P1-O18-Mo5	-9.84(15)	O7-P1-O18-Mo5	112.96(12)
C1-P1-O18-Mo5	-128.20(13)	O12-P2-O21-Mo5	-56.92(12)
O6-P2-O21-Mo5	65.46(11)	C8-P2-O21-Mo5	-177.55(10)

P1-C1-C2-C3	-85.3(2)	P1-C1-C2-C7	90.4(2)
C7-C2-C3-C4	-0.3(4)	C1-C2-C3-C4	175.7(2)
C2-C3-C4-C5	0.1(4)	C3-C4-C5-C6	0.0(4)
C4-C5-C6-C7	0.1(4)	C5-C6-C7-C2	-0.2(4)
C5-C6-C7-C8	180.0(2)	C3-C2-C7-C6	0.3(3)
C1-C2-C7-C6	-175.4(2)	C3-C2-C7-C8	-179.9(2)
C1-C2-C7-C8	4.3(3)	C6-C7-C8-P2	-68.1(3)
C2-C7-C8-P2	112.2(2)	O21-P2-C8-C7	-43.12(19)
O12-P2-C8-C7	-166.09(16)	O6-P2-C8-C7	74.40(18)
C10-N1-C9-C11	1.4(4)	Ni1-N1-C9-C11	177.32(18)
C9-N1-C10-C12	-1.1(4)	Ni1-N1-C10-C12	-176.83(19)
N1-C9-C11-C13	-0.1(4)	N1-C10-C12-C13	-0.6(4)
C9-C11-C13-C12	-1.5(4)	C9-C11-C13-C14	175.8(2)
C10-C12-C13-C11	1.8(4)	C10-C12-C13-C14	-175.4(2)
C11-C13-C14-C15	21.9(4)	C12-C13-C14-C15	-161.0(2)
C11-C13-C14-C16	-155.3(2)	C12-C13-C14-C16	21.8(4)
C16-C14-C15-C17	3.4(4)	C13-C14-C15-C17	-173.8(2)
C15-C14-C16-C18	-3.2(4)	C13-C14-C16-C18	174.1(2)
C18-N2-C17-C15	-2.8(4)	C14-C15-C17-N2	-0.5(4)
C17-N2-C18-C16	3.0(4)	C14-C16-C18-N2	0.1(4)
C20-N3-C19-C21	-1.8(4)	C19-N3-C20-C22	2.3(5)
N3-C19-C21-C23	0.1(4)	N3-C20-C22-C23	-1.0(5)

C19-C21-C23-C22	1.2(4)	C19-C21-C23-C24	-177.1(2)
C20-C22-C23-C21	-0.8(4)	C20-C22-C23-C24	177.5(3)
C21-C23-C24-C26	21.9(4)	C22-C23-C24-C26	-156.4(3)
C21-C23-C24-C25	-159.1(3)	C22-C23-C24-C25	22.7(4)
C26-C24-C25-C27	0.3(4)	C23-C24-C25-C27	-178.8(3)
C25-C24-C26-C28	-1.0(4)	C23-C24-C26-C28	178.1(3)
C28-N4-C27-C25	-1.3(5)	C24-C25-C27-N4	0.8(4)
C27-N4-C28-C26	0.6(5)	C24-C26-C28-N4	0.6(4)

Table S62. Anisotropic atomic displacement parameters ($Å^2$) for (H-4,4'bpy)[{Ni(H-4,4'-bpy)$(H_2O)_4$}Mo_5O_{15}(1,2-$O_3PC_8H_8PO_3$)]·4H_2O (**8**·4H_2O).

The anisotropic atomic displacement factor exponent takes the form: $-2\pi^2[h^2 a^{*2} U_{11} + ... + 2 h k a^* b^* U_{12}]$

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
Mo1	0.01064(10)	0.00630(10)	0.00790(10)	-0.00072(8)	0.00104(8)	-0.00041(8)
Mo2	0.00957(10)	0.00818(10)	0.00752(10)	-0.00086(8)	0.00168(8)	0.00090(8)
Mo3	0.01070(10)	0.00577(10)	0.00696(10)	-0.00031(7)	0.00294(8)	0.00003(8)
Mo4	0.00938(10)	0.00728(10)	0.01011(10)	-0.00097(8)	0.00343(8)	-0.00077(8)
Mo5	0.00994(10)	0.00663(10)	0.00908(10)	-0.00069(8)	0.00222(8)	0.00093(8)
Ni1	0.01181(15)	0.00734(15)	0.00796(14)	-0.00038(12)	0.00350(12)	-0.00074(12)
P1	0.0113(3)	0.0069(3)	0.0065(3)	-0.0003(2)	0.0032(2)	0.0002(2)
P2	0.0096(3)	0.0062(3)	0.0065(3)	0.0004(2)	0.0019(2)	-0.0005(2)
O1	0.0111(8)	0.0095(8)	0.0120(8)	-0.0008(7)	0.0020(7)	-0.0001(7)
O2	0.0137(9)	0.0107(8)	0.0104(8)	-0.0011(7)	0.0024(7)	-0.0009(7)
O3	0.0152(9)	0.0108(8)	0.0086(8)	-0.0013(7)	0.0049(7)	-0.0026(7)
O4	0.0114(8)	0.0105(8)	0.0093(8)	-0.0011(7)	0.0011(7)	0.0006(7)
O5	0.0161(9)	0.0103(8)	0.0128(8)	0.0001(7)	0.0044(7)	-0.0005(7)
O6	0.0101(8)	0.0075(8)	0.0081(8)	-0.0001(6)	0.0014(6)	-0.0005(6)
O7	0.0128(8)	0.0073(8)	0.0075(8)	-0.0006(6)	0.0037(7)	0.0003(6)
O8	0.0166(9)	0.0103(9)	0.0125(8)	-0.0005(7)	0.0009(7)	0.0022(7)
O9	0.0136(9)	0.0170(9)	0.0139(9)	-0.0032(7)	0.0047(7)	-0.0018(7)
O10	0.0107(8)	0.0094(8)	0.0087(8)	-0.0018(7)	0.0039(7)	0.0004(7)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
O11	0.0180(9)	0.0093(8)	0.0127(8)	0.0008(7)	0.0054(7)	0.0020(7)
O12	0.0106(8)	0.0075(8)	0.0087(8)	0.0001(6)	0.0037(6)	0.0000(6)
O13	0.0144(9)	0.0071(8)	0.0104(8)	0.0007(7)	0.0060(7)	0.0001(7)
O14	0.0126(8)	0.0084(8)	0.0108(8)	-0.0017(7)	0.0047(7)	-0.0003(7)
O15	0.0122(9)	0.0123(9)	0.0144(9)	-0.0022(7)	0.0013(7)	0.0005(7)
O16	0.0188(9)	0.0135(9)	0.0239(10)	-0.0010(8)	0.0132(8)	-0.0013(7)
O17	0.0101(8)	0.0103(8)	0.0097(8)	-0.0007(7)	0.0012(7)	-0.0006(7)
O18	0.0138(8)	0.0074(8)	0.0087(8)	-0.0002(6)	0.0037(7)	0.0007(7)
O19	0.0151(9)	0.0109(9)	0.0153(9)	-0.0025(7)	0.0060(7)	0.0004(7)
O20	0.0153(9)	0.0092(8)	0.0154(9)	0.0009(7)	0.0043(7)	0.0023(7)
O21	0.0122(8)	0.0066(8)	0.0100(8)	0.0005(6)	0.0036(7)	0.0001(6)
O22	0.0257(11)	0.0104(9)	0.0134(9)	-0.0019(8)	0.0036(8)	0.0002(8)
O23	0.0170(9)	0.0145(10)	0.0158(9)	-0.0025(8)	0.0070(8)	-0.0013(8)
O24	0.0148(9)	0.0110(9)	0.0121(9)	0.0027(7)	0.0037(7)	-0.0009(7)
O25	0.0187(10)	0.0166(10)	0.0170(10)	0.0005(8)	0.0095(8)	-0.0038(8)
O90	0.0404(13)	0.0221(11)	0.0216(11)	-0.0038(9)	0.0147(10)	-0.0150(9)
O91	0.0244(10)	0.0131(9)	0.0165(10)	-0.0011(8)	0.0100(8)	0.0067(8)
O92	0.0267(11)	0.0353(13)	0.0257(11)	0.0062(10)	0.0121(9)	0.0027(9)
O93	0.0356(12)	0.0218(11)	0.0426(13)	0.0020(10)	0.0245(11)	-0.0019(10)
N1	0.0109(10)	0.0107(10)	0.0118(10)	0.0005(8)	0.0046(8)	-0.0004(8)
N2	0.0213(12)	0.0079(10)	0.0225(12)	0.0049(9)	0.0114(10)	0.0022(9)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
N3	0.0213(13)	0.0438(17)	0.0353(15)	0.0105(13)	0.0149(11)	0.0056(12)
N4	0.075(2)	0.0272(15)	0.0125(13)	0.0067(11)	-0.0012(14)	-0.0219(16)
C1	0.0132(12)	0.0085(11)	0.0088(11)	0.0007(9)	0.0047(9)	-0.0001(9)
C2	0.0123(12)	0.0103(12)	0.0098(11)	-0.0037(9)	0.0066(9)	-0.0006(9)
C3	0.0200(13)	0.0121(12)	0.0105(12)	0.0010(10)	0.0063(10)	-0.0004(10)
C4	0.0241(14)	0.0127(13)	0.0159(13)	-0.0015(10)	0.0112(11)	-0.0077(11)
C5	0.0169(13)	0.0161(13)	0.0129(12)	-0.0045(10)	0.0046(10)	-0.0068(10)
C6	0.0168(13)	0.0153(13)	0.0102(12)	0.0005(10)	0.0047(10)	-0.0009(10)
C7	0.0135(12)	0.0109(12)	0.0097(11)	-0.0021(9)	0.0073(10)	-0.0013(9)
C8	0.0122(12)	0.0086(11)	0.0082(11)	0.0005(9)	0.0029(9)	-0.0006(9)
C9	0.0145(12)	0.0127(12)	0.0115(12)	-0.0016(10)	0.0040(10)	-0.0019(10)
C10	0.0125(12)	0.0121(12)	0.0140(12)	0.0019(10)	0.0016(10)	-0.0014(10)
C11	0.0139(12)	0.0126(12)	0.0136(12)	0.0027(10)	0.0044(10)	0.0015(10)
C12	0.0141(12)	0.0101(12)	0.0168(13)	-0.0008(10)	0.0015(10)	-0.0027(10)
C13	0.0109(12)	0.0122(12)	0.0150(12)	0.0026(10)	0.0067(10)	-0.0001(10)
C14	0.0143(12)	0.0110(12)	0.0145(12)	0.0023(10)	0.0066(10)	0.0011(10)
C15	0.0140(13)	0.0125(13)	0.0204(13)	0.0020(10)	0.0060(10)	0.0001(10)
C16	0.0148(13)	0.0127(12)	0.0184(13)	0.0016(10)	0.0049(10)	-0.0007(10)
C17	0.0151(13)	0.0175(13)	0.0195(13)	0.0034(11)	0.0066(11)	0.0034(10)
C18	0.0186(13)	0.0119(13)	0.0219(13)	-0.0011(11)	0.0090(11)	-0.0022(10)
C19	0.0209(15)	0.0339(17)	0.0329(17)	-0.0009(14)	0.0132(13)	-0.0035(13)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
C20	0.0318(18)	0.0292(17)	0.052(2)	0.0079(16)	0.0220(16)	0.0060(14)
C21	0.0205(14)	0.0268(16)	0.0269(15)	0.0007(13)	0.0114(12)	-0.0009(12)
C22	0.0274(17)	0.0278(17)	0.051(2)	-0.0035(15)	0.0206(16)	-0.0003(13)
C23	0.0206(14)	0.0263(15)	0.0321(16)	-0.0021(13)	0.0196(13)	-0.0010(12)
C24	0.0257(15)	0.0249(15)	0.0210(14)	-0.0051(12)	0.0174(12)	-0.0086(12)
C25	0.050(2)	0.0239(16)	0.0372(18)	-0.0109(14)	0.0349(17)	-0.0155(14)
C26	0.0273(15)	0.0226(15)	0.0130(13)	-0.0024(11)	0.0102(11)	-0.0089(12)
C27	0.075(3)	0.041(2)	0.0129(15)	-0.0137(14)	0.0223(16)	-0.0357(19)
C28	0.0447(19)	0.0246(16)	0.0189(15)	0.0068(12)	0.0060(14)	-0.0126(14)

Table S63. Hydrogen atomic coordinates and isotropic atomic displacement parameters ($Å^2$) for (H-4,4'bpy)[{Ni(H-4,4'-bpy)$(H_2O)_4$}Mo_5O_{15}(1,2-$O_3PC_8H_8PO_3$)]·$4H_2O$ (**8**·$4H_2O$).

	x/a	y/b	z/c	U(eq)
H22C	0.369(2)	0.3391(19)	0.2743(18)	0.021
H22D	0.371(2)	0.3614(18)	0.3390(19)	0.021
H23C	0.471(2)	0.5178(18)	0.2259(18)	0.019
H23D	0.461(2)	0.4418(19)	0.2312(17)	0.019
H24C	0.463(2)	0.5182(17)	0.3907(17)	0.015
H24D	0.4127(19)	0.4782(18)	0.4117(17)	0.015
H25C	0.210(2)	0.4600(18)	0.3196(19)	0.02
H25D	0.168(2)	0.4842(17)	0.2507(18)	0.02
H90A	0.424(2)	0.5546(14)	0.9515(18)	0.032
H90B	0.388(2)	0.4907(18)	0.9711(15)	0.032
H91A	0.0408(19)	0.5067(13)	0.1412(16)	0.021
H91B	0.074(2)	0.4407(16)	0.1285(14)	0.021
H92A	0.4844(17)	0.2482(19)	0.3110(17)	0.034
H92B	0.447(2)	0.217(2)	0.2371(14)	0.034
H93A	0.546(2)	0.6185(12)	0.1825(19)	0.037
H93B	0.554(2)	0.5486(19)	0.1358(17)	0.037
H2'	0.2129(19)	1.0529(11)	0.1115(16)	0.02
H4'	1.032(2)	0.245(2)	0.7898(16)	0.062(13)
H1A	0.3110	0.1843	0.3259	0.012

	x/a	y/b	z/c	U(eq)
H1B	0.3109	0.1059	0.3747	0.012
H3	0.1809	0.0209	0.3384	0.017
H4	0.0502	-0.0280	0.2433	0.02
H5	-0.0187	0.0414	0.1257	0.019
H6	0.0440	0.1595	0.1045	0.017
H8A	0.2375	0.2573	0.2399	0.012
H8B	0.1397	0.2792	0.1774	0.012
H9	0.2121	0.5900	0.1366	0.016
H10	0.3983	0.6525	0.3382	0.016
H11	0.1747	0.7198	0.1030	0.016
H12	0.3648	0.7846	0.3105	0.018
H15	0.1190	0.8365	0.0991	0.019
H16	0.3683	0.8956	0.2539	0.019
H17	0.0988	0.9663	0.0555	0.021
H18	0.3427	1.0242	0.2078	0.02
H19	0.5957	0.1542	0.4137	0.034
H20	0.5996	0.3843	0.4279	0.043
H21	0.7229	0.1441	0.5274	0.029
H22A	0.7274	0.3815	0.5405	0.04
H25A	0.8160	0.3648	0.6656	0.038
H26	0.8638	0.1480	0.5955	0.024

	x/a	y/b	z/c	U(eq)
H27	0.9502	0.3525	0.7739	0.049
H28	0.9978	0.1427	0.7038	0.037

Table S64. Sample and crystal data for [{Ni(tpypyz)(H_2O)}$_2$$Mo_6O_{18}$($H_2O$)(1,4-$O_3PC_8H_8PO_3$]· 6$H_2O$ (**9**·6H_2O).

Identification code	[{Ni(tpypyz)(H_2O)}$_2$$Mo_6O_{18}$($H_2O$) (1,4-$O_3PC_8H_8PO_3$] · 6$H_2O$	
Chemical formula	$C_{56}H_{58}Mo_6N_{12}Ni_2O_{33}P_2$	
Formula weight	2182.14	
Temperature	90(2) K	
Wavelength	0.71073 Å	
Crystal size	0.040 x 0.060 x 0.080 mm	
Crystal system	Monoclinic	
Space group	P21/n	
Unit cell dimensions	a = 14.3824(13) Å	α = 90°
	b = 23.955(2) Å	β = 100.325(2)°
	c = 20.6777(19) Å	γ = 90°
Volume	7008.7(11) Å^3	
Z	4	
Density (calculated)	2.068 g/cm^3	
Absorption coefficient	1.709 mm^{-1}	
F(000)	4320	

Table S65. Data collection and structure refinement for [{Ni(tpypyz)(H_2O)}$_2$$Mo_6O_{18}$($H_2O$)(1,4-$O_3PC_8H_8PO_3$]·6$H_2O$ (**9**·6H_2O).

Theta range for data collection	1.31 to 26.37°	
Index ranges	-17<=h<=17, -29<=k<=29, -25<=l<=25	
Reflections collected	78703	
Independent reflections	14313 [R(int) = 0.1103]	
Max. and min. transmission	0.9350 and 0.8750	
Refinement method	Full-matrix least-squares on F^2	
Refinement program	SHELXL-2013 (Sheldrick, 2013)	
Function minimized	$\Sigma\, w(F_o^2 - F_c^2)^2$	
Data / restraints / parameters	14313 / 379 / 1048	
Goodness-of-fit on F^2	0.943	
Δ/σ_{max}	1.820	
Final R indices	9651 data; I>2σ(I)	R1 = 0.0487, wR2 = 0.0981
	all data	R1 = 0.0909, wR2 = 0.1154
Weighting scheme	$w=1/[\sigma^2(F_o^2)+(0.0580P)^2]$ where $P=(F_o^2+2F_c^2)/3$	
Largest diff. peak and hole	1.348 and -0.805 eÅ^{-3}	
R.M.S. deviation from mean	0.165 eÅ^{-3}	

Table S66. Atomic coordinates and equivalent isotropic atomic displacement parameters ($Å^2$) for $[\{Ni(tpypyz)(H_2O)\}_2Mo_6O_{18}(H_2O)(1,4\text{-}O_3PC_8H_8PO_3]\cdot 6H_2O$ (**9**·$6H_2O$).

U(eq) is defined as one third of the trace of the orthogonalized U_{ij} tensor.

	x/a	**y/b**	**z/c**	**U(eq)**
Mo1	0.54943(4)	0.74094(2)	0.08652(3)	0.00833(12)
Mo2	0.59482(4)	0.87294(2)	0.18266(3)	0.01210(13)
Mo3	0.43381(4)	0.87303(2)	0.27387(3)	0.01289(13)
Mo4	0.32262(4)	0.74131(2)	0.29955(3)	0.00857(12)
Mo5	0.44391(3)	0.62244(2)	0.29172(3)	0.00856(12)
Mo6	0.55699(3)	0.62080(2)	0.16426(3)	0.00810(12)
Ni1	0.74573(5)	0.69167(4)	0.01304(4)	0.01115(18)
Ni2	0.46615(5)	0.87275(3)	0.82840(4)	0.00928(17)
P1	0.35190(10)	0.69866(7)	0.14372(8)	0.0064(3)
P2	0.05924(11)	0.75579(7)	0.75943(8)	0.0082(3)
O1	0.8318(3)	0.6544(2)	0.9563(2)	0.0178(11)
O2	0.8073(3)	0.64462(19)	0.0901(2)	0.0127(10)
O3	0.6654(3)	0.72560(18)	0.0778(2)	0.0112(9)
O4	0.4979(3)	0.75556(19)	0.0081(2)	0.0139(10)
O5	0.5168(3)	0.66213(18)	0.0867(2)	0.0115(9)
O6	0.4104(3)	0.74482(18)	0.1188(2)	0.0098(9)
O7	0.5699(3)	0.81284(18)	0.1216(2)	0.0124(9)
O8	0.7084(3)	0.8639(2)	0.2198(2)	0.0221(11)

	x/a	y/b	z/c	U(eq)
O9	0.5987(3)	0.92592(19)	0.1290(2)	0.0227(11)
O10	0.5370(3)	0.91151(18)	0.2451(2)	0.0173(10)
O11	0.3502(3)	0.92506(19)	0.2615(2)	0.0207(11)
O12	0.4095(3)	0.85853(19)	0.1563(2)	0.0177(10)
O13	0.4701(3)	0.8723(2)	0.3562(2)	0.0246(11)
O14	0.3520(3)	0.81139(18)	0.2641(2)	0.0128(9)
O15	0.3709(3)	0.75500(18)	0.3792(2)	0.0120(9)
O16	0.2033(3)	0.74779(18)	0.2978(2)	0.0127(10)
O17	0.4939(3)	0.6156(2)	0.3723(2)	0.0165(10)
O18	0.3313(3)	0.66177(18)	0.2998(2)	0.0120(9)
O19	0.3908(3)	0.55869(18)	0.2714(2)	0.0146(10)
O20	0.5577(3)	0.61451(17)	0.2567(2)	0.0098(9)
O21	0.2903(3)	0.72345(18)	0.1896(2)	0.0102(9)
O22	0.4108(3)	0.64998(18)	0.1808(2)	0.0104(9)
O23	0.5175(3)	0.55659(18)	0.1356(2)	0.0125(9)
O24	0.6775(3)	0.61823(18)	0.1669(2)	0.0122(9)
O25	0.0396(3)	0.69341(18)	0.7490(2)	0.0113(9)
O26	0.9771(3)	0.78334(17)	0.7860(2)	0.0093(9)
O27	0.0790(3)	0.78348(17)	0.6961(2)	0.0079(9)
O90	0.2445(6)	0.0065(4)	0.1622(5)	0.099(3)
O91	0.2678(5)	0.4083(3)	0.1414(4)	0.071(3)

	x/a	y/b	z/c	U(eq)
O92	0.3413(4)	0.5117(2)	0.1456(3)	0.0305(13)
O93	0.0999(5)	0.7383(3)	0.1576(3)	0.0538(18)
O94	0.1586(5)	0.8476(3)	0.1958(4)	0.070(2)
O95	0.2422(5)	0.9027(3)	0.1063(3)	0.0536(19)
N1	0.6364(4)	0.6402(2)	0.9700(3)	0.0124(11)
N2	0.6727(3)	0.7423(2)	0.9452(2)	0.0074(10)
N3	0.8316(3)	0.7621(2)	0.0236(2)	0.0106(11)
N4	0.5586(3)	0.9240(2)	0.8913(3)	0.0115(11)
N5	0.5571(3)	0.8162(2)	0.8764(2)	0.0076(10)
N6	0.4144(3)	0.7962(2)	0.7841(3)	0.0098(11)
N7	0.5328(3)	0.8945(2)	0.7501(3)	0.0115(11)
N8	0.3748(3)	0.9282(2)	0.7805(3)	0.0096(11)
N9	0.3656(3)	0.8785(2)	0.8891(2)	0.0114(11)
N10	0.3484(4)	0.9672(2)	0.5775(3)	0.0153(12)
N11	0.2507(3)	0.0027(2)	0.7134(2)	0.0111(11)
N12	0.1778(4)	0.0236(2)	0.8641(3)	0.0160(12)
C1	0.6246(5)	0.5878(3)	0.9860(3)	0.0177(14)
C2	0.5479(4)	0.5556(3)	0.9573(3)	0.0163(14)
C3	0.4774(5)	0.5809(3)	0.9127(3)	0.0159(14)
C4	0.4889(4)	0.6359(3)	0.8957(3)	0.0133(13)
C5	0.5705(4)	0.6646(3)	0.9226(3)	0.0095(12)

	x/a	y/b	z/c	U(eq)
C6	0.5927(4)	0.7232(3)	0.9079(3)	0.0084(12)
C7	0.5426(4)	0.7620(3)	0.8636(3)	0.0074(12)
C8	0.6970(4)	0.7961(3)	0.9499(3)	0.0101(12)
C9	0.6287(4)	0.8358(3)	0.9220(3)	0.0091(12)
C10	0.7954(4)	0.8046(3)	0.9847(3)	0.0110(13)
C11	0.9219(4)	0.7643(3)	0.0525(3)	0.0143(14)
C12	0.9803(5)	0.8088(3)	0.0442(3)	0.0194(15)
C13	0.9443(4)	0.8514(3)	0.0032(3)	0.0158(14)
C14	0.8498(4)	0.8501(3)	0.9726(3)	0.0129(13)
C15	0.6228(4)	0.8956(3)	0.9366(3)	0.0119(13)
C16	0.6686(4)	0.9212(3)	0.9927(3)	0.0115(13)
C17	0.6560(4)	0.9783(3)	0.0013(3)	0.0166(14)
C18	0.5963(4)	0.0075(3)	0.9537(3)	0.0172(14)
C19	0.5475(4)	0.9783(3)	0.8997(3)	0.0154(14)
C20	0.4709(4)	0.7521(3)	0.8027(3)	0.0083(12)
C21	0.4668(4)	0.7036(3)	0.7658(3)	0.0116(13)
C22	0.3963(4)	0.6986(3)	0.7112(3)	0.0140(14)
C23	0.3349(4)	0.7430(3)	0.6948(3)	0.0129(14)
C24	0.3469(4)	0.7907(3)	0.7312(3)	0.0127(13)
C25	0.6212(4)	0.8808(3)	0.7450(3)	0.0134(14)
C26	0.6718(4)	0.9089(3)	0.7053(3)	0.0164(15)

	x/a	y/b	z/c	U(eq)
C27	0.6312(4)	0.9544(3)	0.6701(3)	0.0155(14)
C28	0.5373(4)	0.9690(3)	0.6737(3)	0.0135(14)
C29	0.4902(4)	0.9375(3)	0.7139(3)	0.0108(13)
C30	0.3940(4)	0.9509(3)	0.7256(3)	0.0117(13)
C31	0.3250(4)	0.9853(3)	0.6880(3)	0.0119(13)
C32	0.2965(4)	0.9411(3)	0.8052(3)	0.0097(12)
C33	0.2382(4)	0.9838(3)	0.7726(3)	0.0109(13)
C34	0.3667(4)	0.8492(3)	0.9439(3)	0.0141(14)
C35	0.2912(5)	0.8466(3)	0.9763(3)	0.0173(15)
C36	0.2099(4)	0.8746(3)	0.9489(3)	0.0185(15)
C37	0.2073(4)	0.9053(3)	0.8925(3)	0.0135(14)
C38	0.2870(4)	0.9086(3)	0.8637(3)	0.0106(13)
C39	0.3341(4)	0.0057(3)	0.6219(3)	0.0123(13)
C40	0.3372(5)	0.0623(3)	0.6113(3)	0.0187(15)
C41	0.3629(5)	0.0811(3)	0.5532(4)	0.0278(17)
C42	0.3793(5)	0.0420(3)	0.5074(4)	0.0272(17)
C43	0.3683(5)	0.9860(3)	0.5201(3)	0.0227(16)
C44	0.1610(4)	0.0125(3)	0.7989(3)	0.0112(13)
C45	0.0782(4)	0.0272(3)	0.7583(3)	0.0116(13)
C46	0.1092(4)	0.0485(3)	0.8884(3)	0.0194(15)
C47	0.0241(5)	0.0646(3)	0.8509(4)	0.0207(16)

	x/a	y/b	z/c	U(eq)
C48	0.0076(5)	0.0531(3)	0.7842(3)	0.0174(15)
C49	0.2738(4)	0.6654(3)	0.0763(3)	0.0129(13)
C50	0.2450(4)	0.6957(3)	0.0122(3)	0.0133(13)
C51	0.3034(4)	0.6920(3)	0.9653(3)	0.0127(13)
C52	0.1569(4)	0.7210(3)	0.9934(3)	0.0143(14)
C53	0.2749(4)	0.7144(3)	0.9030(3)	0.0149(14)
C54	0.1271(5)	0.7437(3)	0.9308(3)	0.0169(15)
C55	0.1858(4)	0.7399(3)	0.8845(3)	0.0138(14)
C56	0.1668(4)	0.7663(3)	0.8171(3)	0.0106(13)

Table S67. Bond lengths (Å) for [{Ni(tpypyz)(H_2O)}$_2$$Mo_6O_{18}$($H_2O$)(1,4-$O_3PC_8H_8PO_3$]·6$H_2O$ (**9**·6H_2O).

Mo1-O4	1.695(4)	Mo1-O3	1.748(4)
Mo1-O7	1.872(4)	Mo1-O5	1.946(4)
Mo1-O6	2.221(4)	Mo1-O27	2.305(4)
Mo2-O8	1.690(5)	Mo2-O9	1.693(5)
Mo2-O10	1.897(4)	Mo2-O7	1.906(4)
Mo2-O25	2.330(4)	Mo3-O13	1.688(5)
Mo3-O11	1.719(5)	Mo3-O14	1.876(4)
Mo3-O10	1.930(4)	Mo3-O25	2.323(4)
Mo3-O12	2.417(4)	Mo4-O15	1.701(4)
Mo4-O16	1.717(4)	Mo4-O14	1.910(4)
Mo4-O18	1.910(4)	Mo4-O21	2.277(4)
Mo4-O26	2.364(4)	Mo5-O17	1.700(4)
Mo5-O19	1.725(4)	Mo5-O18	1.906(4)
Mo5-O20	1.915(4)	Mo5-O26	2.315(4)
Mo5-O22	2.352(4)	Mo6-O23	1.709(4)
Mo6-O24	1.725(4)	Mo6-O5	1.884(4)
Mo6-O20	1.915(4)	Mo6-O22	2.298(4)
Mo6-O27	2.391(4)	Ni1-N2	2.003(5)
Ni1-O2	2.022(5)	Ni1-O1	2.055(4)
Ni1-N1	2.070(5)	Ni1-N3	2.080(5)

Ni1-O3	2.085(4)	Ni2-N8	2.001(5)
Ni2-N5	2.017(5)	Ni2-N9	2.083(5)
Ni2-N4	2.085(5)	Ni2-N7	2.090(5)
Ni2-N6	2.124(5)	P1-O21	1.530(4)
P1-O6	1.533(4)	P1-O22	1.560(4)
P1-C49	1.810(6)	P2-O25	1.529(5)
P2-O26	1.538(4)	P2-O27	1.539(4)
P2-C56	1.793(6)	O1-H1C	0.84(2)
O1-H1D	0.85(2)	O2-H2C	0.85(2)
O2-H2D	0.84(2)	O25-Mo3	2.323(4)
O25-Mo2	2.330(4)	O26-Mo5	2.314(4)
O26-Mo4	2.364(4)	O27-Mo1	2.305(4)
O27-Mo6	2.391(4)	O90-H90A	0.93(4)
O90-H90B	0.85(2)	O91-H91B	0.84(2)
O91-H91A	0.84(2)	O92-H92A	0.82(2)
O92-H92B	0.84(2)	O93-H93A	0.86(2)
O93-H93B	0.89(2)	O94-H94A	0.88(2)
O94-H94B	0.85(2)	O95-H95A	0.86(2)
O95-H95B	0.84(2)	N1-C1	1.317(8)
N1-C5	1.366(8)	N2-C8	1.333(8)
N2-C6	1.345(8)	N3-C11	1.329(8)
N3-C10	1.343(8)	N4-C19	1.327(8)

N4-C15	1.371(8)	N5-C7	1.333(8)
N5-C9	1.350(8)	N6-C24	1.333(8)
N6-C20	1.348(8)	N7-C25	1.336(7)
N7-C29	1.354(8)	N8-C30	1.331(8)
N8-C32	1.354(7)	N9-C34	1.330(8)
N9-C38	1.365(8)	N10-C39	1.343(8)
N10-C43	1.348(8)	N11-C31	1.339(7)
N11-C33	1.348(8)	N12-C46	1.328(8)
N12-C44	1.351(8)	C1-C2	1.389(9)
C1-H1	0.95	C2-C3	1.383(9)
C2-H2	0.95	C3-C4	1.382(9)
C3-H3	0.95	C4-C5	1.387(9)
C4-H4	0.95	C5-C6	1.485(8)
C6-C7	1.409(9)	C7-C20	1.496(9)
C8-C9	1.415(9)	C8-C10	1.483(8)
C9-C15	1.468(9)	C10-C14	1.391(9)
C11-C12	1.387(9)	C11-H11	0.95
C12-C13	1.369(10)	C12-H12	0.95
C13-C14	1.392(9)	C13-H13	0.95
C14-H14	0.95	C15-C16	1.372(9)
C16-C17	1.396(9)	C16-H16	0.95
C17-C18	1.377(10)	C17-H17	0.95

C18-C19	1.395(9)	C18-H18	0.95
C19-H19	0.95	C20-C21	1.385(9)
C21-C22	1.381(9)	C21-H21	0.95
C22-C23	1.384(9)	C22-H22	0.95
C23-C24	1.363(9)	C23-H23	0.95
C24-H24	0.95	C25-C26	1.367(8)
C25-H25	0.95	C26-C27	1.382(9)
C26-H26	0.95	C27-C28	1.410(8)
C27-H27	0.95	C28-C29	1.386(8)
C28-H28	0.95	C29-C30	1.481(8)
C30-C31	1.411(9)	C31-C39	1.480(8)
C32-C33	1.415(9)	C32-C38	1.465(9)
C33-C44	1.490(8)	C34-C35	1.374(8)
C34-H34	0.95	C35-C36	1.378(9)
C35-H35	0.95	C36-C37	1.374(9)
C36-H36	0.95	C37-C38	1.385(8)
C37-H37	0.95	C39-C40	1.374(9)
C40-C41	1.395(9)	C40-H40	0.95
C41-C42	1.381(11)	C41-H41	0.95
C42-C43	1.381(11)	C42-H42	0.95
C43-H43	0.95	C44-C45	1.374(9)
C45-C48	1.379(8)	C45-H45	0.95

C46-C47	1.381(9)	C46-H46	0.95
C47-C48	1.385(10)	C47-H47	0.95
C48-H48	0.95	C49-C50	1.503(9)
C49-H49A	0.99	C49-H49B	0.99
C50-C52	1.395(9)	C50-C51	1.395(8)
C51-C53	1.387(9)	C51-H51	0.95
C52-C54	1.398(9)	C52-H52	0.95
C53-C55	1.409(9)	C53-H53	0.95
C54-C55	1.390(8)	C54-H54	0.95
C55-C56	1.509(9)	C56-H56A	0.99
C56-H56B	0.99		

Table S68. Bond angles (°) for [{Ni(tpypyz)(H_2O)}$_2$$Mo_6O_{18}$($H_2O$)(1,4-$O_3PC_8H_8PO_3$]·6$H_2O$ (**9**·6H_2O).

O4-Mo1-O3	101.93(19)	O4-Mo1-O7	101.1(2)
O3-Mo1-O7	98.45(19)	O4-Mo1-O5	98.0(2)
O3-Mo1-O5	91.80(18)	O7-Mo1-O5	155.95(17)
O4-Mo1-O6	90.95(17)	O3-Mo1-O6	165.17(18)
O7-Mo1-O6	86.10(16)	O5-Mo1-O6	79.00(16)
O4-Mo1-O27	164.90(17)	O3-Mo1-O27	92.08(17)
O7-Mo1-O27	82.18(16)	O5-Mo1-O27	75.73(16)
O6-Mo1-O27	74.48(14)	O8-Mo2-O9	104.6(2)
O8-Mo2-O10	105.0(2)	O9-Mo2-O10	98.7(2)
O8-Mo2-O7	104.8(2)	O9-Mo2-O7	99.0(2)
O10-Mo2-O7	139.94(18)	O8-Mo2-O25	92.75(19)
O9-Mo2-O25	162.1(2)	O10-Mo2-O25	72.19(17)
O7-Mo2-O25	80.27(16)	O13-Mo3-O11	103.8(2)
O13-Mo3-O14	100.1(2)	O11-Mo3-O14	98.4(2)
O13-Mo3-O10	101.5(2)	O11-Mo3-O10	99.5(2)
O14-Mo3-O10	147.65(18)	O13-Mo3-O25	96.9(2)
O11-Mo3-O25	158.89(19)	O14-Mo3-O25	81.94(16)
O10-Mo3-O25	71.84(16)	O13-Mo3-O12	167.1(2)
O11-Mo3-O12	89.07(19)	O14-Mo3-O12	78.65(17)
O10-Mo3-O12	74.91(18)	O25-Mo3-O12	70.23(15)

O15-Mo4-O16	103.6(2)	O15-Mo4-O14	97.1(2)
O16-Mo4-O14	101.64(19)	O15-Mo4-O18	100.1(2)
O16-Mo4-O18	98.92(19)	O14-Mo4-O18	149.24(17)
O15-Mo4-O21	167.88(17)	O16-Mo4-O21	88.38(17)
O14-Mo4-O21	78.54(17)	O18-Mo4-O21	79.44(17)
O15-Mo4-O26	86.22(17)	O16-Mo4-O26	167.77(18)
O14-Mo4-O26	84.06(15)	O18-Mo4-O26	71.85(15)
O21-Mo4-O26	82.12(14)	O17-Mo5-O19	104.1(2)
O17-Mo5-O18	100.29(19)	O19-Mo5-O18	96.49(19)
O17-Mo5-O20	96.97(19)	O19-Mo5-O20	101.02(18)
O18-Mo5-O20	151.57(18)	O17-Mo5-O26	95.15(19)
O19-Mo5-O26	159.57(19)	O18-Mo5-O26	73.07(16)
O20-Mo5-O26	83.06(16)	O17-Mo5-O22	163.54(19)
O19-Mo5-O22	90.42(18)	O18-Mo5-O22	85.43(16)
O20-Mo5-O22	72.30(16)	O26-Mo5-O22	71.59(15)
O23-Mo6-O24	104.4(2)	O23-Mo6-O5	98.4(2)
O24-Mo6-O5	101.49(18)	O23-Mo6-O20	102.57(19)
O24-Mo6-O20	98.03(18)	O5-Mo6-O20	146.72(17)
O23-Mo6-O22	94.00(17)	O24-Mo6-O22	161.18(18)
O5-Mo6-O22	79.47(16)	O20-Mo6-O22	73.57(16)
O23-Mo6-O27	168.04(17)	O24-Mo6-O27	86.72(17)
O5-Mo6-O27	74.68(17)	O20-Mo6-O27	79.84(16)

O22-Mo6-O27	75.33(14)	N2-Ni1-O2	171.85(18)
N2-Ni1-O1	99.00(19)	O2-Ni1-O1	89.13(18)
N2-Ni1-N1	78.3(2)	O2-Ni1-N1	101.5(2)
O1-Ni1-N1	89.1(2)	N2-Ni1-N3	79.0(2)
O2-Ni1-N3	101.6(2)	O1-Ni1-N3	90.30(19)
N1-Ni1-N3	156.9(2)	N2-Ni1-O3	86.16(18)
O2-Ni1-O3	85.71(17)	O1-Ni1-O3	174.81(19)
N1-Ni1-O3	92.48(18)	N3-Ni1-O3	90.18(18)
N8-Ni2-N5	179.3(2)	N8-Ni2-N9	77.9(2)
N5-Ni2-N9	101.91(19)	N8-Ni2-N4	101.9(2)
N5-Ni2-N4	78.7(2)	N9-Ni2-N4	91.11(19)
N8-Ni2-N7	78.6(2)	N5-Ni2-N7	101.65(19)
N9-Ni2-N7	156.2(2)	N4-Ni2-N7	90.4(2)
N8-Ni2-N6	102.2(2)	N5-Ni2-N6	77.2(2)
N9-Ni2-N6	95.10(19)	N4-Ni2-N6	155.8(2)
N7-Ni2-N6	93.19(19)	O21-P1-O6	110.0(2)
O21-P1-O22	108.0(2)	O6-P1-O22	114.9(2)
O21-P1-C49	107.5(3)	O6-P1-C49	111.1(3)
O22-P1-C49	104.9(3)	O25-P2-O26	109.6(2)
O25-P2-O27	111.0(2)	O26-P2-O27	112.9(2)
O25-P2-C56	110.2(3)	O26-P2-C56	109.2(3)
O27-P2-C56	103.7(3)	Ni1-O1-H1C	114.(5)

Ni1-O1-H1D	115.(5)	H1C-O1-H1D	109.(4)
Ni1-O2-H2C	119.(5)	Ni1-O2-H2D	110.(4)
H2C-O2-H2D	101.(4)	Mo1-O3-Ni1	142.8(2)
Mo6-O5-Mo1	118.3(2)	P1-O6-Mo1	130.4(2)
Mo1-O7-Mo2	161.8(2)	Mo2-O10-Mo3	115.6(2)
Mo3-O14-Mo4	145.9(3)	Mo5-O18-Mo4	123.3(2)
Mo5-O20-Mo6	121.2(2)	P1-O21-Mo4	130.9(2)
P1-O22-Mo6	125.8(2)	P1-O22-Mo5	133.3(2)
Mo6-O22-Mo5	91.67(15)	P2-O25-Mo3	138.6(2)
P2-O25-Mo2	132.5(2)	Mo3-O25-Mo2	88.21(15)
P2-O26-Mo5	128.0(2)	P2-O26-Mo4	137.2(2)
Mo5-O26-Mo4	91.76(14)	P2-O27-Mo1	134.2(2)
P2-O27-Mo6	127.8(2)	Mo1-O27-Mo6	88.89(14)
H90A-O90-H90B	103.(5)	H91B-O91-H91A	107.(5)
H92A-O92-H92B	113.(5)	H93A-O93-H93B	102.(4)
H94A-O94-H94B	103.(5)	H95A-O95-H95B	107.(5)
C1-N1-C5	118.9(6)	C1-N1-Ni1	125.6(5)
C5-N1-Ni1	115.5(4)	C8-N2-C6	123.7(5)
C8-N2-Ni1	115.8(4)	C6-N2-Ni1	118.9(4)
C11-N3-C10	118.8(6)	C11-N3-Ni1	126.4(5)
C10-N3-Ni1	113.1(4)	C19-N4-C15	118.7(6)
C19-N4-Ni2	125.5(4)	C15-N4-Ni2	114.2(4)

C7-N5-C9	123.5(5)	C7-N5-Ni2	119.4(4)
C9-N5-Ni2	117.1(4)	C24-N6-C20	118.1(6)
C24-N6-Ni2	125.9(4)	C20-N6-Ni2	114.0(4)
C25-N7-C29	119.2(5)	C25-N7-Ni2	125.1(4)
C29-N7-Ni2	113.2(4)	C30-N8-C32	122.8(5)
C30-N8-Ni2	118.4(4)	C32-N8-Ni2	118.8(4)
C34-N9-C38	119.3(5)	C34-N9-Ni2	125.1(4)
C38-N9-Ni2	114.8(4)	C39-N10-C43	117.1(6)
C31-N11-C33	119.8(5)	C46-N12-C44	117.0(6)
N1-C1-C2	123.4(7)	N1-C1-H1	118.3
C2-C1-H1	118.3	C3-C2-C1	118.3(7)
C3-C2-H2	120.9	C1-C2-H2	120.9
C4-C3-C2	118.7(6)	C4-C3-H3	120.7
C2-C3-H3	120.7	C3-C4-C5	120.1(6)
C3-C4-H4	119.9	C5-C4-H4	119.9
N1-C5-C4	120.3(6)	N1-C5-C6	113.8(5)
C4-C5-C6	125.8(6)	N2-C6-C7	115.8(6)
N2-C6-C5	113.4(5)	C7-C6-C5	130.8(6)
N5-C7-C6	117.9(6)	N5-C7-C20	112.5(5)
C6-C7-C20	129.5(6)	N2-C8-C9	117.9(6)
N2-C8-C10	112.5(5)	C9-C8-C10	129.6(6)
N5-C9-C8	115.8(6)	N5-C9-C15	114.8(5)

C8-C9-C15	129.4(6)	N3-C10-C14	122.1(6)
N3-C10-C8	114.8(5)	C14-C10-C8	122.7(6)
N3-C11-C12	122.7(7)	N3-C11-H11	118.7
C12-C11-H11	118.7	C13-C12-C11	118.6(6)
C13-C12-H12	120.7	C11-C12-H12	120.7
C12-C13-C14	119.6(6)	C12-C13-H13	120.2
C14-C13-H13	120.2	C10-C14-C13	118.1(6)
C10-C14-H14	121.0	C13-C14-H14	121.0
N4-C15-C16	121.2(6)	N4-C15-C9	113.6(6)
C16-C15-C9	124.9(6)	C15-C16-C17	119.4(6)
C15-C16-H16	120.3	C17-C16-H16	120.3
C18-C17-C16	119.1(6)	C18-C17-H17	120.5
C16-C17-H17	120.5	C17-C18-C19	118.5(6)
C17-C18-H18	120.7	C19-C18-H18	120.7
N4-C19-C18	122.8(6)	N4-C19-H19	118.6
C18-C19-H19	118.6	N6-C20-C21	122.4(6)
N6-C20-C7	113.9(5)	C21-C20-C7	123.5(6)
C22-C21-C20	118.4(6)	C22-C21-H21	120.8
C20-C21-H21	120.8	C21-C22-C23	118.5(6)
C21-C22-H22	120.7	C23-C22-H22	120.7
C24-C23-C22	119.8(6)	C24-C23-H23	120.1
C22-C23-H23	120.1	N6-C24-C23	122.5(6)

N6-C24-H24	118.8	C23-C24-H24	118.8
N7-C25-C26	122.8(6)	N7-C25-H25	118.6
C26-C25-H25	118.6	C25-C26-C27	119.1(6)
C25-C26-H26	120.5	C27-C26-H26	120.5
C26-C27-C28	119.1(6)	C26-C27-H27	120.5
C28-C27-H27	120.5	C29-C28-C27	118.2(6)
C29-C28-H28	120.9	C27-C28-H28	120.9
N7-C29-C28	121.6(5)	N7-C29-C30	114.7(5)
C28-C29-C30	123.5(6)	N8-C30-C31	118.4(5)
N8-C30-C29	113.2(5)	C31-C30-C29	128.4(6)
N11-C31-C30	119.9(5)	N11-C31-C39	117.9(6)
C30-C31-C39	122.2(5)	N8-C32-C33	116.6(5)
N8-C32-C38	113.4(5)	C33-C32-C38	130.0(5)
N11-C33-C32	120.8(5)	N11-C33-C44	114.0(5)
C32-C33-C44	125.2(5)	N9-C34-C35	123.3(6)
N9-C34-H34	118.4	C35-C34-H34	118.4
C34-C35-C36	117.7(6)	C34-C35-H35	121.2
C36-C35-H35	121.2	C37-C36-C35	120.1(6)
C37-C36-H36	120.0	C35-C36-H36	120.0
C36-C37-C38	119.8(6)	C36-C37-H37	120.1
C38-C37-H37	120.1	N9-C38-C37	119.7(6)
N9-C38-C32	114.0(5)	C37-C38-C32	126.3(6)

N10-C39-C40	123.7(6)	N10-C39-C31	117.0(6)
C40-C39-C31	118.9(6)	C39-C40-C41	118.5(7)
C39-C40-H40	120.8	C41-C40-H40	120.8
C42-C41-C40	118.4(7)	C42-C41-H41	120.8
C40-C41-H41	120.8	C43-C42-C41	119.2(7)
C43-C42-H42	120.4	C41-C42-H42	120.4
N10-C43-C42	122.8(7)	N10-C43-H43	118.6
C42-C43-H43	118.6	N12-C44-C45	122.8(6)
N12-C44-C33	116.3(5)	C45-C44-C33	121.0(6)
C44-C45-C48	119.7(6)	C44-C45-H45	120.2
C48-C45-H45	120.2	N12-C46-C47	123.8(7)
N12-C46-H46	118.1	C47-C46-H46	118.1
C46-C47-C48	118.8(6)	C46-C47-H47	120.6
C48-C47-H47	120.6	C45-C48-C47	118.0(6)
C45-C48-H48	121.0	C47-C48-H48	121.0
C50-C49-P1	120.2(5)	C50-C49-H49A	107.3
P1-C49-H49A	107.3	C50-C49-H49B	107.3
P1-C49-H49B	107.3	H49A-C49-H49B	106.9
C52-C50-C51	117.7(6)	C52-C50-C49	123.2(6)
C51-C50-C49	118.5(6)	C53-C51-C50	120.4(6)
C53-C51-H51	119.8	C50-C51-H51	119.8
C50-C52-C54	122.6(6)	C50-C52-H52	118.7

C54-C52-H52	118.7	C51-C53-C55	121.6(6)
C51-C53-H53	119.2	C55-C53-H53	119.2
C55-C54-C52	119.3(6)	C55-C54-H54	120.3
C52-C54-H54	120.3	C54-C55-C53	118.4(6)
C54-C55-C56	125.2(6)	C53-C55-C56	116.0(5)
C55-C56-P2	123.2(4)	C55-C56-H56A	106.5
P2-C56-H56A	106.5	C55-C56-H56B	106.5
P2-C56-H56B	106.5	H56A-C56-H56B	106.5

Table S69. Anisotropic atomic displacement parameters ($Å^2$) for $[\{Ni(tpypyz)(H_2O)\}_2Mo_6O_{18}(H_2O)(1,4\text{-}O_3PC_8H_8PO_3]\cdot 6H_2O$ (**9**·$6H_2O$).

The anisotropic atomic displacement factor exponent takes the form: $-2\pi^2[\, h^2\, a^{*2}\, U_{11} + ... + 2\, h\, k\, a^*\, b^*\, U_{12}\,]$

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
Mo1	0.0087(3)	0.0097(3)	0.0067(3)	0.0012(2)	0.0020(2)	0.0018(2)
Mo2	0.0141(3)	0.0082(3)	0.0149(3)	-0.0001(2)	0.0051(2)	0.0010(2)
Mo3	0.0133(3)	0.0112(3)	0.0153(3)	-0.0034(3)	0.0059(2)	-0.0010(2)
Mo4	0.0092(3)	0.0100(3)	0.0070(3)	-0.0008(2)	0.0027(2)	-0.0003(2)
Mo5	0.0083(2)	0.0099(3)	0.0074(3)	0.0002(2)	0.0012(2)	0.0000(2)
Mo6	0.0088(2)	0.0085(3)	0.0070(3)	0.0002(2)	0.0015(2)	0.0008(2)
Ni1	0.0107(4)	0.0129(5)	0.0099(4)	0.0014(4)	0.0018(3)	0.0029(3)
Ni2	0.0093(4)	0.0093(4)	0.0090(4)	0.0017(4)	0.0010(3)	0.0021(3)
P1	0.0043(7)	0.0077(8)	0.0062(8)	-0.0003(7)	-0.0013(6)	0.0005(6)
P2	0.0080(7)	0.0086(9)	0.0079(8)	0.0004(7)	0.0013(6)	0.0002(6)
O1	0.023(3)	0.020(3)	0.014(3)	-0.002(2)	0.011(2)	0.002(2)
O2	0.013(2)	0.014(2)	0.012(2)	0.0026(19)	0.0035(18)	0.0033(18)
O3	0.012(2)	0.014(2)	0.008(2)	0.0002(18)	0.0024(17)	0.0003(17)
O4	0.012(2)	0.019(3)	0.012(2)	0.0045(19)	0.0034(18)	0.0049(19)
O5	0.016(2)	0.013(2)	0.005(2)	0.0007(18)	0.0018(18)	0.0024(18)
O6	0.010(2)	0.014(2)	0.005(2)	0.0021(18)	0.0003(17)	0.0025(17)
O7	0.020(2)	0.011(2)	0.008(2)	0.0022(18)	0.0048(18)	-0.0002(18)
O8	0.016(2)	0.022(3)	0.029(3)	-0.004(2)	0.006(2)	0.000(2)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
O9	0.038(3)	0.010(3)	0.025(3)	0.004(2)	0.016(2)	0.002(2)
O10	0.019(2)	0.009(2)	0.026(3)	-0.005(2)	0.009(2)	-0.0023(18)
O11	0.022(2)	0.012(3)	0.031(3)	-0.004(2)	0.013(2)	0.002(2)
O12	0.029(3)	0.015(3)	0.012(2)	-0.0002(19)	0.010(2)	0.002(2)
O13	0.019(2)	0.035(3)	0.019(3)	-0.006(2)	0.001(2)	-0.003(2)
O14	0.011(2)	0.012(2)	0.018(2)	0.0031(19)	0.0083(18)	0.0024(17)
O15	0.015(2)	0.013(2)	0.008(2)	0.0014(18)	0.0044(18)	-0.0021(18)
O16	0.012(2)	0.014(3)	0.014(2)	-0.0029(19)	0.0053(18)	0.0003(18)
O17	0.013(2)	0.028(3)	0.007(2)	0.002(2)	0.0011(17)	0.004(2)
O18	0.010(2)	0.013(2)	0.013(2)	0.0016(19)	0.0016(18)	0.0000(17)
O19	0.015(2)	0.013(2)	0.017(3)	-0.003(2)	0.0081(19)	-0.0017(18)
O20	0.011(2)	0.011(2)	0.007(2)	-0.0013(18)	0.0011(17)	0.0002(17)
O21	0.013(2)	0.015(2)	0.003(2)	0.0012(18)	0.0006(17)	0.0049(18)
O22	0.011(2)	0.013(2)	0.007(2)	0.0018(18)	0.0012(17)	0.0009(17)
O23	0.012(2)	0.014(2)	0.012(2)	0.0008(19)	0.0011(18)	0.0030(18)
O24	0.013(2)	0.012(3)	0.011(2)	0.0019(19)	0.0003(17)	0.0028(18)
O25	0.013(2)	0.010(2)	0.011(2)	0.0014(19)	0.0047(18)	0.0041(18)
O26	0.010(2)	0.008(2)	0.011(2)	0.0018(18)	0.0023(17)	-0.0007(17)
O27	0.009(2)	0.009(2)	0.005(2)	0.0032(18)	0.0003(16)	0.0027(17)
O90	0.088(6)	0.084(7)	0.142(8)	-0.050(7)	0.070(6)	0.001(6)
O91	0.103(6)	0.071(6)	0.060(5)	-0.045(4)	0.068(5)	-0.057(5)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
O92	0.025(3)	0.040(4)	0.027(3)	-0.014(3)	0.005(3)	-0.006(3)
O93	0.045(4)	0.068(5)	0.048(4)	0.010(4)	0.008(3)	0.009(4)
O94	0.041(4)	0.077(6)	0.087(7)	0.022(5)	-0.005(4)	0.010(4)
O95	0.075(5)	0.055(5)	0.030(4)	-0.004(4)	0.009(3)	0.032(4)
N1	0.014(2)	0.011(3)	0.013(3)	0.000(2)	0.006(2)	0.001(2)
N2	0.005(2)	0.012(3)	0.005(3)	-0.002(2)	-0.0006(18)	0.0029(19)
N3	0.008(2)	0.016(3)	0.007(3)	-0.003(2)	0.000(2)	0.001(2)
N4	0.013(2)	0.011(3)	0.012(3)	0.001(2)	0.005(2)	0.001(2)
N5	0.009(2)	0.012(3)	0.003(3)	0.000(2)	0.0041(19)	0.0000(19)
N6	0.004(2)	0.013(3)	0.013(3)	0.003(2)	0.0042(19)	0.0044(19)
N7	0.012(2)	0.010(3)	0.012(3)	-0.001(2)	0.001(2)	0.001(2)
N8	0.007(2)	0.009(3)	0.012(3)	-0.004(2)	-0.0010(19)	-0.001(2)
N9	0.014(2)	0.012(3)	0.008(3)	-0.005(2)	0.002(2)	0.000(2)
N10	0.018(3)	0.016(3)	0.012(3)	-0.001(2)	0.003(2)	-0.005(2)
N11	0.011(2)	0.011(3)	0.008(3)	-0.004(2)	-0.006(2)	-0.002(2)
N12	0.020(3)	0.013(3)	0.016(3)	-0.004(2)	0.004(2)	-0.003(2)
C1	0.022(3)	0.014(3)	0.018(4)	0.004(3)	0.007(3)	0.005(3)
C2	0.021(3)	0.013(4)	0.017(4)	0.000(3)	0.010(3)	0.002(3)
C3	0.020(3)	0.018(4)	0.010(4)	-0.001(3)	0.005(3)	-0.006(3)
C4	0.019(3)	0.016(3)	0.006(3)	-0.003(3)	0.004(2)	0.000(3)
C5	0.017(3)	0.007(3)	0.005(3)	-0.003(2)	0.002(2)	0.000(2)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
C6	0.014(3)	0.011(3)	0.001(3)	0.000(2)	0.002(2)	0.000(2)
C7	0.008(3)	0.007(3)	0.009(3)	0.000(2)	0.008(2)	-0.002(2)
C8	0.014(3)	0.014(3)	0.004(3)	-0.003(2)	0.005(2)	0.002(2)
C9	0.008(3)	0.014(3)	0.007(3)	0.002(2)	0.004(2)	0.002(2)
C10	0.009(3)	0.012(3)	0.011(3)	-0.004(3)	0.001(2)	-0.002(2)
C11	0.011(3)	0.021(4)	0.011(4)	-0.004(3)	0.002(2)	0.000(3)
C12	0.014(3)	0.032(4)	0.011(4)	-0.012(3)	-0.001(3)	-0.001(3)
C13	0.012(3)	0.018(4)	0.019(4)	-0.013(3)	0.007(3)	-0.003(3)
C14	0.014(3)	0.018(4)	0.007(3)	-0.005(3)	0.003(2)	0.002(3)
C15	0.008(3)	0.010(3)	0.019(4)	0.000(3)	0.005(2)	-0.003(2)
C16	0.008(3)	0.019(3)	0.007(3)	0.000(3)	-0.001(2)	0.003(3)
C17	0.020(3)	0.018(4)	0.013(4)	-0.008(3)	0.005(3)	-0.004(3)
C18	0.023(4)	0.011(4)	0.018(4)	-0.004(3)	0.005(3)	0.000(3)
C19	0.013(3)	0.014(3)	0.019(4)	0.000(3)	0.002(3)	0.004(3)
C20	0.008(3)	0.012(3)	0.006(3)	-0.001(2)	0.002(2)	0.001(2)
C21	0.014(3)	0.008(3)	0.014(3)	0.001(3)	0.004(2)	0.004(2)
C22	0.022(3)	0.016(4)	0.005(3)	-0.004(3)	0.004(3)	-0.001(3)
C23	0.005(3)	0.019(4)	0.012(4)	-0.001(3)	-0.002(2)	-0.001(2)
C24	0.010(3)	0.016(3)	0.012(3)	0.005(3)	0.002(2)	0.003(3)
C25	0.015(3)	0.016(4)	0.009(3)	-0.003(3)	0.002(3)	0.003(3)
C26	0.012(3)	0.025(4)	0.012(4)	-0.002(3)	0.002(3)	0.004(3)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
C27	0.015(3)	0.022(4)	0.011(4)	-0.003(3)	0.004(3)	-0.005(3)
C28	0.012(3)	0.022(4)	0.006(3)	-0.002(3)	0.000(2)	0.002(3)
C29	0.010(3)	0.011(3)	0.010(3)	-0.005(3)	-0.001(2)	-0.002(2)
C30	0.013(3)	0.011(3)	0.011(3)	0.002(3)	0.003(2)	0.004(2)
C31	0.017(3)	0.009(3)	0.009(3)	-0.003(3)	0.000(2)	-0.004(3)
C32	0.009(3)	0.007(3)	0.011(3)	-0.004(2)	-0.003(2)	-0.005(2)
C33	0.012(3)	0.009(3)	0.012(3)	-0.002(3)	0.001(2)	0.000(2)
C34	0.015(3)	0.014(4)	0.011(3)	0.002(3)	0.000(3)	0.005(3)
C35	0.022(3)	0.020(4)	0.010(4)	0.004(3)	0.002(3)	0.000(3)
C36	0.013(3)	0.023(4)	0.021(4)	-0.004(3)	0.008(3)	-0.002(3)
C37	0.013(3)	0.014(4)	0.014(4)	-0.003(3)	0.003(3)	-0.001(3)
C38	0.013(3)	0.011(3)	0.007(3)	-0.004(2)	-0.001(2)	-0.001(2)
C39	0.010(3)	0.019(4)	0.006(3)	0.002(3)	-0.004(2)	0.001(3)
C40	0.022(3)	0.012(3)	0.023(4)	0.004(3)	0.004(3)	0.001(3)
C41	0.031(4)	0.028(4)	0.021(4)	0.011(3)	-0.003(3)	-0.003(3)
C42	0.029(4)	0.033(4)	0.019(4)	0.008(3)	0.001(3)	-0.007(3)
C43	0.024(4)	0.034(4)	0.010(4)	-0.004(3)	0.003(3)	-0.011(3)
C44	0.012(3)	0.006(3)	0.016(3)	0.002(3)	0.003(2)	0.000(2)
C45	0.015(3)	0.003(3)	0.015(3)	0.005(3)	-0.003(2)	0.000(2)
C46	0.019(3)	0.021(4)	0.017(4)	-0.008(3)	-0.001(3)	0.008(3)
C47	0.017(3)	0.016(4)	0.030(4)	-0.006(3)	0.008(3)	0.002(3)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
C48	0.015(3)	0.010(4)	0.025(4)	-0.003(3)	-0.001(3)	-0.002(3)
C49	0.015(3)	0.013(4)	0.010(3)	-0.003(3)	-0.002(2)	-0.002(3)
C50	0.013(3)	0.011(3)	0.014(3)	-0.005(3)	-0.002(2)	-0.003(3)
C51	0.008(3)	0.013(4)	0.016(3)	-0.001(3)	-0.002(2)	0.003(3)
C52	0.014(3)	0.018(4)	0.013(3)	-0.004(3)	0.009(3)	0.004(3)
C53	0.014(3)	0.018(4)	0.012(3)	0.003(3)	0.000(3)	0.005(3)
C54	0.016(3)	0.017(4)	0.018(4)	-0.001(3)	0.002(3)	0.001(3)
C55	0.013(3)	0.015(4)	0.013(3)	-0.006(3)	0.002(2)	-0.002(3)
C56	0.008(3)	0.010(3)	0.013(3)	-0.002(3)	-0.001(2)	-0.001(2)

Table S70. Hydrogen atomic coordinates and isotropic atomic displacement parameters ($Å^2$) for $[\{Ni(tpypyz)(H_2O)\}_2Mo_6O_{18}(H_2O)(1,4\text{-}O_3PC_8H_8PO_3]\cdot 6H_2O$ (**9**·$6H_2O$).

	x/a	y/b	z/c	U(eq)
H1C	0.805(4)	0.630(2)	0.930(3)	0.021
H1D	0.862(4)	0.677(2)	0.936(3)	0.021
H2C	0.827(4)	0.6123(13)	1.083(3)	0.015
H2D	0.768(4)	0.637(2)	1.115(3)	0.015
H90A	0.210(7)	0.021(6)	0.124(3)	0.118
H90B	0.203(6)	0.004(6)	0.187(4)	0.118
H91B	0.253(7)	0.405(5)	0.179(2)	0.086
H91A	0.225(6)	0.428(5)	0.119(4)	0.086
H92A	0.320(5)	0.533(3)	0.115(2)	0.037
H92B	0.346(5)	0.527(3)	0.1824(17)	0.037
H93A	0.086(7)	0.733(4)	0.196(2)	0.065
H93B	0.130(7)	0.771(2)	0.163(4)	0.065
H94A	0.151(7)	0.816(3)	0.174(5)	0.084
H94B	0.211(4)	0.844(4)	0.222(4)	0.084
H95A	0.204(6)	0.883(3)	1.125(5)	0.064
H95B	0.237(7)	0.9360(13)	1.118(5)	0.064
H1	0.6710	0.5712	1.0188	0.021
H2	0.5440	0.5172	0.9682	0.02
H3	0.4221	0.5607	0.8939	0.019

	x/a	y/b	z/c	U(eq)
H4	0.4407	0.6542	0.8657	0.016
H11	0.9473	0.7342	1.0800	0.017
H12	1.0441	0.8097	1.0665	0.023
H13	0.9835	0.8818	0.9956	0.019
H14	0.8232	0.8795	0.9444	0.015
H16	0.7086	0.9002	1.0255	0.014
H17	0.6882	0.9967	1.0395	0.02
H18	0.5885	1.0467	0.9575	0.021
H19	0.5045	0.9982	0.8676	0.018
H21	0.5115	0.6745	0.7777	0.014
H22	0.3900	0.6655	0.6855	0.017
H23	0.2845	0.7401	0.6583	0.015
H24	0.3056	0.8213	0.7183	0.015
H25	0.6502	0.8501	0.7701	0.016
H26	0.7339	0.8972	0.7020	0.02
H27	0.6661	0.9756	0.6439	0.019
H28	0.5072	0.9996	0.6493	0.016
H34	0.4223	0.8290	0.9616	0.017
H35	0.2950	0.8263	1.0161	0.021
H36	0.1557	0.8726	0.9691	0.022
H37	0.1510	0.9241	0.8732	0.016

	x/a	y/b	z/c	U(eq)
H40	0.3221	1.0879	0.6429	0.022
H41	0.3689	1.1199	0.5452	0.033
H42	0.3980	1.0535	0.4677	0.033
H43	0.3751	0.9597	0.4869	0.027
H45	0.0697	1.0194	0.7125	0.014
H46	0.1191	1.0556	0.9343	0.023
H47	-0.0221	1.0833	0.8704	0.025
H48	-0.0507	1.0628	0.7571	0.021
H49A	0.3040	0.6300	1.0667	0.015
H49B	0.2151	0.6554	1.0924	0.015
H51	0.3631	0.6741	0.9761	0.015
H52	0.1155	0.7228	1.0244	0.017
H53	0.3165	0.7125	0.8722	0.018
H54	0.0674	0.7615	0.9201	0.02
H56A	0.2189	0.7545	0.7947	0.013
H56B	0.1736	0.8071	0.8240	0.013

Table S71. Sample and crystal data for $[\{Ni_2(tpypyz)(H_2O)_4\}Mo_4O_{10}(1,4\text{-}O_3PC_8H_8PO_3)_2]\cdot 4H_2O$ (**10**·$4H_2O$).

Identification code	$[\{Ni_2(tpypyz)(H_2O)_4\}Mo_4O_{10}(1,4\text{-}O_3PC_8H_8PO_3)_2]\cdot 4H_2O$	
Chemical formula	$C_{20}H_{24}Mo_2N_3NiO_{15}P_2$	
Formula weight	858.95	
Temperature	90(2) K	
Wavelength	0.71073 Å	
Crystal size	0.125 x 0.183 x 0.192 mm	
Crystal system	Monoclinic	
Space group	C2/c	
Unit cell dimensions	a = 24.135(4) Å	α = 90°
	b = 11.8418(18) Å	β = 112.140(4)°
	c = 20.308(3) Å	γ = 90°
Volume	5376.1(14) $Å^3$	
Z	8	
Density (calculated)	2.122 g/cm^3	
Absorption coefficient	1.813 mm^{-1}	
F(000)	3416	

Table S72. Data collection and structure refinement for [{Ni_2(tpypyz)(H_2O)$_4$}Mo_4O_{10}(1,4-$O_3PC_8H_8PO_3$)$_2$]·4H_2O (**10**·4H_2O).

Theta range for data collection	1.82 to 26.36°	
Index ranges	-30<=h<=29, -13<=k<=14, -25<=l<=25	
Reflections collected	20391	
Independent reflections	5490 [R(int) = 0.1092]	
Coverage of independent reflections	100.0%	
Max. and min. transmission	0.8050 and 0.7220	
Refinement method	Full-matrix least-squares on F^2	
Refinement program	SHELXL-2013 (Sheldrick, 2013)	
Function minimized	$\Sigma\, w(F_o^2 - F_c^2)^2$	
Data / restraints / parameters	5490 / 127 / 416	
Goodness-of-fit on F^2	1.023	
Δ/σ_{max}	0.001	
Final R indices	4573 data; I>2σ(I)	R1 = 0.0428, wR2 = 0.1143
	all data	R1 = 0.0508, wR2 = 0.1189
Weighting scheme	$w=1/[\sigma^2(F_o^2)+(0.0766P)^2]$ where $P=(F_o^2+2F_c^2)/3$	
Largest diff. peak and hole	2.114 and -1.279 eÅ^{-3}	
R.M.S. deviation from mean	0.187 eÅ^{-3}	

Table S73. Atomic coordinates and equivalent isotropic atomic displacement parameters ($Å^2$) for $[\{Ni_2(tpypyz)(H_2O)_4\}Mo_4O_{10}(1,4\text{-}O_3PC_8H_8PO_3)_2]\cdot 4H_2O$ (**10**·$4H_2O$).

U(eq) is defined as one third of the trace of the orthogonalized U_{ij} tensor.

	x/a	**y/b**	**z/c**	**U(eq)**
Mo1	0.29382(2)	0.30454(3)	0.65747(2)	0.01032(12)
Mo2	0.34786(2)	0.09466(3)	0.58809(2)	0.00955(12)
Ni1	0.41055(2)	0.98536(4)	0.34326(3)	0.01109(15)
P1	0.31253(5)	0.00557(9)	0.42012(5)	0.0099(2)
P2	0.30859(4)	0.34317(9)	0.50230(5)	0.0093(2)
O1	0.26518(13)	0.2121(3)	0.70207(15)	0.0140(6)
O2	0.33311(13)	0.3993(3)	0.71987(16)	0.0167(6)
O3	0.35815(12)	0.2127(2)	0.65542(15)	0.0123(6)
O4	0.33139(13)	0.9889(2)	0.63489(15)	0.0135(6)
O5	0.42151(13)	0.0709(3)	0.60311(15)	0.0163(6)
O6	0.31542(12)	0.0109(2)	0.49724(15)	0.0111(6)
O7	0.28484(12)	0.1152(2)	0.38010(15)	0.0127(6)
O8	0.37382(13)	0.9857(3)	0.41763(15)	0.0149(6)
O9	0.49105(14)	0.0209(3)	0.42905(17)	0.0191(7)
O10	0.32710(13)	0.9728(3)	0.26460(15)	0.0136(6)
O11	0.34434(12)	0.2332(2)	0.51646(14)	0.0117(6)
O12	0.24699(12)	0.3314(2)	0.44097(15)	0.0119(6)
O13	0.30331(12)	0.3894(2)	0.56995(15)	0.0117(6)

	x/a	y/b	z/c	U(eq)
O90A	0.5219(4)	0.3310(18)	0.4188(5)	0.034(4)
O90B	0.5260(3)	0.2571(17)	0.4248(4)	0.034(4)
O91	0.44584(17)	0.2140(3)	0.48178(19)	0.0309(8)
N1	0.40703(15)	0.1559(3)	0.30806(18)	0.0124(7)
N2	0.45981(15)	0.9784(3)	0.28027(17)	0.0112(7)
N3	0.43362(16)	0.8129(3)	0.34675(18)	0.0131(7)
C1	0.37146(18)	0.2380(4)	0.3139(2)	0.0141(8)
C2	0.36545(19)	0.3412(4)	0.2785(2)	0.0176(9)
C3	0.39458(19)	0.3568(4)	0.2327(2)	0.0173(9)
C4	0.43100(18)	0.2712(4)	0.2247(2)	0.0142(8)
C5	0.43791(18)	0.1736(4)	0.2646(2)	0.0120(8)
C6	0.47557(18)	0.0773(4)	0.2602(2)	0.0125(8)
C7	0.48297(17)	0.8788(4)	0.2720(2)	0.0114(8)
C8	0.47266(18)	0.7857(4)	0.3149(2)	0.0118(8)
C9	0.50332(19)	0.6843(4)	0.3273(2)	0.0154(9)
C10	0.49036(19)	0.6018(4)	0.3689(2)	0.0162(9)
C11	0.44763(19)	0.6262(4)	0.3969(2)	0.0179(9)
C12	0.42214(19)	0.7322(4)	0.3868(2)	0.0171(9)
C13	0.26080(19)	0.8930(3)	0.3809(2)	0.0138(8)
C14	0.28288(18)	0.7747(4)	0.4072(2)	0.0129(8)
C15	0.27577(19)	0.6896(3)	0.3584(2)	0.0137(8)

	x/a	y/b	z/c	U(eq)
C16	0.31031(18)	0.7487(4)	0.4796(2)	0.0145(8)
C17	0.29733(19)	0.5813(4)	0.3796(2)	0.0143(8)
C18	0.33165(19)	0.6410(4)	0.5008(2)	0.0151(8)
C19	0.32661(18)	0.5555(4)	0.4512(2)	0.0140(8)
C20	0.35330(18)	0.4403(4)	0.4745(2)	0.0130(8)

Table S74. Bond lengths (Å) for [{Ni_2(tpypyz)(H_2O)$_4$}Mo_4O_{10}(1,4-$O_3PC_8H_8PO_3$)$_2$]·4H_2O (**10**·4H_2O).

Mo1-O2	1.691(3)	Mo1-O1	1.723(3)
Mo1-O3	1.909(3)	Mo1-O7	1.999(3)
Mo1-O13	2.128(3)	Mo1-O12	2.464(3)
Mo2-O4	1.707(3)	Mo2-O5	1.710(3)
Mo2-O3	1.905(3)	Mo2-O6	1.978(3)
Mo2-O11	2.173(3)	Mo2-O12	2.310(3)
Ni1-O8	2.019(3)	Ni1-N2	2.050(3)
Ni1-O10	2.051(3)	Ni1-O9	2.106(3)
Ni1-N3	2.111(4)	Ni1-N1	2.133(4)
P1-O8	1.517(3)	P1-O6	1.542(3)
P1-O7	1.544(3)	P1-C13	1.794(4)
P2-O13	1.528(3)	P2-O11	1.528(3)
P2-O12	1.544(3)	P2-C20	1.806(4)
O7-Mo1	1.999(3)	O9-H9C	0.85(2)
O9-H9D	0.84(2)	O10-H10C	0.82(2)
O10-H10D	0.83(2)	O11-Mo2	2.173(3)
O12-Mo2	2.310(3)	O12-Mo1	2.465(3)
O13-Mo1	2.128(3)	O91-H91A	0.85(2)
O91-H91B	0.86(2)	N1-C1	1.332(5)
N1-C5	1.370(5)	N2-C6	1.342(5)

N2-C7	1.342(5)	N3-C12	1.350(5)
N3-C8	1.368(5)	C1-C2	1.398(6)
C1-H1	0.95	C2-C3	1.373(6)
C2-H2	0.95	C3-C4	1.391(6)
C3-H3	0.95	C4-C5	1.385(6)
C4-H4	0.95	C5-C6	1.481(6)
C6-C6	1.389(8)	C7-C7	1.423(8)
C7-C8	1.483(6)	C8-C9	1.383(6)
C9-C10	1.401(6)	C9-H9	0.95
C10-C11	1.382(6)	C10-H10	0.95
C11-C12	1.378(7)	C11-H11	0.95
C12-H12	0.95	C13-C14	1.523(6)
C13-H13A	0.99	C13-H13B	0.99
C14-C15	1.377(6)	C14-C16	1.399(6)
C15-C17	1.390(6)	C15-H15	0.95
C16-C18	1.382(6)	C16-H16	0.95
C17-C19	1.389(6)	C17-H17	0.95
C18-C19	1.401(6)	C18-H18	0.95
C19-C20	1.507(6)	C20-H20A	0.99
C20-H20B	0.99		

Table S75. Bond angles (°) for [{Ni_2(tpypyz)(H_2O)$_4$}Mo_4O_{10}(1,4-$O_3PC_8H_8PO_3$)$_2$]·4H_2O (**10**·4H_2O).

O2-Mo1-O1	104.51(14)	O2-Mo1-O3	99.41(14)
O1-Mo1-O3	98.07(13)	O2-Mo1-O7	98.87(13)
O1-Mo1-O7	89.49(13)	O3-Mo1-O7	157.76(12)
O2-Mo1-O13	96.08(13)	O1-Mo1-O13	158.43(13)
O3-Mo1-O13	84.62(12)	O7-Mo1-O13	80.94(11)
O2-Mo1-O12	168.90(12)	O1-Mo1-O12	83.96(12)
O3-Mo1-O12	71.93(11)	O7-Mo1-O12	88.23(11)
O13-Mo1-O12	76.52(10)	O4-Mo2-O5	102.96(14)
O4-Mo2-O3	97.67(13)	O5-Mo2-O3	98.48(13)
O4-Mo2-O6	93.33(13)	O5-Mo2-O6	95.92(13)
O3-Mo2-O6	159.42(12)	O4-Mo2-O11	165.47(12)
O5-Mo2-O11	91.13(13)	O3-Mo2-O11	83.57(12)
O6-Mo2-O11	81.51(11)	O4-Mo2-O12	90.23(12)
O5-Mo2-O12	166.29(13)	O3-Mo2-O12	75.74(11)
O6-Mo2-O12	86.91(11)	O11-Mo2-O12	75.98(10)
O8-Ni1-N2	171.20(13)	O8-Ni1-O10	90.21(12)
N2-Ni1-O10	98.12(12)	O8-Ni1-O9	84.78(12)
N2-Ni1-O9	87.31(13)	O10-Ni1-O9	171.09(13)
O8-Ni1-N3	99.29(13)	N2-Ni1-N3	76.95(14)
O10-Ni1-N3	97.18(13)	O9-Ni1-N3	90.91(13)

O8-Ni1-N1	106.00(13)	N2-Ni1-N1	77.79(14)
O10-Ni1-N1	84.52(13)	O9-Ni1-N1	89.77(13)
N3-Ni1-N1	154.66(13)	O8-P1-O6	111.62(16)
O8-P1-O7	110.30(16)	O6-P1-O7	110.19(16)
O8-P1-C13	113.21(19)	O6-P1-C13	103.78(18)
O7-P1-C13	107.49(18)	O13-P2-O11	111.63(16)
O13-P2-O12	112.09(16)	O11-P2-O12	112.23(16)
O13-P2-C20	108.35(18)	O11-P2-C20	103.87(17)
O12-P2-C20	108.21(18)	Mo2-O3-Mo1	123.14(15)
P1-O6-Mo2	144.89(18)	P1-O7-Mo1	139.54(18)
P1-O8-Ni1	137.19(18)	Ni1-O9-H9C	119.(4)
Ni1-O9-H9D	123.(4)	H9C-O9-H9D	89.(5)
Ni1-O10-H10C	110.(4)	Ni1-O10-H10D	108.(4)
H10C-O10-H10D	110.(5)	P2-O11-Mo2	130.26(16)
P2-O12-Mo2	137.04(17)	P2-O12-Mo1	133.00(16)
Mo2-O12-Mo1	89.19(9)	P2-O13-Mo1	130.80(17)
H91A-O91-H91B	102.(6)	C1-N1-C5	118.2(4)
C1-N1-Ni1	127.3(3)	C5-N1-Ni1	113.5(3)
C6-N2-C7	123.3(4)	C6-N2-Ni1	116.9(3)
C7-N2-Ni1	118.7(3)	C12-N3-C8	117.0(4)
C12-N3-Ni1	126.4(3)	C8-N3-Ni1	115.6(3)
N1-C1-C2	122.5(4)	N1-C1-H1	118.7

C2-C1-H1	118.7	C3-C2-C1	119.1(4)
C3-C2-H2	120.5	C1-C2-H2	120.5
C2-C3-C4	119.3(4)	C2-C3-H3	120.4
C4-C3-H3	120.4	C5-C4-C3	118.8(4)
C5-C4-H4	120.6	C3-C4-H4	120.6
N1-C5-C4	122.0(4)	N1-C5-C6	114.6(4)
C4-C5-C6	123.3(4)	N2-C6-C6	117.4(2)
N2-C6-C5	113.8(4)	C6-C6-C5	128.8(2)
N2-C7-C7	116.4(2)	N2-C7-C8	114.2(3)
C7-C7-C8	129.3(2)	N3-C8-C9	122.3(4)
N3-C8-C7	113.6(4)	C9-C8-C7	123.8(4)
C8-C9-C10	119.3(4)	C8-C9-H9	120.3
C10-C9-H9	120.3	C11-C10-C9	118.2(4)
C11-C10-H10	120.9	C9-C10-H10	120.9
C12-C11-C10	119.4(4)	C12-C11-H11	120.3
C10-C11-H11	120.3	N3-C12-C11	123.5(4)
N3-C12-H12	118.3	C11-C12-H12	118.3
C14-C13-P1	115.8(3)	C14-C13-H13A	108.3
P1-C13-H13A	108.3	C14-C13-H13B	108.3
P1-C13-H13B	108.3	H13A-C13-H13B	107.4
C15-C14-C16	118.4(4)	C15-C14-C13	119.3(4)
C16-C14-C13	122.3(4)	C14-C15-C17	121.4(4)

C14-C15-H15	119.3	C17-C15-H15	119.3
C18-C16-C14	120.3(4)	C18-C16-H16	119.9
C14-C16-H16	119.9	C19-C17-C15	120.7(4)
C19-C17-H17	119.6	C15-C17-H17	119.6
C16-C18-C19	121.4(4)	C16-C18-H18	119.3
C19-C18-H18	119.3	C17-C19-C18	117.7(4)
C17-C19-C20	121.1(4)	C18-C19-C20	121.1(4)
C19-C20-P2	116.1(3)	C19-C20-H20A	108.3
P2-C20-H20A	108.3	C19-C20-H20B	108.3
P2-C20-H20B	108.3	H20A-C20-H20B	107.4

Table S76. Torsion angles (°) for [{Ni_2(tpypyz)(H_2O)$_4$}Mo_4O_{10}(1,4-$O_3PC_8H_8PO_3$)$_2$]·4H_2O (**10**·4H_2O).

O8-P1-O6-Mo2	63.2(3)	O7-P1-O6-Mo2	-59.7(3)
C13-P1-O6-Mo2	-174.5(3)	O8-P1-O7-Mo1	167.6(2)
O6-P1-O7-Mo1	-68.7(3)	C13-P1-O7-Mo1	43.8(3)
O6-P1-O8-Ni1	-171.4(2)	O7-P1-O8-Ni1	-48.6(3)
C13-P1-O8-Ni1	71.9(3)	O13-P2-O11-Mo2	36.0(3)
O12-P2-O11-Mo2	-90.8(2)	C20-P2-O11-Mo2	152.5(2)
O13-P2-O12-Mo2	20.4(3)	O11-P2-O12-Mo2	147.0(2)
C20-P2-O12-Mo2	-99.0(3)	O13-P2-O12-Mo1	-146.26(19)
O11-P2-O12-Mo1	-19.7(3)	C20-P2-O12-Mo1	94.3(2)
O11-P2-O13-Mo1	-36.0(3)	O12-P2-O13-Mo1	90.9(2)
C20-P2-O13-Mo1	-149.8(2)	C5-N1-C1-C2	-1.1(6)
Ni1-N1-C1-C2	-168.7(3)	N1-C1-C2-C3	3.7(6)
C1-C2-C3-C4	-2.3(6)	C2-C3-C4-C5	-1.6(6)
C1-N1-C5-C4	-3.0(6)	Ni1-N1-C5-C4	166.3(3)
C1-N1-C5-C6	-179.0(4)	Ni1-N1-C5-C6	-9.7(4)
C3-C4-C5-N1	4.3(6)	C3-C4-C5-C6	180.0(4)
C7-N2-C6-C6	-9.9(7)	Ni1-N2-C6-C6	158.1(4)
C7-N2-C6-C5	171.4(4)	Ni1-N2-C6-C5	-20.7(4)
N1-C5-C6-N2	19.8(5)	C4-C5-C6-N2	-156.2(4)
N1-C5-C6-C6	-158.8(5)	C4-C5-C6-C6	25.3(8)

C6-N2-C7-C7	-11.9(7)	Ni1-N2-C7-C7	-179.7(4)
C6-N2-C7-C8	163.9(4)	Ni1-N2-C7-C8	-3.9(4)
C12-N3-C8-C9	-5.4(6)	Ni1-N3-C8-C9	163.7(3)
C12-N3-C8-C7	-179.9(3)	Ni1-N3-C8-C7	-10.8(4)
N2-C7-C8-N3	9.6(5)	C7-C7-C8-N3	-175.3(5)
N2-C7-C8-C9	-164.8(4)	C7-C7-C8-C9	10.3(8)
N3-C8-C9-C10	5.8(6)	C7-C8-C9-C10	179.7(4)
C8-C9-C10-C11	-0.9(6)	C9-C10-C11-C12	-4.0(6)
C8-N3-C12-C11	0.2(6)	Ni1-N3-C12-C11	-167.6(3)
C10-C11-C12-N3	4.6(7)	O8-P1-C13-C14	51.1(4)
O6-P1-C13-C14	-70.1(3)	O7-P1-C13-C14	173.2(3)
P1-C13-C14-C15	-132.1(3)	P1-C13-C14-C16	47.9(5)
C16-C14-C15-C17	-2.7(6)	C13-C14-C15-C17	177.3(4)
C15-C14-C16-C18	2.4(6)	C13-C14-C16-C18	-177.6(4)
C14-C15-C17-C19	0.3(6)	C14-C16-C18-C19	0.1(6)
C15-C17-C19-C18	2.2(6)	C15-C17-C19-C20	-176.4(4)
C16-C18-C19-C17	-2.4(6)	C16-C18-C19-C20	176.2(4)
C17-C19-C20-P2	-98.3(4)	C18-C19-C20-P2	83.1(4)
O13-P2-C20-C19	-65.7(3)	O11-P2-C20-C19	175.5(3)
O12-P2-C20-C19	56.0(3)		

Table S77. Anisotropic atomic displacement parameters ($Å^2$) for $[\{Ni_2(tpypyz)(H_2O)_4\}Mo_4O_{10}(1,4\text{-}O_3PC_8H_8PO_3)_2]\cdot 4H_2O$ (**10**·$4H_2O$).

The anisotropic atomic displacement factor exponent takes the form: $-2\pi^2[\, h^2\, a^{*2}\, U_{11} + ... + 2\, h\, k\, a^*\, b^*\, U_{12}\,]$

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
Mo1	0.0149(2)	0.0137(2)	0.00270(18)	0.00007(13)	0.00368(14)	0.00099(13)
Mo2	0.0127(2)	0.0136(2)	0.00254(19)	0.00088(13)	0.00304(14)	0.00148(13)
Ni1	0.0139(3)	0.0155(3)	0.0046(3)	-0.0005(2)	0.0043(2)	0.0010(2)
P1	0.0144(5)	0.0134(5)	0.0031(5)	0.0003(4)	0.0045(4)	0.0016(4)
P2	0.0138(5)	0.0116(5)	0.0022(5)	0.0006(4)	0.0025(4)	0.0006(4)
O1	0.0192(15)	0.0189(15)	0.0057(14)	0.0016(11)	0.0069(12)	0.0028(12)
O2	0.0215(16)	0.0187(16)	0.0085(15)	-0.0058(12)	0.0042(12)	0.0001(12)
O3	0.0141(14)	0.0168(15)	0.0055(13)	-0.0005(11)	0.0030(11)	0.0019(11)
O4	0.0212(15)	0.0165(15)	0.0039(13)	-0.0004(11)	0.0061(12)	0.0008(12)
O5	0.0165(15)	0.0237(17)	0.0079(14)	0.0004(12)	0.0037(12)	0.0038(12)
O6	0.0153(14)	0.0140(15)	0.0040(13)	0.0002(10)	0.0036(11)	0.0014(11)
O7	0.0148(14)	0.0157(15)	0.0088(14)	0.0018(12)	0.0056(12)	0.0016(11)
O8	0.0182(15)	0.0226(16)	0.0042(13)	0.0041(12)	0.0047(12)	0.0064(12)
O9	0.0176(15)	0.0250(17)	0.0098(15)	-0.0016(13)	-0.0006(12)	0.0065(13)
O10	0.0176(14)	0.0190(16)	0.0039(14)	-0.0008(12)	0.0037(12)	-0.0025(12)
O11	0.0158(14)	0.0151(14)	0.0053(13)	0.0012(11)	0.0052(11)	-0.0006(11)
O12	0.0132(14)	0.0175(15)	0.0044(13)	-0.0015(11)	0.0029(11)	-0.0018(11)
O13	0.0157(14)	0.0156(15)	0.0039(13)	0.0005(11)	0.0039(11)	-0.0006(11)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
O90A	0.041(5)	0.036(11)	0.023(5)	0.007(4)	0.008(4)	0.000(5)
O90B	0.034(4)	0.055(12)	0.015(4)	-0.002(4)	0.011(3)	-0.005(4)
O91	0.033(2)	0.042(2)	0.0196(18)	-0.0040(16)	0.0124(16)	0.0107(18)
N1	0.0126(16)	0.0161(17)	0.0081(17)	-0.0028(14)	0.0035(13)	-0.0017(13)
N2	0.0127(16)	0.0160(17)	0.0033(15)	-0.0005(13)	0.0013(13)	-0.0011(13)
N3	0.0180(17)	0.0196(18)	0.0018(16)	0.0003(13)	0.0037(13)	-0.0006(14)
C1	0.0148(19)	0.017(2)	0.011(2)	-0.0019(17)	0.0060(16)	0.0009(16)
C2	0.015(2)	0.020(2)	0.017(2)	-0.0005(18)	0.0050(17)	0.0022(17)
C3	0.019(2)	0.016(2)	0.015(2)	0.0016(17)	0.0049(17)	0.0013(17)
C4	0.015(2)	0.018(2)	0.010(2)	-0.0014(16)	0.0049(16)	-0.0008(16)
C5	0.0114(18)	0.016(2)	0.0056(19)	-0.0021(15)	-0.0003(15)	-0.0008(15)
C6	0.017(2)	0.016(2)	0.0026(18)	0.0007(15)	0.0018(16)	-0.0014(16)
C7	0.0082(18)	0.016(2)	0.0058(18)	-0.0007(16)	-0.0019(15)	-0.0022(15)
C8	0.0156(19)	0.015(2)	0.0051(18)	0.0012(16)	0.0048(15)	-0.0009(16)
C9	0.019(2)	0.016(2)	0.013(2)	-0.0019(17)	0.0081(17)	-0.0012(16)
C10	0.020(2)	0.019(2)	0.011(2)	0.0010(17)	0.0075(17)	-0.0008(17)
C11	0.021(2)	0.022(2)	0.012(2)	0.0036(18)	0.0082(18)	-0.0008(18)
C12	0.019(2)	0.024(2)	0.010(2)	0.0055(18)	0.0076(17)	0.0010(18)
C13	0.017(2)	0.016(2)	0.0075(19)	-0.0032(16)	0.0036(16)	-0.0024(16)
C14	0.0124(19)	0.016(2)	0.0106(19)	0.0016(16)	0.0052(16)	0.0026(16)
C15	0.017(2)	0.019(2)	0.0048(18)	-0.0007(15)	0.0039(16)	-0.0019(16)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
C16	0.020(2)	0.017(2)	0.0074(19)	-0.0019(16)	0.0061(16)	-0.0014(17)
C17	0.020(2)	0.016(2)	0.0070(19)	-0.0037(16)	0.0058(16)	-0.0037(16)
C18	0.019(2)	0.020(2)	0.0050(19)	-0.0014(16)	0.0032(16)	-0.0026(17)
C19	0.015(2)	0.018(2)	0.011(2)	0.0011(16)	0.0075(16)	-0.0019(16)
C20	0.0132(19)	0.015(2)	0.013(2)	0.0018(16)	0.0079(16)	-0.0003(16)

Table S78. Hydrogen atomic coordinates and isotropic atomic displacement parameters ($Å^2$) for $[\{Ni_2(tpypyz)(H_2O)_4\}Mo_4O_{10}(1,4\text{-}O_3PC_8H_8PO_3)_2]\cdot 4H_2O$ (**10**·$4H_2O$).

	x/a	y/b	z/c	U(eq)
H9C	0.5235(15)	0.993(4)	0.429(3)	0.023
H9D	0.507(2)	1.085(3)	0.436(3)	0.023
H10C	0.327(2)	0.998(4)	0.2271(17)	0.016
H10D	0.318(2)	0.9047(19)	0.260(3)	0.016
H91A	0.440(3)	0.148(3)	0.465(3)	0.037
H91B	0.4145(17)	0.225(5)	0.491(3)	0.037
H1	0.3493	1.2259	0.3433	0.017
H2	0.3415	1.3997	0.2860	0.021
H3	0.3899	1.4253	0.2068	0.021
H4	0.4508	1.2795	0.1924	0.017
H9	0.5329	0.6706	0.3079	0.018
H10	0.5104	0.5311	0.3776	0.019
H11	0.4359	0.5705	0.4227	0.021
H12	0.3951	0.7493	0.4093	0.02
H13A	0.2239	0.9074	0.3899	0.017
H13B	0.2500	0.8951	0.3288	0.017
H15	0.2557	0.7053	0.3092	0.016
H16	0.3143	0.8053	0.5142	0.017
H17	0.2920	0.5244	0.3448	0.017

	x/a	y/b	z/c	U(eq)
H18	0.3501	0.6246	0.5501	0.018
H20A	0.3923	0.4500	0.5144	0.016
H20B	0.3614	0.4054	0.4347	0.016

Table S79. Sample and crystal data for [{Ni(2,2'-dpa)(H_2O)}$_2$$Mo_5O_{15}$(1,4-$O_3PC_8H_8PO_3$)]·2$H_2O$ (**11**·2H_2O).

Identification code	[{Ni(2,2'-dpa)(H_2O)}$_2$$Mo_5O_{15}$(1,4-$O_3PC_8H_8PO_3$)]·2$H_2O$	
Chemical formula	$C_{28}H_{34}Mo_5N_6Ni_2O_{25}P_2$	
Formula weight	1513.67	
Temperature	90(2) K	
Wavelength	0.71073 Å	
Crystal size	0.068 x 0.101 x 0.209 mm	
Crystal system	Monoclinic	
Space group	C2/c	
Unit cell dimensions	a = 17.3753(12) Å	α = 90°
	b = 11.3661(8) Å	β = 103.1180(10)°
	c = 22.3579(15) Å	γ = 90°
Volume	4300.2(5) Å^3	
Z	4	
Density (calculated)	2.338 g/cm^3	
Absorption coefficient	2.440 mm^{-1}	
F(000)	2960	

Table S80. Data collection and structure refinement for [{Ni(2,2'-dpa)(H_2O)}$_2$$Mo_5O_{15}$(1,4-$O_3PC_8H_8PO_3$)]·2$H_2O$ (**11**·2H_2O).

Theta range for data collection	1.87 to 26.37°	
Index ranges	-21<=h<=21, -14<=k<=14, -27<=l<=27	
Reflections collected	33502	
Independent reflections	4381 [R(int) = 0.0651]	
Coverage of independent reflections	100.0%	
Absorption correction	multi-scan	
Max. and min. transmission	0.8520 and 0.6300	
Refinement method	Full-matrix least-squares on F^2	
Refinement program	SHELXL-2013 (Sheldrick, 2013)	
Function minimized	$\Sigma\, w(F_o^2 - F_c^2)^2$	
Data / restraints / parameters	4381 / 96 / 317	
Goodness-of-fit on F^2	1.033	
Δ/σ_{max}	0.001	
Final R indices	3679 data; I>2σ(I)	R1 = 0.0317, wR2 = 0.0636
	all data	R1 = 0.0442, wR2 = 0.0672
Weighting scheme	$w=1/[\sigma^2(F_o^2)+(0.0288P)^2+17.3963P]$ where $P=(F_o^2+2F_c^2)/3$	
Largest diff. peak and hole	1.561 and -0.752 eÅ^{-3}	
R.M.S. deviation from mean	0.130 eÅ^{-3}	

Table S81. Atomic coordinates and equivalent isotropic atomic displacement parameters ($Å^2$) for $[\{Ni(2,2'\text{-dpa})(H_2O)\}_2Mo_5O_{15}(1,4\text{-}O_3PC_8H_8PO_3)]\cdot 2H_2O$ (**11**·$2H_2O$).

U(eq) is defined as one third of the trace of the orthogonalized U_{ij} tensor.

	x/a	**y/b**	**z/c**	**U(eq)**
Mo1	0.39635(2)	0.42202(3)	0.70708(2)	0.00753(9)
Mo2	0.35739(2)	0.70639(3)	0.66782(2)	0.00662(9)
Mo3	0.5	0.87807(4)	0.75	0.00847(11)
Ni1	0.19148(3)	0.90381(5)	0.67989(2)	0.00937(12)
P1	0.04754(6)	0.11857(9)	0.67987(5)	0.0067(2)
O1	0.5	0.3785(3)	0.75	0.0090(8)
O2	0.39153(17)	0.3588(2)	0.63794(13)	0.0122(6)
O3	0.33670(16)	0.3303(3)	0.73937(13)	0.0111(6)
O4	0.32306(15)	0.5516(2)	0.68583(12)	0.0079(6)
O5	0.33608(16)	0.7009(3)	0.58966(12)	0.0123(6)
O6	0.43634(16)	0.8248(2)	0.67418(12)	0.0089(6)
O7	0.56439(17)	0.9700(3)	0.72573(14)	0.0151(6)
O8	0.28043(16)	0.7856(2)	0.68478(12)	0.0111(6)
O9	0.27095(17)	0.0102(3)	0.73826(14)	0.0153(7)
O10	0.09931(15)	0.0105(2)	0.69591(12)	0.0077(6)
O11	0.07622(15)	0.2179(2)	0.72595(12)	0.0082(6)
O12	0.96087(15)	0.0843(2)	0.68083(12)	0.0080(6)
O90	0.3391(10)	0.0844(16)	0.3800(7)	0.306(8)

	x/a	y/b	z/c	U(eq)
N1	0.2212(2)	0.9859(3)	0.60692(15)	0.0109(7)
N2	0.1796(2)	0.8239(3)	0.54005(16)	0.0130(7)
N3	0.1159(2)	0.7933(3)	0.62235(16)	0.0126(7)
C1	0.2556(2)	0.0940(4)	0.6142(2)	0.0155(9)
C2	0.2886(3)	0.1464(4)	0.5712(2)	0.0190(10)
C3	0.2896(3)	0.0855(4)	0.5174(2)	0.0191(10)
C4	0.2552(3)	0.9773(4)	0.5086(2)	0.0158(9)
C5	0.2188(2)	0.9310(4)	0.55338(19)	0.0119(8)
C6	0.1230(2)	0.7695(4)	0.56482(19)	0.0118(8)
C7	0.0744(3)	0.6867(4)	0.5279(2)	0.0144(9)
C8	0.0166(3)	0.6313(4)	0.5502(2)	0.0176(10)
C9	0.0084(3)	0.6560(4)	0.6088(2)	0.0189(10)
C10	0.0593(3)	0.7358(4)	0.6431(2)	0.0161(9)
C11	0.0464(2)	0.1678(4)	0.60291(17)	0.0103(8)
C12	0.0224(2)	0.0793(4)	0.55058(17)	0.0114(8)
C13	0.0630(2)	0.0748(4)	0.50367(18)	0.0107(8)
C14	0.0415(2)	0.9975(4)	0.45441(18)	0.0118(9)

Table S82. Bond lengths (Å) for [{Ni(2,2'-dpa)(H_2O)}$_2$$Mo_5O_{15}$(1,4-$O_3PC_8H_8PO_3$)]·2$H_2O$ (**11**·2H_2O).

Mo1-O2	1.689(3)	Mo1-O3	1.738(3)
Mo1-O1	1.9036(11)	Mo1-O4	1.935(3)
Mo1-O12	2.303(3)	Mo1-O10	2.376(3)
Mo2-O5	1.703(3)	Mo2-O8	1.724(3)
Mo2-O6	1.904(3)	Mo2-O4	1.929(3)
Mo2-O12	2.237(3)	Mo2-O11	2.397(3)
Mo3-O7	1.707(3)	Mo3-O7	1.707(3)
Mo3-O6	1.901(3)	Mo3-O6	1.901(3)
Mo3-O11	2.383(3)	Mo3-O11	2.383(3)
Ni1-O8	2.032(3)	Ni1-N1	2.045(3)
Ni1-N3	2.048(4)	Ni1-O9	2.062(3)
Ni1-O10	2.104(3)	Ni1-O3	2.144(3)
P1-O10	1.516(3)	P1-O11	1.534(3)
P1-O12	1.560(3)	P1-C11	1.805(4)
O1-Mo1	1.9036(11)	O3-Ni1	2.144(3)
O9-H9C	0.824(19)	O9-H9D	0.828(19)
O10-Mo1	2.376(3)	O11-Mo3	2.383(3)
O11-Mo2	2.397(3)	O12-Mo2	2.237(3)
O12-Mo1	2.303(3)	N1-C5	1.342(5)
N1-C1	1.360(6)	N2-C6	1.380(5)

N2-C5	1.393(5)	N2-H2'	0.872(19)
N3-C6	1.347(5)	N3-C10	1.348(5)
C1-C2	1.364(6)	C1-H1	0.95
C2-C3	1.391(6)	C2-H2	0.95
C3-C4	1.362(6)	C3-H3	0.95
C4-C5	1.402(6)	C4-H4	0.95
C6-C7	1.402(6)	C7-C8	1.372(6)
C7-H7	0.95	C8-C9	1.378(6)
C8-H8	0.95	C9-C10	1.373(6)
C9-H9	0.95	C10-H10	0.95
C11-C12	1.528(6)	C11-H11A	0.99
C11-H11B	0.99	C12-C13	1.391(5)
C12-C14	1.397(6)	C13-C14	1.392(6)
C13-H13	0.95	C14-C12	1.397(6)
C14-H14	0.95		

Table S83. Bond angles (°) for [{Ni(2,2'-dpa)(H_2O)}$_2$$Mo_5O_{15}$(1,4-$O_3PC_8H_8PO_3$)]·2$H_2O$ (**11**·2H_2O).

O2-Mo1-O3	102.39(13)	O2-Mo1-O1	101.28(11)
O3-Mo1-O1	102.71(12)	O2-Mo1-O4	101.88(13)
O3-Mo1-O4	97.89(12)	O1-Mo1-O4	144.70(14)
O2-Mo1-O12	91.91(12)	O3-Mo1-O12	163.12(11)
O1-Mo1-O12	82.93(12)	O4-Mo1-O12	70.05(10)
O2-Mo1-O10	178.99(12)	O3-Mo1-O10	76.89(11)
O1-Mo1-O10	79.60(9)	O4-Mo1-O10	77.56(10)
O12-Mo1-O10	88.69(9)	O5-Mo2-O8	104.24(13)
O5-Mo2-O6	95.04(13)	O8-Mo2-O6	101.29(13)
O5-Mo2-O4	100.09(13)	O8-Mo2-O4	97.95(12)
O6-Mo2-O4	151.74(11)	O5-Mo2-O12	95.27(12)
O8-Mo2-O12	159.36(11)	O6-Mo2-O12	83.38(11)
O4-Mo2-O12	71.63(10)	O5-Mo2-O11	164.22(11)
O8-Mo2-O11	88.64(11)	O6-Mo2-O11	73.19(10)
O4-Mo2-O11	86.85(10)	O12-Mo2-O11	73.34(9)
O7-Mo3-O7	104.5(2)	O7-Mo3-O6	101.67(13)
O7-Mo3-O6	100.82(13)	O7-Mo3-O6	100.82(13)
O7-Mo3-O6	101.67(13)	O6-Mo3-O6	142.87(17)
O7-Mo3-O11	167.75(12)	O7-Mo3-O11	87.60(12)
O6-Mo3-O11	73.58(10)	O6-Mo3-O11	78.25(11)

O7-Mo3-O11	87.60(12)	O7-Mo3-O11	167.75(12)
O6-Mo3-O11	78.25(11)	O6-Mo3-O11	73.58(10)
O11-Mo3-O11	80.41(13)	O8-Ni1-N1	91.01(12)
O8-Ni1-N3	89.99(13)	N1-Ni1-N3	91.19(14)
O8-Ni1-O9	87.81(12)	N1-Ni1-O9	89.04(13)
N3-Ni1-O9	177.79(13)	O8-Ni1-O10	166.24(11)
N1-Ni1-O10	102.29(12)	N3-Ni1-O10	93.22(12)
O9-Ni1-O10	88.88(11)	O8-Ni1-O3	90.77(11)
N1-Ni1-O3	175.39(13)	N3-Ni1-O3	93.06(12)
O9-Ni1-O3	86.78(12)	O10-Ni1-O3	75.71(10)
O10-P1-O11	110.64(16)	O10-P1-O12	108.49(16)
O11-P1-O12	110.00(15)	O10-P1-C11	110.76(17)
O11-P1-C11	110.10(18)	O12-P1-C11	106.76(17)
Mo1-O1-Mo1	149.9(2)	Mo1-O3-Ni1	112.80(14)
Mo2-O4-Mo1	121.88(13)	Mo3-O6-Mo2	123.73(14)
Mo2-O8-Ni1	162.15(17)	Ni1-O9-H9C	124.(3)
Ni1-O9-H9D	117.(3)	H9C-O9-H9D	106.(5)
P1-O10-Ni1	149.36(17)	P1-O10-Mo1	116.93(14)
Ni1-O10-Mo1	92.50(10)	P1-O11-Mo3	126.98(15)
P1-O11-Mo2	129.44(15)	Mo3-O11-Mo2	89.16(9)
P1-O12-Mo2	126.37(16)	P1-O12-Mo1	136.94(16)
Mo2-O12-Mo1	96.12(10)	C5-N1-C1	116.8(4)

C5-N1-Ni1	122.5(3)	C1-N1-Ni1	120.0(3)
C6-N2-C5	131.6(4)	C6-N2-H2'	113.(3)
C5-N2-H2'	110.(3)	C6-N3-C10	117.7(4)
C6-N3-Ni1	122.5(3)	C10-N3-Ni1	119.7(3)
N1-C1-C2	123.6(4)	N1-C1-H1	118.2
C2-C1-H1	118.2	C1-C2-C3	118.8(4)
C1-C2-H2	120.6	C3-C2-H2	120.6
C4-C3-C2	119.0(4)	C4-C3-H3	120.5
C2-C3-H3	120.5	C3-C4-C5	119.3(4)
C3-C4-H4	120.3	C5-C4-H4	120.3
N1-C5-N2	120.8(4)	N1-C5-C4	122.3(4)
N2-C5-C4	116.9(4)	N3-C6-N2	121.4(4)
N3-C6-C7	121.6(4)	N2-C6-C7	117.0(4)
C8-C7-C6	119.1(4)	C8-C7-H7	120.4
C6-C7-H7	120.4	C7-C8-C9	119.7(4)
C7-C8-H8	120.2	C9-C8-H8	120.2
C10-C9-C8	118.2(4)	C10-C9-H9	120.9
C8-C9-H9	120.9	N3-C10-C9	123.7(4)
N3-C10-H10	118.1	C9-C10-H10	118.1
C12-C11-P1	117.9(3)	C12-C11-H11A	107.8
P1-C11-H11A	107.8	C12-C11-H11B	107.8
P1-C11-H11B	107.8	H11A-C11-H11B	107.2

C13-C12-C14	116.5(4)	C13-C12-C11	120.3(4)
C14-C12-C11	123.1(3)	C12-C13-C14	122.0(4)
C12-C13-H13	119.0	C14-C13-H13	119.0
C13-C14-C12	121.5(4)	C13-C14-H14	119.2
C12-C14-H14	119.2		

Table S84. Torsion angles (°) for [{Ni(2,2'-dpa)(H_2O)}$_2$$Mo_5O_{15}$(1,4-$O_3PC_8H_8PO_3$)]·2$H_2O$ (**11**·2H_2O).

O2-Mo1-O3-Ni1	-167.65(14)	O1-Mo1-O3-Ni1	87.59(15)
O4-Mo1-O3-Ni1	-63.57(15)	O12-Mo1-O3-Ni1	-20.4(5)
O10-Mo1-O3-Ni1	11.63(12)	O5-Mo2-O8-Ni1	-32.5(6)
O6-Mo2-O8-Ni1	65.7(6)	O4-Mo2-O8-Ni1	-135.1(6)
O12-Mo2-O8-Ni1	167.1(4)	O11-Mo2-O8-Ni1	138.3(6)
O11-P1-O10-Ni1	-101.1(3)	O12-P1-O10-Ni1	138.1(3)
C11-P1-O10-Ni1	21.3(4)	O11-P1-O10-Mo1	61.41(19)
O12-P1-O10-Mo1	-59.34(19)	C11-P1-O10-Mo1	-176.21(17)
O10-P1-O11-Mo3	-156.07(16)	O12-P1-O11-Mo3	-36.2(2)
C11-P1-O11-Mo3	81.2(2)	O10-P1-O11-Mo2	-29.5(2)
O12-P1-O11-Mo2	90.3(2)	C11-P1-O11-Mo2	-152.27(19)
O10-P1-O12-Mo2	175.30(16)	O11-P1-O12-Mo2	54.1(2)
C11-P1-O12-Mo2	-65.3(2)	O10-P1-O12-Mo1	6.1(3)
O11-P1-O12-Mo1	-115.0(2)	C11-P1-O12-Mo1	125.5(2)
C5-N1-C1-C2	-1.7(6)	Ni1-N1-C1-C2	169.1(3)
N1-C1-C2-C3	-1.9(7)	C1-C2-C3-C4	2.1(7)
C2-C3-C4-C5	1.1(7)	C1-N1-C5-N2	-175.2(4)
Ni1-N1-C5-N2	14.3(5)	C1-N1-C5-C4	5.0(6)
Ni1-N1-C5-C4	-165.5(3)	C6-N2-C5-N1	18.7(7)
C6-N2-C5-C4	-161.5(4)	C3-C4-C5-N1	-4.8(7)

C3-C4-C5-N2	175.4(4)	C10-N3-C6-N2	-179.7(4)
Ni1-N3-C6-N2	-3.7(5)	C10-N3-C6-C7	-1.0(6)
Ni1-N3-C6-C7	175.0(3)	C5-N2-C6-N3	-24.7(7)
C5-N2-C6-C7	156.5(4)	N3-C6-C7-C8	2.1(6)
N2-C6-C7-C8	-179.1(4)	C6-C7-C8-C9	-1.4(7)
C7-C8-C9-C10	-0.2(7)	C6-N3-C10-C9	-0.7(6)
Ni1-N3-C10-C9	-176.9(3)	C8-C9-C10-N3	1.3(7)
O10-P1-C11-C12	55.0(4)	O11-P1-C11-C12	177.7(3)
O12-P1-C11-C12	-62.9(3)	P1-C11-C12-C13	-138.7(3)
P1-C11-C12-C14	43.5(5)	C14-C12-C13-C14	-0.3(7)
C11-C12-C13-C14	-178.3(4)	C12-C13-C14-C12	0.4(7)

Table S85. Anisotropic atomic displacement parameters ($Å^2$) for [{Ni(2,2'-dpa)(H_2O)}$_2$$Mo_5O_{15}$(1,4-$O_3PC_8H_8PO_3$)]·2$H_2O$ (**11**·2H_2O).

The anisotropic atomic displacement factor exponent takes the form: $-2\pi^2[h^2 a^{*2} U_{11} + ... + 2 h k a^* b^* U_{12}]$

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
Mo1	0.00614(17)	0.00815(18)	0.00789(17)	-0.00136(14)	0.00072(13)	-0.00053(14)
Mo2	0.00490(17)	0.00900(18)	0.00579(16)	-0.00007(14)	0.00083(13)	0.00146(14)
Mo3	0.0060(2)	0.0076(2)	0.0110(2)	0	0.00035(19)	0
Ni1	0.0073(3)	0.0109(3)	0.0097(3)	-0.0013(2)	0.0015(2)	0.0012(2)
P1	0.0053(5)	0.0095(5)	0.0051(5)	-0.0001(4)	0.0012(4)	0.0011(4)
O1	0.0059(19)	0.009(2)	0.012(2)	0	0.0011(16)	0
O2	0.0138(15)	0.0106(15)	0.0116(14)	-0.0042(12)	0.0015(12)	-0.0017(12)
O3	0.0087(14)	0.0121(15)	0.0123(14)	0.0003(12)	0.0022(12)	-0.0010(12)
O4	0.0043(13)	0.0089(14)	0.0099(14)	0.0003(11)	0.0004(11)	-0.0009(11)
O5	0.0135(15)	0.0170(16)	0.0053(13)	-0.0010(12)	-0.0004(11)	-0.0003(13)
O6	0.0077(14)	0.0088(14)	0.0100(14)	0.0027(11)	0.0014(11)	0.0006(11)
O7	0.0102(15)	0.0124(15)	0.0211(16)	0.0051(13)	-0.0002(13)	-0.0019(12)
O8	0.0084(14)	0.0127(15)	0.0127(14)	-0.0026(12)	0.0036(12)	0.0021(12)
O9	0.0090(15)	0.0223(17)	0.0148(16)	-0.0052(13)	0.0033(13)	0.0008(14)
O10	0.0055(13)	0.0099(14)	0.0072(13)	0.0008(11)	0.0003(11)	0.0020(11)
O11	0.0070(14)	0.0081(14)	0.0106(14)	0.0017(11)	0.0042(11)	0.0008(11)
O12	0.0058(13)	0.0099(14)	0.0082(13)	0.0004(11)	0.0012(11)	0.0011(11)
O90	0.296(18)	0.34(2)	0.272(17)	-0.111(16)	0.050(14)	-0.054(16)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
N1	0.0076(17)	0.0124(18)	0.0114(17)	-0.0011(14)	-0.0003(14)	0.0017(14)
N2	0.0167(19)	0.0132(19)	0.0092(17)	-0.0025(15)	0.0034(15)	0.0003(15)
N3	0.0100(17)	0.0124(18)	0.0148(18)	-0.0036(15)	0.0013(14)	0.0020(14)
C1	0.010(2)	0.017(2)	0.017(2)	-0.0015(18)	-0.0021(17)	0.0002(18)
C2	0.017(2)	0.019(2)	0.020(2)	-0.0021(19)	0.0012(19)	-0.0081(19)
C3	0.016(2)	0.026(3)	0.018(2)	0.003(2)	0.0074(19)	-0.002(2)
C4	0.016(2)	0.020(2)	0.012(2)	-0.0034(18)	0.0044(18)	0.0012(19)
C5	0.007(2)	0.012(2)	0.014(2)	0.0012(17)	-0.0013(16)	0.0016(16)
C6	0.009(2)	0.009(2)	0.015(2)	0.0032(16)	-0.0013(17)	0.0045(16)
C7	0.015(2)	0.013(2)	0.015(2)	-0.0001(17)	0.0043(18)	0.0029(17)
C8	0.015(2)	0.009(2)	0.027(2)	-0.0071(19)	0.0012(19)	-0.0006(18)
C9	0.013(2)	0.019(2)	0.025(2)	-0.002(2)	0.0059(19)	-0.0029(18)
C10	0.014(2)	0.015(2)	0.020(2)	-0.0008(18)	0.0034(18)	-0.0009(18)
C11	0.012(2)	0.013(2)	0.0067(18)	0.0034(16)	0.0025(16)	0.0019(17)
C12	0.012(2)	0.016(2)	0.0057(18)	0.0018(16)	0.0005(16)	0.0052(18)
C13	0.008(2)	0.013(2)	0.0105(19)	0.0030(17)	-0.0008(16)	0.0003(17)
C14	0.014(2)	0.016(2)	0.007(2)	-0.0003(16)	0.0051(17)	0.0040(18)

Table S86. Hydrogen atomic coordinates and isotropic atomic displacement parameters ($Å^2$) for [{Ni(2,2'-dpa)(H_2O)}$_2$$Mo_5O_{15}$(1,4-$O_3PC_8H_8PO_3$)]·2$H_2O$ (**11**·2H_2O).

	x/a	y/b	z/c	U(eq)
H9C	0.3190(12)	0.999(4)	0.747(2)	0.018
H9D	0.259(3)	1.030(4)	0.7707(14)	0.018
H2'	0.177(3)	0.805(4)	0.5018(11)	0.016
H1	0.2567	1.1353	0.6513	0.019
H2	0.3106	1.2232	0.5778	0.023
H3	0.3137	1.1188	0.4873	0.023
H4	0.2560	0.9336	0.4726	0.019
H7	0.0814	0.6691	0.4879	0.017
H8	-0.0177	0.5763	0.5254	0.021
H9	-0.0314	0.6187	0.6250	0.023
H10	0.0543	0.7514	0.6838	0.019
H11A	0.0100	1.2358	0.5938	0.012
H11B	0.0999	1.1970	0.6025	0.012
H13	0.1066	1.1260	0.5053	0.013
H14	0.0710	0.9970	0.4235	0.014

Table S87. Sample and crystal data for [{Cu(o-phen)}$_2$Mo$_2$F$_3$O$_4$(OH)(H$_2$O)$_4$(1,3-O$_3$PC$_8$H$_8$PO$_3$)]·H$_2$O (**12**·H$_2$O).

Identification code	[{Cu(o-phen)}$_2$Mo$_2$F$_3$O$_4$(OH)(H$_2$O)$_4$(1,3-O$_3$PC$_8$H$_8$PO$_3$)]·H$_2$O	
Chemical formula	C$_{32}$H$_{35}$Cu$_2$F$_3$Mo$_2$N$_4$O$_{16}$P$_2$	
Formula weight	1169.54	
Temperature	90(2) K	
Wavelength	0.71073 Å	
Crystal size	0.095 x 0.137 x 0.206 mm	
Crystal system	Triclinic	
Space group	P -1	
Unit cell dimensions	a = 10.9305(12) Å	α = 85.669(2)°
	b = 12.2497(14) Å	β = 76.269(2)°
	c = 16.2618(18) Å	γ = 66.823(2)°
Volume	1944.1(4) Å^3	
Z	2	
Density (calculated)	1.998 g/cm^3	
Absorption coefficient	1.885 mm^{-1}	
F(000)	1164	

Table S88. Data collection and structure refinement for [{Cu(o-phen)}$_2$Mo$_2$F$_3$O$_4$(OH)(H$_2$O)$_4$(1,3-O$_3$PC$_8$H$_8$PO$_3$)]·H$_2$O (**12**·H$_2$O).

Theta range for data collection	2.08 to 28.08°	
Index ranges	-14<=h<=14, -16<=k<=16, -21<=l<=21	
Reflections collected	23327	
Independent reflections	9391 [R(int) = 0.0364]	
Coverage of independent reflections	99.3%	
Max. and min. transmission	0.8410 and 0.6970	
Refinement method	Full-matrix least-squares on F^2	
Refinement program	SHELXL-2013 (Sheldrick, 2013)	
Function minimized	$\Sigma\, w(F_o^2 - F_c^2)^2$	
Data / restraints / parameters	9391 / 15 / 580	
Goodness-of-fit on F^2	1.047	
Δ/σ_{max}	0.010	
Final R indices	7374 data; I>2σ(I)	R1 = 0.0411, wR2 = 0.1162
	all data	R1 = 0.0582, wR2 = 0.1279
Weighting scheme	$w=1/[\sigma^2(F_o^2)+(0.0829P)^2]$ where $P=(F_o^2+2F_c^2)/3$	
Largest diff. peak and hole	1.052 and -1.237 eÅ^{-3}	
R.M.S. deviation from mean	0.167 eÅ^{-3}	

Table S89. Atomic coordinates and equivalent isotropic atomic displacement parameters ($Å^2$) for $[\{Cu(o\text{-}phen)\}_2Mo_2F_3O_4(OH)(H_2O)_4(1,3\text{-}O_3PC_8H_8PO_3)]\cdot H_2O$ (**12**·H_2O).

U(eq) is defined as one third of the trace of the orthogonalized U_{ij} tensor.

	x/a	**y/b**	**z/c**	**U(eq)**
Mo1	0.56915(3)	0.20034(3)	0.92401(2)	0.01328(9)
Mo2	0.81760(3)	0.78912(3)	0.52914(2)	0.01975(10)
Cu1	0.55088(4)	0.20194(4)	0.68758(3)	0.01354(11)
Cu2	0.83247(4)	0.79359(4)	0.79035(3)	0.01228(11)
P1	0.81963(9)	0.05493(8)	0.75925(6)	0.01085(18)
P2	0.57877(9)	0.93440(8)	0.70170(6)	0.01169(18)
F1	0.5921(2)	0.06330(19)	0.00239(13)	0.0165(4)
F2	0.3975(2)	0.2772(2)	0.00332(14)	0.0221(5)
F3	0.8090(3)	0.9307(2)	0.46181(16)	0.0336(6)
O1	0.4440(3)	0.1076(2)	0.89138(16)	0.0156(5)
O2	0.7325(3)	0.0764(2)	0.85061(16)	0.0158(5)
O3	0.7393(3)	0.1199(2)	0.69391(16)	0.0149(5)
O4	0.8920(3)	0.9217(2)	0.74142(16)	0.0131(5)
O5	0.5010(3)	0.0668(2)	0.72511(15)	0.0133(5)
O6	0.5658(4)	0.1538(3)	0.5410(2)	0.0401(8)
O7	0.6505(3)	0.8652(2)	0.76879(16)	0.0152(5)
O8	0.6741(3)	0.9203(2)	0.61336(16)	0.0171(5)
O9	0.9569(3)	0.8438(3)	0.5801(2)	0.0303(7)

	x/a	y/b	z/c	U(eq)
O10	0.7748(3)	0.8399(2)	0.92871(17)	0.0178(5)
O11	0.8313(3)	0.6869(3)	0.6155(2)	0.0341(8)
O12	0.7006(3)	0.7630(3)	0.4891(2)	0.0340(7)
O13	0.5264(3)	0.2878(2)	0.83976(17)	0.0215(6)
O14	0.6561(3)	0.2608(3)	0.96553(18)	0.0258(6)
O15	0.9746(3)	0.7043(3)	0.4560(2)	0.0365(8)
O90	0.8489(5)	0.1122(4)	0.5165(2)	0.0507(10)
N1	0.3580(3)	0.3062(3)	0.68511(19)	0.0158(6)
N2	0.5824(3)	0.3506(3)	0.64874(19)	0.0155(6)
N3	0.0220(3)	0.6960(3)	0.81000(19)	0.0150(6)
N4	0.8096(3)	0.6385(3)	0.8216(2)	0.0156(6)
C1	0.2489(4)	0.2787(4)	0.7032(2)	0.0187(8)
C2	0.1208(4)	0.3595(4)	0.6887(3)	0.0220(8)
C3	0.1104(4)	0.4675(4)	0.6551(3)	0.0242(9)
C4	0.2243(4)	0.4992(4)	0.6364(2)	0.0215(8)
C5	0.3467(4)	0.4143(3)	0.6526(2)	0.0167(7)
C6	0.2233(5)	0.6104(4)	0.6026(3)	0.0269(9)
C7	0.3384(5)	0.6355(3)	0.5871(3)	0.0252(9)
C8	0.4645(4)	0.5502(3)	0.6033(2)	0.0211(8)
C9	0.4672(4)	0.4393(3)	0.6344(2)	0.0152(7)
C10	0.5860(5)	0.5684(4)	0.5887(3)	0.0248(9)

	x/a	y/b	z/c	U(eq)
C11	0.7025(4)	0.4788(4)	0.6045(3)	0.0234(8)
C12	0.6977(4)	0.3704(3)	0.6333(2)	0.0198(8)
C13	0.1263(4)	0.7282(3)	0.8033(3)	0.0195(8)
C14	0.2457(4)	0.6547(4)	0.8300(3)	0.0270(9)
C15	0.2558(4)	0.5473(4)	0.8649(3)	0.0249(9)
C16	0.1450(4)	0.5114(3)	0.8743(2)	0.0192(8)
C17	0.0307(4)	0.5901(3)	0.8456(2)	0.0154(7)
C18	0.1440(5)	0.4009(4)	0.9102(3)	0.0268(9)
C19	0.0346(5)	0.3733(3)	0.9169(3)	0.0253(9)
C20	0.9161(4)	0.4510(3)	0.8872(2)	0.0217(8)
C21	0.9158(4)	0.5594(3)	0.8519(2)	0.0161(7)
C22	0.8009(5)	0.4252(4)	0.8893(3)	0.0266(9)
C23	0.6959(4)	0.5053(4)	0.8565(3)	0.0283(10)
C24	0.7027(4)	0.6115(4)	0.8240(3)	0.0207(8)
C25	0.4512(4)	0.8782(3)	0.6924(2)	0.0157(7)
C26	0.3162(4)	0.9296(3)	0.7560(2)	0.0141(7)
C27	0.3063(4)	0.9161(4)	0.8429(2)	0.0182(7)
C28	0.1978(4)	0.9954(3)	0.7276(2)	0.0141(7)
C29	0.1815(4)	0.9679(4)	0.8993(2)	0.0213(8)
C30	0.0705(4)	0.0481(3)	0.7844(2)	0.0137(7)
C31	0.0638(4)	0.0339(4)	0.8711(2)	0.0210(8)

	x/a	y/b	z/c	U(eq)
C32	0.9459(4)	0.1189(3)	0.7502(2)	0.0153(7)

Table S90. Bond lengths (Å) for [{Cu(o-phen)}$_2$Mo$_2$F$_3$O$_4$(OH)(H$_2$O)$_4$(1,3-O$_3$PC$_8$H$_8$PO$_3$)]·H$_2$O (**12**·H$_2$O).

Mo1-O14	1.686(3)	Mo1-O13	1.712(3)
Mo1-F2	1.927(2)	Mo1-F1	2.001(2)
Mo1-O2	2.011(3)	Mo1-O1	2.259(3)
Mo2-O12	1.706(3)	Mo2-O11	1.802(3)
Mo2-O15	1.804(3)	Mo2-F3	1.962(3)
Mo2-O8	2.061(3)	Mo2-O9	2.222(3)
Cu1-O3	1.928(3)	Cu1-O5	1.951(3)
Cu1-N1	1.998(3)	Cu1-N2	2.012(3)
Cu1-O6	2.450(4)	Cu2-O7	1.939(3)
Cu2-O4	1.972(2)	Cu2-N4	2.024(3)
Cu2-N3	2.032(3)	Cu2-O10	2.241(3)
P1-O3	1.516(2)	P1-O4	1.523(3)
P1-O2	1.541(3)	P1-C32	1.810(4)
P2-O7	1.505(3)	P2-O5	1.534(3)
P2-O8	1.538(3)	P2-C25	1.820(4)
O1-H1C	0.830(18)	O1-H1D	0.827(18)
O6-H6C	0.885(19)	O6-H6D	0.870(19)
O9-H9C	0.874(19)	O9-H9D	0.868(19)
O10-H10C	0.814(19)	O10-H10D	0.820(19)
O90-H90A	0.897(19)	O90-H90B	0.886(19)

N1-C1	1.325(5)	N1-C5	1.358(5)
N2-C12	1.339(5)	N2-C9	1.359(5)
N3-C13	1.326(5)	N3-C17	1.357(5)
N4-C24	1.327(5)	N4-C21	1.358(5)
C1-C2	1.423(5)	C1-H1	0.95
C2-C3	1.365(6)	C2-H2	0.95
C3-C4	1.406(6)	C3-H3	0.95
C4-C5	1.404(5)	C4-C6	1.427(6)
C5-C9	1.426(5)	C6-C7	1.372(7)
C6-H6	0.95	C7-C8	1.434(6)
C7-H7	0.95	C8-C10	1.395(6)
C8-C9	1.405(5)	C10-C11	1.381(6)
C10-H10	0.95	C11-C12	1.390(5)
C11-H11	0.95	C12-H12	0.95
C13-C14	1.409(5)	C13-H13	0.95
C14-C15	1.366(6)	C14-H14	0.95
C15-C16	1.416(6)	C15-H15	0.95
C16-C17	1.403(5)	C16-C18	1.435(5)
C17-C21	1.426(5)	C18-C19	1.344(6)
C18-H18	0.95	C19-C20	1.441(6)
C19-H19	0.95	C20-C21	1.404(5)
C20-C22	1.405(6)	C22-C23	1.376(6)

C22-H22	0.95	C23-C24	1.389(6)
C23-H23	0.95	C24-H24	0.95
C25-C26	1.513(5)	C25-H25A	0.99
C25-H25B	0.99	C26-C27	1.393(5)
C26-C28	1.395(5)	C27-C29	1.385(5)
C27-H27	0.95	C28-C30	1.407(5)
C28-H28	0.95	C29-C31	1.388(6)
C29-H29	0.95	C30-C31	1.396(5)
C30-C32	1.507(5)	C31-H31	0.95
C32-C30	1.507(5)	C32-H32A	0.99
C32-H32B	0.99		

Table S91. Bond angles (°) for [{Cu(o-phen)}$_2$Mo$_2$F$_3$O$_4$(OH)(H$_2$O)$_4$(1,3-O$_3$PC$_8$H$_8$PO$_3$)]·H$_2$O (**12**·H$_2$O).

O14-Mo1-O13	104.37(14)	O14-Mo1-F2	95.29(12)
O13-Mo1-F2	96.48(12)	O14-Mo1-F1	96.55(12)
O13-Mo1-F1	158.83(12)	F2-Mo1-F1	84.62(9)
O14-Mo1-O2	96.92(13)	O13-Mo1-O2	93.02(12)
F2-Mo1-O2	162.17(10)	F1-Mo1-O2	81.13(10)
O14-Mo1-O1	169.99(12)	O13-Mo1-O1	84.69(12)
F2-Mo1-O1	79.22(10)	F1-Mo1-O1	74.72(9)
O2-Mo1-O1	86.70(10)	O12-Mo2-O11	99.67(17)
O12-Mo2-O15	101.35(15)	O11-Mo2-O15	98.62(16)
O12-Mo2-F3	95.06(14)	O11-Mo2-F3	161.67(14)
O15-Mo2-F3	89.03(15)	O12-Mo2-O8	94.86(13)
O11-Mo2-O8	88.29(13)	O15-Mo2-O8	160.98(14)
F3-Mo2-O8	79.61(11)	O12-Mo2-O9	173.86(14)
O11-Mo2-O9	84.63(14)	O15-Mo2-O9	82.21(13)
F3-Mo2-O9	79.91(12)	O8-Mo2-O9	80.82(11)
O3-Cu1-O5	94.52(10)	O3-Cu1-N1	172.48(12)
O5-Cu1-N1	91.85(12)	O3-Cu1-N2	91.24(12)
O5-Cu1-N2	174.23(12)	N1-Cu1-N2	82.38(13)
O3-Cu1-O6	100.30(12)	O5-Cu1-O6	89.34(11)
N1-Cu1-O6	83.74(13)	N2-Cu1-O6	89.92(12)

O7-Cu2-O4	96.23(10)	O7-Cu2-N4	91.22(12)
O4-Cu2-N4	166.98(12)	O7-Cu2-N3	171.82(12)
O4-Cu2-N3	90.60(11)	N4-Cu2-N3	81.25(12)
O7-Cu2-O10	97.59(10)	O4-Cu2-O10	101.16(10)
N4-Cu2-O10	88.38(11)	N3-Cu2-O10	85.44(11)
O3-P1-O4	112.92(15)	O3-P1-O2	113.28(14)
O4-P1-O2	109.00(14)	O3-P1-C32	103.91(16)
O4-P1-C32	109.23(16)	O2-P1-C32	108.25(16)
O7-P2-O5	112.43(15)	O7-P2-O8	113.55(15)
O5-P2-O8	109.46(14)	O7-P2-C25	108.25(16)
O5-P2-C25	106.30(16)	O8-P2-C25	106.40(16)
Mo1-O1-H1C	109.(3)	Mo1-O1-H1D	114.(3)
H1C-O1-H1D	106.(3)	P1-O2-Mo1	141.17(16)
P1-O3-Cu1	138.20(16)	P1-O4-Cu2	128.65(15)
P2-O5-Cu1	130.12(15)	Cu1-O6-H6C	97.(4)
Cu1-O6-H6D	112.(4)	H6C-O6-H6D	106.(4)
P2-O7-Cu2	140.01(16)	P2-O8-Mo2	140.11(16)
Mo2-O9-H9C	98.(4)	Mo2-O9-H9D	117.(4)
H9C-O9-H9D	95.(3)	Cu2-O10-H10C	102.(3)
Cu2-O10-H10D	111.(3)	H10C-O10-H10D	108.(4)
H90A-O90-H90B	98.(4)	C1-N1-C5	119.1(3)
C1-N1-Cu1	128.5(3)	C5-N1-Cu1	112.1(2)

C12-N2-C9	118.4(3)	C12-N2-Cu1	129.6(3)
C9-N2-Cu1	112.0(2)	C13-N3-C17	118.3(3)
C13-N3-Cu2	128.9(3)	C17-N3-Cu2	112.1(2)
C24-N4-C21	118.6(3)	C24-N4-Cu2	128.9(3)
C21-N4-Cu2	112.1(2)	N1-C1-C2	121.5(4)
N1-C1-H1	119.3	C2-C1-H1	119.3
C3-C2-C1	119.2(4)	C3-C2-H2	120.4
C1-C2-H2	120.4	C2-C3-C4	120.4(4)
C2-C3-H3	119.8	C4-C3-H3	119.8
C5-C4-C3	116.8(4)	C5-C4-C6	118.6(4)
C3-C4-C6	124.6(4)	N1-C5-C4	123.1(4)
N1-C5-C9	116.8(3)	C4-C5-C9	120.1(3)
C7-C6-C4	121.2(4)	C7-C6-H6	119.4
C4-C6-H6	119.4	C6-C7-C8	121.1(4)
C6-C7-H7	119.4	C8-C7-H7	119.4
C10-C8-C9	117.4(4)	C10-C8-C7	124.7(4)
C9-C8-C7	117.9(4)	N2-C9-C8	122.7(4)
N2-C9-C5	116.3(3)	C8-C9-C5	120.9(3)
C11-C10-C8	119.8(4)	C11-C10-H10	120.1
C8-C10-H10	120.1	C10-C11-C12	119.4(4)
C10-C11-H11	120.3	C12-C11-H11	120.3
N2-C12-C11	122.2(4)	N2-C12-H12	118.9

C11-C12-H12	118.9	N3-C13-C14	121.9(4)
N3-C13-H13	119.1	C14-C13-H13	119.1
C15-C14-C13	120.1(4)	C15-C14-H14	120.0
C13-C14-H14	120.0	C14-C15-C16	119.3(4)
C14-C15-H15	120.3	C16-C15-H15	120.3
C17-C16-C15	116.6(3)	C17-C16-C18	119.2(4)
C15-C16-C18	124.2(4)	N3-C17-C16	123.8(3)
N3-C17-C21	116.3(3)	C16-C17-C21	119.8(3)
C19-C18-C16	120.5(4)	C19-C18-H18	119.7
C16-C18-H18	119.7	C18-C19-C20	121.9(4)
C18-C19-H19	119.1	C20-C19-H19	119.1
C21-C20-C22	117.3(4)	C21-C20-C19	118.1(4)
C22-C20-C19	124.6(4)	N4-C21-C20	122.8(4)
N4-C21-C17	116.8(3)	C20-C21-C17	120.4(3)
C23-C22-C20	119.0(4)	C23-C22-H22	120.5
C20-C22-H22	120.5	C22-C23-C24	120.1(4)
C22-C23-H23	120.0	C24-C23-H23	120.0
N4-C24-C23	122.2(4)	N4-C24-H24	118.9
C23-C24-H24	118.9	C26-C25-P2	115.2(2)
C26-C25-H25A	108.5	P2-C25-H25A	108.5
C26-C25-H25B	108.5	P2-C25-H25B	108.5
H25A-C25-H25B	107.5	C27-C26-C28	118.5(3)

C27-C26-C25	122.1(3)	C28-C26-C25	119.4(3)
C29-C27-C26	120.4(3)	C29-C27-H27	119.8
C26-C27-H27	119.8	C26-C28-C30	121.5(3)
C26-C28-H28	119.2	C30-C28-H28	119.2
C27-C29-C31	121.1(3)	C27-C29-H29	119.4
C31-C29-H29	119.4	C31-C30-C28	118.7(3)
C31-C30-C32	121.9(3)	C28-C30-C32	119.3(3)
C29-C31-C30	119.7(4)	C29-C31-H31	120.1
C30-C31-H31	120.1	C30-C32-P1	117.2(2)
C30-C32-H32A	108.0	P1-C32-H32A	108.0
C30-C32-H32B	108.0	P1-C32-H32B	108.0
H32A-C32-H32B	107.3		

Table S92. Torsion angles (°) for [{Cu(o-phen)}$_2$Mo$_2$F$_3$O$_4$(OH)(H$_2$O)$_4$(1,3-O$_3$PC$_8$H$_8$PO$_3$)]·H$_2$O (**12**·H$_2$O).

O3-P1-O2-Mo1	-30.5(3)	O4-P1-O2-Mo1	-157.1(2)
C32-P1-O2-Mo1	84.2(3)	O4-P1-O3-Cu1	110.7(2)
O2-P1-O3-Cu1	-13.8(3)	C32-P1-O3-Cu1	-131.0(2)
O3-P1-O4-Cu2	-97.6(2)	O2-P1-O4-Cu2	29.2(2)
C32-P1-O4-Cu2	147.29(19)	O7-P2-O5-Cu1	100.7(2)
O8-P2-O5-Cu1	-26.4(2)	C25-P2-O5-Cu1	-140.99(19)
O5-P2-O7-Cu2	-103.8(3)	O8-P2-O7-Cu2	21.2(3)
C25-P2-O7-Cu2	139.1(2)	O7-P2-O8-Mo2	51.5(3)
O5-P2-O8-Mo2	178.1(2)	C25-P2-O8-Mo2	-67.5(3)
C5-N1-C1-C2	0.5(5)	Cu1-N1-C1-C2	-172.4(3)
N1-C1-C2-C3	0.3(6)	C1-C2-C3-C4	-1.1(6)
C2-C3-C4-C5	1.1(6)	C2-C3-C4-C6	-178.8(4)
C1-N1-C5-C4	-0.5(5)	Cu1-N1-C5-C4	173.5(3)
C1-N1-C5-C9	179.9(3)	Cu1-N1-C5-C9	-6.1(4)
C3-C4-C5-N1	-0.3(6)	C6-C4-C5-N1	179.6(3)
C3-C4-C5-C9	179.3(3)	C6-C4-C5-C9	-0.8(5)
C5-C4-C6-C7	-0.9(6)	C3-C4-C6-C7	179.0(4)
C4-C6-C7-C8	0.8(6)	C6-C7-C8-C10	179.4(4)
C6-C7-C8-C9	1.0(6)	C12-N2-C9-C8	0.6(5)
Cu1-N2-C9-C8	-177.2(3)	C12-N2-C9-C5	-179.4(3)

Cu1-N2-C9-C5	2.8(4)	C10-C8-C9-N2	-1.2(5)
C7-C8-C9-N2	177.4(3)	C10-C8-C9-C5	178.8(3)
C7-C8-C9-C5	-2.6(5)	N1-C5-C9-N2	2.2(5)
C4-C5-C9-N2	-177.4(3)	N1-C5-C9-C8	-177.8(3)
C4-C5-C9-C8	2.6(5)	C9-C8-C10-C11	0.2(6)
C7-C8-C10-C11	-178.3(4)	C8-C10-C11-C12	1.3(6)
C9-N2-C12-C11	1.0(6)	Cu1-N2-C12-C11	178.4(3)
C10-C11-C12-N2	-2.0(6)	C17-N3-C13-C14	1.6(6)
Cu2-N3-C13-C14	171.4(3)	N3-C13-C14-C15	-0.7(7)
C13-C14-C15-C16	-0.4(7)	C14-C15-C16-C17	0.6(6)
C14-C15-C16-C18	-179.8(4)	C13-N3-C17-C16	-1.3(5)
Cu2-N3-C17-C16	-172.8(3)	C13-N3-C17-C21	179.7(3)
Cu2-N3-C17-C21	8.2(4)	C15-C16-C17-N3	0.2(6)
C18-C16-C17-N3	-179.4(4)	C15-C16-C17-C21	179.2(3)
C18-C16-C17-C21	-0.4(6)	C17-C16-C18-C19	-0.5(6)
C15-C16-C18-C19	180.0(4)	C16-C18-C19-C20	1.3(7)
C18-C19-C20-C21	-1.3(6)	C18-C19-C20-C22	177.4(4)
C24-N4-C21-C20	-1.2(5)	Cu2-N4-C21-C20	172.3(3)
C24-N4-C21-C17	177.9(3)	Cu2-N4-C21-C17	-8.6(4)
C22-C20-C21-N4	0.8(6)	C19-C20-C21-N4	179.6(4)
C22-C20-C21-C17	-178.3(4)	C19-C20-C21-C17	0.4(5)
N3-C17-C21-N4	0.2(5)	C16-C17-C21-N4	-178.9(3)

N3-C17-C21-C20	179.4(3)	C16-C17-C21-C20	0.3(5)
C21-C20-C22-C23	0.7(6)	C19-C20-C22-C23	-178.0(4)
C20-C22-C23-C24	-1.8(7)	C21-N4-C24-C23	0.1(6)
Cu2-N4-C24-C23	-172.1(3)	C22-C23-C24-N4	1.4(7)
O7-P2-C25-C26	83.4(3)	O5-P2-C25-C26	-37.6(3)
O8-P2-C25-C26	-154.2(3)	P2-C25-C26-C27	-61.2(4)
P2-C25-C26-C28	117.1(3)	C28-C26-C27-C29	-0.5(6)
C25-C26-C27-C29	177.8(3)	C27-C26-C28-C30	0.4(5)
C25-C26-C28-C30	-178.0(3)	C26-C27-C29-C31	0.1(6)
C26-C28-C30-C31	0.1(5)	C26-C28-C30-C32	179.6(3)
C27-C29-C31-C30	0.4(6)	C28-C30-C31-C29	-0.5(6)
C32-C30-C31-C29	180.0(4)	O3-P1-C32-C30	-160.1(3)
O4-P1-C32-C30	-39.3(3)	O2-P1-C32-C30	79.3(3)

Table S93. Anisotropic atomic displacement parameters ($Å^2$) for [{Cu(o-phen)}$_2$$Mo_2F_3O_4(OH)(H_2O)_4$(1,3-$O_3PC_8H_8PO_3$)]·$H_2O$ (**12**·H_2O).

The anisotropic atomic displacement factor exponent takes the form: $-2\pi^2[h^2 a^{*2} U_{11} + ... + 2 h k a^* b^* U_{12}]$

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
Mo1	0.01154(16)	0.01372(16)	0.01369(15)	0.00074(11)	0.00036(12)	-0.00588(12)
Mo2	0.01504(18)	0.02022(18)	0.02060(18)	-0.00129(13)	-0.00327(13)	-0.00342(14)
Cu1	0.0080(2)	0.0141(2)	0.0162(2)	0.00527(16)	-0.00181(17)	-0.00325(17)
Cu2	0.0087(2)	0.0127(2)	0.0153(2)	0.00317(16)	-0.00298(16)	-0.00440(17)
P1	0.0066(4)	0.0120(4)	0.0128(4)	0.0025(3)	-0.0009(3)	-0.0035(3)
P2	0.0071(4)	0.0146(4)	0.0115(4)	-0.0004(3)	-0.0008(3)	-0.0029(3)
F1	0.0174(11)	0.0193(11)	0.0155(10)	0.0054(8)	-0.0048(9)	-0.0103(9)
F2	0.0191(12)	0.0226(12)	0.0191(11)	-0.0042(9)	0.0044(9)	-0.0067(10)
F3	0.0407(16)	0.0449(16)	0.0229(13)	0.0107(11)	-0.0105(12)	-0.0246(13)
O1	0.0175(13)	0.0191(13)	0.0111(12)	0.0016(10)	-0.0022(10)	-0.0090(11)
O2	0.0099(12)	0.0179(13)	0.0150(12)	0.0014(10)	0.0016(10)	-0.0031(10)
O3	0.0080(12)	0.0199(13)	0.0140(12)	0.0042(10)	-0.0013(10)	-0.0039(10)
O4	0.0114(12)	0.0108(11)	0.0173(12)	0.0020(9)	-0.0027(10)	-0.0051(10)
O5	0.0099(12)	0.0162(12)	0.0114(11)	-0.0006(9)	-0.0016(9)	-0.0030(10)
O6	0.041(2)	0.0361(19)	0.0333(18)	0.0027(15)	-0.0050(17)	-0.0067(17)
O7	0.0096(12)	0.0186(13)	0.0163(12)	0.0032(10)	-0.0036(10)	-0.0043(10)
O8	0.0136(13)	0.0185(13)	0.0139(12)	-0.0005(10)	0.0004(10)	-0.0025(11)
O9	0.0224(16)	0.0351(17)	0.0257(16)	-0.0058(13)	-0.0065(13)	-0.0014(14)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
O10	0.0157(14)	0.0161(13)	0.0176(13)	0.0017(10)	0.0010(11)	-0.0049(11)
O11	0.0342(19)	0.0209(15)	0.0349(18)	0.0066(13)	-0.0051(15)	-0.0003(14)
O12	0.0308(18)	0.0416(19)	0.0332(17)	-0.0104(14)	0.0008(14)	-0.0207(16)
O13	0.0181(14)	0.0162(13)	0.0219(14)	0.0078(11)	0.0005(11)	-0.0023(11)
O14	0.0291(16)	0.0318(16)	0.0247(15)	-0.0035(12)	0.0004(13)	-0.0233(14)
O15	0.0142(15)	0.058(2)	0.0276(16)	-0.0278(15)	0.0017(13)	-0.0022(15)
O90	0.065(3)	0.055(2)	0.038(2)	0.0031(18)	-0.002(2)	-0.035(2)
N1	0.0125(15)	0.0157(15)	0.0157(15)	-0.0006(12)	-0.0025(12)	-0.0021(12)
N2	0.0145(15)	0.0153(15)	0.0143(14)	0.0028(12)	-0.0025(12)	-0.0042(12)
N3	0.0126(15)	0.0140(14)	0.0160(15)	0.0008(12)	-0.0014(12)	-0.0039(12)
N4	0.0134(15)	0.0140(14)	0.0176(15)	0.0001(12)	-0.0020(12)	-0.0043(12)
C1	0.0117(17)	0.0231(19)	0.0198(18)	-0.0044(15)	-0.0036(15)	-0.0043(15)
C2	0.0119(18)	0.029(2)	0.0222(19)	-0.0074(16)	-0.0026(15)	-0.0035(16)
C3	0.017(2)	0.025(2)	0.023(2)	-0.0063(16)	-0.0063(16)	0.0018(16)
C4	0.020(2)	0.0215(19)	0.0159(18)	-0.0005(14)	-0.0071(15)	0.0010(16)
C5	0.0157(18)	0.0157(17)	0.0139(16)	-0.0008(13)	-0.0027(14)	-0.0012(14)
C6	0.029(2)	0.020(2)	0.020(2)	0.0009(15)	-0.0062(17)	0.0030(17)
C7	0.029(2)	0.0132(18)	0.023(2)	0.0032(15)	-0.0037(18)	0.0008(16)
C8	0.026(2)	0.0139(18)	0.0159(18)	-0.0010(14)	-0.0011(16)	-0.0023(16)
C9	0.0132(17)	0.0145(17)	0.0120(16)	-0.0031(13)	0.0015(13)	-0.0012(14)
C10	0.033(2)	0.0163(18)	0.0195(19)	0.0006(15)	0.0042(17)	-0.0101(17)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
C11	0.025(2)	0.022(2)	0.024(2)	0.0002(16)	0.0002(17)	-0.0135(17)
C12	0.0177(19)	0.0205(19)	0.0175(18)	-0.0004(14)	-0.0001(15)	-0.0057(16)
C13	0.0126(18)	0.0145(17)	0.030(2)	0.0021(15)	-0.0056(16)	-0.0033(15)
C14	0.0123(19)	0.026(2)	0.042(3)	0.0032(19)	-0.0098(18)	-0.0056(17)
C15	0.0140(19)	0.022(2)	0.033(2)	0.0012(17)	-0.0097(17)	0.0011(16)
C16	0.0174(19)	0.0160(18)	0.0192(18)	0.0012(14)	-0.0056(15)	-0.0007(15)
C17	0.0134(18)	0.0152(17)	0.0129(16)	-0.0005(13)	0.0006(14)	-0.0026(14)
C18	0.029(2)	0.0182(19)	0.027(2)	0.0069(16)	-0.0067(18)	-0.0033(17)
C19	0.028(2)	0.0134(18)	0.025(2)	0.0064(15)	-0.0003(17)	-0.0027(16)
C20	0.025(2)	0.0153(18)	0.0192(18)	0.0024(14)	0.0032(16)	-0.0074(16)
C21	0.0171(18)	0.0143(17)	0.0133(16)	-0.0012(13)	0.0002(14)	-0.0041(14)
C22	0.027(2)	0.0173(19)	0.032(2)	0.0011(16)	0.0042(18)	-0.0108(17)
C23	0.020(2)	0.029(2)	0.040(3)	-0.0008(19)	0.0000(19)	-0.0177(19)
C24	0.0153(19)	0.025(2)	0.0230(19)	-0.0005(15)	-0.0016(15)	-0.0105(16)
C25	0.0101(16)	0.0197(18)	0.0160(17)	-0.0039(14)	-0.0006(14)	-0.0049(14)
C26	0.0110(17)	0.0158(17)	0.0173(17)	0.0007(13)	-0.0031(14)	-0.0073(14)
C27	0.0134(18)	0.028(2)	0.0158(17)	0.0037(15)	-0.0042(14)	-0.0112(16)
C28	0.0133(18)	0.0151(17)	0.0152(17)	-0.0001(13)	-0.0028(14)	-0.0070(14)
C29	0.0175(19)	0.036(2)	0.0127(17)	0.0025(15)	-0.0026(15)	-0.0133(17)
C30	0.0103(16)	0.0187(17)	0.0165(17)	0.0029(13)	-0.0033(14)	-0.0106(14)
C31	0.0147(18)	0.035(2)	0.0161(18)	-0.0012(16)	-0.0001(15)	-0.0141(17)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
C32	0.0097(17)	0.0161(17)	0.0204(18)	0.0022(14)	-0.0037(14)	-0.0055(14)

Table S94. Hydrogen atomic coordinates and isotropic atomic displacement parameters ($Å^2$) for $[\{Cu(o\text{-}phen)\}_2Mo_2F_3O_4(OH)(H_2O)_4(1,3\text{-}O_3PC_8H_8PO_3)]\cdot H_2O$ (**12**·H_2O).

	x/a	y/b	z/c	U(eq)
H1C	0.469(4)	1.092(3)	0.8398(12)	0.019
H1D	0.455(5)	1.043(2)	0.915(2)	0.019
H6C	0.617(4)	1.077(2)	0.542(4)	0.048
H6D	0.488(3)	1.157(4)	0.533(4)	0.048
H9C	0.930(6)	0.917(2)	0.563(3)	0.036
H9D	0.934(5)	0.862(4)	0.6337(13)	0.036
H10C	0.742(4)	0.9121(17)	0.928(3)	0.021
H10D	0.715(3)	0.817(3)	0.954(3)	0.021
H90A	0.849(7)	0.042(3)	0.504(3)	0.061
H90B	0.830(7)	0.107(5)	0.5724(13)	0.061
H1	0.2563	1.2035	0.7264	0.022
H2	0.0435	1.3386	0.7023	0.026
H3	0.0258	1.5214	0.6443	0.029
H6	0.1415	1.6681	0.5906	0.032
H7	0.3345	1.7106	0.5653	0.03
H10	0.5886	1.6423	0.5679	0.03
H11	0.7852	1.4910	0.5958	0.028
H12	0.7790	1.3084	0.6423	0.024
H13	1.1205	0.8030	0.7798	0.023

	x/a	y/b	z/c	U(eq)
H14	1.3192	0.6799	0.8238	0.032
H15	1.3363	0.4973	0.8826	0.03
H18	1.2213	0.3469	0.9293	0.032
H19	1.0355	0.3005	0.9419	0.03
H22	0.7958	0.3534	0.9131	0.032
H23	0.6186	0.4880	0.8561	0.034
H24	0.6283	0.6666	0.8027	0.025
H25A	0.4339	0.8946	0.6348	0.019
H25B	0.4895	0.7909	0.6985	0.019
H27	0.3857	0.8711	0.8636	0.022
H28	0.2032	1.0047	0.6685	0.017
H29	0.1764	0.9580	0.9584	0.026
H31	-0.0209	1.0693	0.9106	0.025
H32A	0.9769	1.1331	0.6896	0.018
H32B	0.9003	1.1973	0.7797	0.018

Table S95. Sample and crystal data for [{Cu(terpy)}$_2$(H$_2$O)(OH)Mo$_2$F$_4$O$_3$(1,2-O$_3$PC$_8$H$_8$PO$_3$)][{Cu(terpy)(H$_2$O)}Mo$_2$F$_3$O$_4$(1,2-O$_3$PC$_8$H$_8$PO$_3$)]·H$_2$O (**13**·H$_2$O).

Identification code	[{Cu(terpy)}$_2$(H$_2$O)(OH)Mo$_2$F$_4$O$_3$(1,2-O$_3$PC$_8$H$_8$PO$_3$)][{Cu(terpy)(H$_2$O)}Mo$_2$F$_3$O$_4$(1,2-O$_3$PC$_8$H$_8$PO$_3$)] ·H$_2$O	
Chemical formula	C$_{61}$H$_{55}$Cu$_3$F$_6$Mo$_4$N$_9$O$_{24}$P$_4$	
Formula weight	2110.40	
Temperature	90(2) K	
Wavelength	0.71073 Å	
Crystal size	0.044 x 0.071 x 0.397 mm	
Crystal system	Triclinic	
Space group	P -1	
Unit cell dimensions	a = 8.9990(6) Å	α = 113.584(2)°
	b = 20.6937(16) Å	β = 96.090(2)°
	c = 21.1594(16) Å	γ = 95.628(2)°
Volume	3547.9(5) Å^3	
Z	2	
Density (calculated)	1.975 g/cm^3	
Absorption coefficient	1.758 mm^{-1}	
F(000)	2090	

Table S96. Data collection and structure refinement for [{Cu(terpy)}$_2$(H$_2$O)(OH)Mo$_2$F$_4$O$_3$(1,2-O$_3$PC$_8$H$_8$PO$_3$)][{Cu(terpy)(H$_2$O)}Mo$_2$F$_3$O$_4$(1,2-O$_3$PC$_8$H$_8$PO$_3$)]·H$_2$O (**13**·H$_2$O).

Theta range for data collection	1.06 to 25.68°	
Index ranges	-10<=h<=10, -25<=k<=24, -25<=l<=25	
Reflections collected	41328	
Independent reflections	13390 [R(int) = 0.0405]	
Coverage of independent reflections	99.7%	
Absorption correction	multi-scan	
Max. and min. transmission	0.9270 and 0.5420	
Structure solution technique	direct methods	
Structure solution program	SHELXL-2013 (Sheldrick, 2013)	
Refinement method	Full-matrix least-squares on F^2	
Refinement program	SHELXL-2013 (Sheldrick, 2013)	
Function minimized	$\Sigma\, w(F_o^2 - F_c^2)^2$	
Data / restraints / parameters	13390 / 344 / 1018	
Goodness-of-fit on F^2	1.035	
Δ/σ_{max}	0.013	
Final R indices	10415 data; I>2σ(I)	R1 = 0.0325, wR2 = 0.0634
	all data	R1 = 0.0507, wR2 = 0.0678
Weighting scheme	$w=1/[\sigma^2(F_o^2)+(0.0291P)^2+0.8323P]$ where $P=(F_o^2+2F_c^2)/3$	
Largest diff. peak and hole	0.889 and -1.014 eÅ^{-3}	
R.M.S. deviation from mean	0.107 eÅ^{-3}	

Table S97. Atomic coordinates and equivalent isotropic atomic displacement parameters ($Å^2$) for $[\{Cu(terpy)\}_2(H_2O)(OH)Mo_2F_4O_3(1,2\text{-}O_3PC_8H_8PO_3)][\{Cu(terpy)(H_2O)\}Mo_2F_3O_4(1,2\text{-}O_3PC_8H_8PO_3)]\cdot H_2O$ (**13**·H_2O).

U(eq) is defined as one third of the trace of the orthogonalized U_{ij} tensor.

	x/a	**y/b**	**z/c**	**U(eq)**
Mo1	0.04966(4)	0.42190(2)	0.30873(2)	0.01314(8)
Mo2	0.97843(4)	0.47270(2)	0.14326(2)	0.01237(8)
Mo3	0.23185(4)	0.05123(2)	0.66722(2)	0.01146(8)
Mo4	0.18973(4)	0.97680(2)	0.81607(2)	0.01179(8)
Cu1	0.52338(5)	0.37916(2)	0.12160(2)	0.00898(10)
Cu2	0.28678(5)	0.75783(2)	0.52027(2)	0.01039(10)
Cu3	0.81654(5)	0.08178(2)	0.82944(2)	0.00903(10)
P1	0.18126(10)	0.57577(5)	0.30002(5)	0.0088(2)
P2	0.74535(10)	0.48499(5)	0.25949(5)	0.00730(19)
P3	0.29569(10)	0.88987(5)	0.65822(5)	0.0090(2)
P4	0.90897(10)	0.97719(5)	0.69566(5)	0.00738(19)
F1	0.0217(2)	0.40977(10)	0.20317(10)	0.0101(4)
F2	0.9594(2)	0.32689(11)	0.26907(11)	0.0185(5)
F3	0.2532(2)	0.04586(10)	0.76782(10)	0.0105(4)
F4	0.1941(3)	0.14178(11)	0.72046(11)	0.0223(5)
F5	0.8145(2)	0.40355(11)	0.09328(11)	0.0190(5)
F6	0.3845(2)	0.00692(12)	0.86552(11)	0.0206(5)
O1	0.0446(3)	0.43830(13)	0.39441(13)	0.0159(6)

	x/a	y/b	z/c	U(eq)
O2	0.2341(3)	0.40543(12)	0.29916(12)	0.0092(5)
O3	0.1051(3)	0.52550(12)	0.32960(12)	0.0101(5)
O4	0.1483(3)	0.54127(12)	0.21933(12)	0.0102(5)
O5	0.1363(3)	0.64848(13)	0.33220(12)	0.0121(5)
O6	0.9569(3)	0.52999(13)	0.10546(12)	0.0122(5)
O7	0.8459(3)	0.51220(12)	0.21797(12)	0.0096(5)
O8	0.8281(3)	0.44287(12)	0.29364(12)	0.0090(5)
O9	0.5948(3)	0.44320(12)	0.21658(12)	0.0097(5)
O10	0.2974(3)	0.35031(14)	0.15383(14)	0.0153(6)
O11	0.2016(3)	0.05196(13)	0.58692(13)	0.0139(6)
O12	0.2344(3)	0.94571(12)	0.63479(12)	0.0097(5)
O13	0.2808(3)	0.90713(13)	0.73446(12)	0.0129(6)
O14	0.2171(3)	0.81560(12)	0.60943(12)	0.0109(5)
O15	0.0063(3)	0.95261(12)	0.74312(12)	0.0114(5)
O16	0.1180(3)	0.05225(12)	0.87517(12)	0.0079(5)
O17	0.1303(3)	0.91363(12)	0.84339(12)	0.0114(5)
O18	0.7893(3)	0.01735(12)	0.73305(12)	0.0098(5)
O19	0.0027(3)	0.02040(12)	0.66584(12)	0.0095(5)
O20	0.5751(3)	0.09398(15)	0.82258(13)	0.0154(6)
O21	0.0819(3)	0.69566(13)	0.45979(12)	0.0119(5)
O22	0.1145(2)	0.42368(11)	0.09550(11)	0.0053(5)

	x/a	y/b	z/c	U(eq)
O23	0.4320(3)	0.07110(13)	0.68685(12)	0.0125(6)
O90	0.9421(3)	0.57953(15)	0.47891(14)	0.0201(6)
N1	0.4492(3)	0.45033(15)	0.08654(15)	0.0095(6)
N2	0.4444(3)	0.31577(15)	0.02468(15)	0.0096(6)
N3	0.5824(3)	0.28560(15)	0.11956(15)	0.0100(6)
N4	0.2811(3)	0.82980(15)	0.47789(15)	0.0111(7)
N5	0.4796(3)	0.75197(15)	0.48550(14)	0.0091(6)
N6	0.3627(3)	0.68308(16)	0.54862(15)	0.0115(7)
N7	0.7985(3)	0.01332(15)	0.87712(15)	0.0110(6)
N8	0.8712(3)	0.14918(15)	0.92613(15)	0.0106(6)
N9	0.8769(3)	0.17262(15)	0.81658(15)	0.0112(7)
C1	0.4475(4)	0.51993(19)	0.12418(19)	0.0123(8)
C2	0.3770(4)	0.5618(2)	0.09699(19)	0.0153(8)
C3	0.3066(4)	0.5313(2)	0.02836(19)	0.0160(8)
C4	0.3074(4)	0.4596(2)	0.98865(19)	0.0129(8)
C5	0.3784(4)	0.42068(19)	0.01909(18)	0.0098(7)
C6	0.3779(4)	0.34255(19)	0.98313(18)	0.0112(8)
C7	0.3140(4)	0.2978(2)	0.91438(19)	0.0158(8)
C8	0.3173(4)	0.2249(2)	0.8914(2)	0.0183(9)
C9	0.3825(4)	0.1980(2)	0.93595(19)	0.0153(8)
C10	0.4484(4)	0.24554(19)	0.00320(18)	0.0107(8)

	x/a	y/b	z/c	U(eq)
C11	0.5260(4)	0.22780(19)	0.05827(19)	0.0109(8)
C12	0.5383(4)	0.15913(19)	0.0506(2)	0.0135(8)
C13	0.6088(4)	0.1490(2)	0.1064(2)	0.0152(8)
C14	0.6702(4)	0.20748(19)	0.1678(2)	0.0143(8)
C15	0.6545(4)	0.2751(2)	0.17203(19)	0.0131(8)
C16	0.3814(4)	0.57793(19)	0.32333(19)	0.0112(8)
C17	0.4925(4)	0.62524(18)	0.30376(18)	0.0097(8)
C18	0.4451(4)	0.67689(18)	0.28309(18)	0.0117(8)
C19	0.5443(4)	0.71900(18)	0.26351(18)	0.0124(8)
C20	0.6948(4)	0.70894(19)	0.26437(18)	0.0118(8)
C21	0.7443(4)	0.65846(18)	0.28566(18)	0.0119(8)
C22	0.6450(4)	0.61644(18)	0.30604(17)	0.0083(7)
C23	0.7113(4)	0.56427(18)	0.32992(18)	0.0084(7)
C24	0.2918(4)	0.6497(2)	0.58250(19)	0.0145(8)
C25	0.3516(4)	0.5982(2)	0.5989(2)	0.0173(9)
C26	0.4901(4)	0.5804(2)	0.5805(2)	0.0180(9)
C27	0.5651(4)	0.6153(2)	0.54623(19)	0.0164(8)
C28	0.4992(4)	0.66669(18)	0.53156(18)	0.0104(8)
C29	0.5700(4)	0.70947(19)	0.49751(18)	0.0105(8)
C30	0.7146(4)	0.70977(19)	0.48078(18)	0.0120(8)
C31	0.7619(4)	0.75610(19)	0.45103(18)	0.0133(8)

	x/a	y/b	z/c	U(eq)
C32	0.6672(4)	0.80046(19)	0.43944(18)	0.0117(8)
C33	0.5230(4)	0.79703(18)	0.45724(17)	0.0094(7)
C34	0.4074(4)	0.84090(19)	0.45125(18)	0.0112(8)
C35	0.4214(4)	0.88931(19)	0.42135(19)	0.0150(8)
C36	0.3029(4)	0.9267(2)	0.41824(19)	0.0181(9)
C37	0.1748(4)	0.9153(2)	0.44501(19)	0.0164(8)
C38	0.1686(4)	0.86716(19)	0.47496(18)	0.0133(8)
C39	0.7634(4)	0.94207(19)	0.84633(19)	0.0122(8)
C40	0.7606(4)	0.9009(2)	0.88442(19)	0.0146(8)
C41	0.7906(4)	0.9345(2)	0.95624(19)	0.0152(8)
C42	0.8261(4)	0.0085(2)	0.98884(19)	0.0151(8)
C43	0.8307(4)	0.04665(19)	0.94773(18)	0.0108(8)
C44	0.8754(4)	0.12440(19)	0.97574(18)	0.0113(8)
C45	0.9199(4)	0.1717(2)	0.04592(19)	0.0166(8)
C46	0.9572(4)	0.2442(2)	0.0622(2)	0.0207(9)
C47	0.9535(4)	0.2681(2)	0.0094(2)	0.0196(9)
C48	0.9096(4)	0.21845(19)	0.94068(19)	0.0139(8)
C49	0.9039(4)	0.23258(19)	0.87712(19)	0.0130(8)
C50	0.9230(4)	0.2999(2)	0.8768(2)	0.0179(9)
C51	0.9168(4)	0.3055(2)	0.8134(2)	0.0213(9)
C52	0.8943(4)	0.2441(2)	0.7522(2)	0.0180(9)

	x/a	y/b	z/c	U(eq)
C53	0.8745(4)	0.1785(2)	0.7561(2)	0.0140(8)
C54	0.4941(4)	0.89960(19)	0.65216(18)	0.0101(8)
C55	0.5884(4)	0.84823(18)	0.66617(18)	0.0089(7)
C56	0.5331(4)	0.80321(19)	0.69626(18)	0.0116(8)
C57	0.6212(4)	0.75882(19)	0.71220(19)	0.0157(8)
C58	0.7678(4)	0.75761(19)	0.6977(2)	0.0163(8)
C59	0.8247(4)	0.80087(19)	0.66749(19)	0.0149(8)
C60	0.7370(4)	0.84640(18)	0.65178(18)	0.0108(8)
C61	0.8140(4)	0.89593(18)	0.62431(18)	0.0097(8)

Table S98. Bond lengths (Å) for [{Cu(terpy)}$_2$(H_2O)(OH)$Mo_2F_4O_3$(1,2-$O_3PC_8H_8PO_3$)][{Cu(terpy)(H_2O)}$Mo_2F_3O_4$(1,2-$O_3PC_8H_8PO_3$)]·H_2O (**13**·H_2O).

Mo1-O1	1.712(2)	Mo1-O2	1.744(2)
Mo1-F2	1.858(2)	Mo1-O3	2.006(2)
Mo1-O8	2.100(2)	Mo1-F1	2.1294(19)
Mo2-O6	1.689(2)	Mo2-O22	1.799(2)
Mo2-F5	1.840(2)	Mo2-O7	2.036(2)
Mo2-O4	2.041(2)	Mo2-F1	2.1844(19)
Mo3-O11	1.698(2)	Mo3-O23	1.774(2)
Mo3-F4	1.847(2)	Mo3-O12	2.013(2)
Mo3-O19	2.092(2)	Mo3-F3	2.1650(19)
Mo4-O17	1.692(2)	Mo4-O16	1.796(2)
Mo4-F6	1.852(2)	Mo4-O15	2.010(2)
Mo4-O13	2.067(2)	Mo4-F3	2.1309(19)
Cu1-O9	1.907(2)	Cu1-N2	1.946(3)
Cu1-N1	2.032(3)	Cu1-N3	2.043(3)
Cu1-O10	2.310(3)	Cu2-N5	1.951(3)
Cu2-O14	1.994(2)	Cu2-N6	2.020(3)
Cu2-N4	2.024(3)	Cu2-O21	2.100(2)
Cu3-O18	1.911(2)	Cu3-N8	1.934(3)
Cu3-N9	2.035(3)	Cu3-N7	2.045(3)
Cu3-O20	2.208(3)	P1-O5	1.500(2)

P1-O4	1.546(2)	P1-O3	1.554(2)
P1-C16	1.808(4)	P2-O9	1.513(2)
P2-O8	1.534(2)	P2-O7	1.540(2)
P2-C23	1.801(3)	P3-O14	1.513(3)
P3-O13	1.530(2)	P3-O12	1.553(2)
P3-C54	1.801(3)	P4-O18	1.518(2)
P4-O19	1.525(2)	P4-O15	1.531(3)
P4-C61	1.803(4)	F1-Mo2	2.1844(19)
F3-Mo3	2.1650(19)	O4-Mo2	2.041(2)
O8-Mo1	2.100(2)	O10-H10C	0.807(19)
O10-H10D	0.824(18)	O13-Mo4	2.067(2)
O19-Mo3	2.092(2)	O20-H20C	0.838(19)
O20-H20D	0.814(19)	O90-H90A	0.817(19)
O90-H90B	0.826(19)	N1-C1	1.341(4)
N1-C5	1.360(4)	N2-C6	1.335(5)
N2-C10	1.344(4)	N3-C15	1.330(5)
N3-C11	1.368(5)	N4-C38	1.344(4)
N4-C34	1.360(4)	N5-C29	1.333(4)
N5-C33	1.343(4)	N6-C24	1.344(4)
N6-C28	1.353(4)	N7-C39	1.340(4)
N7-C43	1.355(4)	N8-C44	1.339(5)
N8-C48	1.340(5)	N9-C53	1.332(5)

N9-C49	1.357(4)	C1-C2	1.384(5)
C1-H1	0.95	C2-C3	1.379(5)
C2-H2	0.95	C3-C4	1.384(5)
C3-H3	0.95	C4-C5	1.378(5)
C4-H4	0.95	C5-C6	1.485(5)
C6-C7	1.394(5)	C7-C8	1.392(5)
C7-H7	0.95	C8-C9	1.382(5)
C8-H8	0.95	C9-C10	1.390(5)
C9-H9	0.95	C10-C11	1.483(5)
C11-C12	1.382(5)	C12-C13	1.380(5)
C12-H12	0.95	C13-C14	1.385(5)
C13-H13	0.95	C14-C15	1.387(5)
C14-H14	0.95	C15-H15	0.95
C16-C17	1.532(5)	C16-H16A	0.99
C16-H16B	0.99	C17-C18	1.392(5)
C17-C22	1.401(5)	C18-C19	1.393(5)
C18-H18	0.95	C19-C20	1.390(5)
C19-H19	0.95	C20-C21	1.385(5)
C20-H20	0.95	C21-C22	1.403(5)
C21-H21	0.95	C22-C23	1.508(5)
C23-H23A	0.99	C23-H23B	0.99
C24-C25	1.383(5)	C24-H24	0.95

C25-C26	1.385(5)	C25-H25	0.95
C26-C27	1.389(5)	C26-H26	0.95
C27-C28	1.389(5)	C27-H27	0.95
C28-C29	1.485(5)	C29-C30	1.384(5)
C30-C31	1.399(5)	C30-H30	0.95
C31-C32	1.385(5)	C31-H31	0.95
C32-C33	1.392(5)	C32-H32	0.95
C33-C34	1.474(5)	C34-C35	1.385(5)
C35-C36	1.388(5)	C35-H35	0.95
C36-C37	1.377(5)	C36-H36	0.95
C37-C38	1.378(5)	C37-H37	0.95
C38-H38	0.95	C39-C40	1.388(5)
C39-H39	0.95	C40-C41	1.377(5)
C40-H40	0.95	C41-C42	1.391(5)
C41-H41	0.95	C42-C43	1.391(5)
C42-H42	0.95	C43-C44	1.470(5)
C44-C45	1.399(5)	C45-C46	1.395(5)
C45-H45	0.95	C46-C47	1.389(5)
C46-H46	0.95	C47-C48	1.390(5)
C47-H47	0.95	C48-C49	1.483(5)
C49-C50	1.389(5)	C50-C51	1.388(5)
C50-H50	0.95	C51-C52	1.381(5)

C51-H51	0.95	C52-C53	1.389(5)
C52-H52	0.95	C53-H53	0.95
C54-C55	1.521(5)	C54-H54A	0.99
C54-H54B	0.99	C55-C56	1.404(5)
C55-C60	1.403(5)	C56-C57	1.386(5)
C56-H56	0.95	C57-C58	1.385(5)
C57-H57	0.95	C58-C59	1.383(5)
C58-H58	0.95	C59-C60	1.401(5)
C59-H59	0.95	C60-C61	1.514(5)
C61-H61A	0.99	C61-H61B	0.99

Table S99. Bond angles (°) for [{Cu(terpy)}$_2$(H_2O)(OH)$Mo_2F_4O_3$(1,2-$O_3PC_8H_8PO_3$)][{Cu(terpy)(H_2O)}$Mo_2F_3O_4$(1,2-$O_3PC_8H_8PO_3$)]·H_2O (**13**·H_2O).

O1-Mo1-O2	102.77(12)	O1-Mo1-F2	97.97(11)
O2-Mo1-F2	96.36(10)	O1-Mo1-O3	93.51(11)
O2-Mo1-O3	92.59(10)	F2-Mo1-O3	163.50(10)
O1-Mo1-O8	92.59(11)	O2-Mo1-O8	164.31(10)
F2-Mo1-O8	84.50(9)	O3-Mo1-O8	83.18(9)
O1-Mo1-F1	170.17(10)	O2-Mo1-F1	86.67(9)
F2-Mo1-F1	83.50(8)	O3-Mo1-F1	83.21(8)
O8-Mo1-F1	77.84(8)	O6-Mo2-O22	100.54(10)
O6-Mo2-F5	99.70(11)	O22-Mo2-F5	95.02(10)
O6-Mo2-O7	97.81(10)	O22-Mo2-O7	161.07(10)
F5-Mo2-O7	86.43(9)	O6-Mo2-O4	93.31(11)
O22-Mo2-O4	90.49(10)	F5-Mo2-O4	164.68(9)
O7-Mo2-O4	83.81(9)	O6-Mo2-F1	173.02(10)
O22-Mo2-F1	81.55(8)	F5-Mo2-F1	86.70(9)
O7-Mo2-F1	79.69(8)	O4-Mo2-F1	79.97(8)
O11-Mo3-O23	100.86(11)	O11-Mo3-F4	99.26(11)
O23-Mo3-F4	96.79(11)	O11-Mo3-O12	97.18(11)
O23-Mo3-O12	92.43(10)	F4-Mo3-O12	159.26(9)
O11-Mo3-O19	93.53(10)	O23-Mo3-O19	165.16(10)
F4-Mo3-O19	84.30(9)	O12-Mo3-O19	82.15(9)

O11-Mo3-F3	175.64(10)	O23-Mo3-F3	83.28(9)
F4-Mo3-F3	81.46(8)	O12-Mo3-F3	81.22(8)
O19-Mo3-F3	82.25(8)	O17-Mo4-O16	100.81(11)
O17-Mo4-F6	99.84(11)	O16-Mo4-F6	93.54(10)
O17-Mo4-O15	94.56(11)	O16-Mo4-O15	91.53(10)
F6-Mo4-O15	163.55(9)	O17-Mo4-O13	93.94(10)
O16-Mo4-O13	164.89(10)	F6-Mo4-O13	87.12(10)
O15-Mo4-O13	83.97(10)	O17-Mo4-F3	172.30(10)
O16-Mo4-F3	84.51(9)	F6-Mo4-F3	85.27(9)
O15-Mo4-F3	79.65(8)	O13-Mo4-F3	80.49(8)
O9-Cu1-N2	177.89(11)	O9-Cu1-N1	98.50(11)
N2-Cu1-N1	79.82(12)	O9-Cu1-N3	101.68(11)
N2-Cu1-N3	80.08(12)	N1-Cu1-N3	159.55(12)
O9-Cu1-O10	88.97(10)	N2-Cu1-O10	89.94(11)
N1-Cu1-O10	95.67(10)	N3-Cu1-O10	88.04(10)
N5-Cu2-O14	135.71(11)	N5-Cu2-N6	79.48(12)
O14-Cu2-N6	97.66(11)	N5-Cu2-N4	79.90(12)
O14-Cu2-N4	97.50(11)	N6-Cu2-N4	159.37(11)
N5-Cu2-O21	123.37(11)	O14-Cu2-O21	100.91(9)
N6-Cu2-O21	96.82(11)	N4-Cu2-O21	93.96(11)
O18-Cu3-N8	172.65(11)	O18-Cu3-N9	96.92(11)
N8-Cu3-N9	80.11(12)	O18-Cu3-N7	101.97(11)

N8-Cu3-N7	80.13(12)	N9-Cu3-N7	159.36(12)
O18-Cu3-O20	91.79(10)	N8-Cu3-O20	95.04(11)
N9-Cu3-O20	92.50(11)	N7-Cu3-O20	95.12(11)
O5-P1-O4	113.68(14)	O5-P1-O3	110.56(13)
O4-P1-O3	110.40(13)	O5-P1-C16	111.81(16)
O4-P1-C16	105.95(15)	O3-P1-C16	103.92(15)
O9-P2-O8	111.63(14)	O9-P2-O7	112.77(14)
O8-P2-O7	111.97(13)	O9-P2-C23	108.58(15)
O8-P2-C23	106.32(15)	O7-P2-C23	105.08(14)
O14-P3-O13	112.03(14)	O14-P3-O12	109.96(14)
O13-P3-O12	111.37(13)	O14-P3-C54	110.77(15)
O13-P3-C54	108.04(16)	O12-P3-C54	104.39(15)
O18-P4-O19	111.66(13)	O18-P4-O15	110.85(14)
O19-P4-O15	112.78(14)	O18-P4-C61	108.00(15)
O19-P4-C61	108.32(15)	O15-P4-C61	104.86(15)
Mo1-F1-Mo2	139.47(9)	Mo4-F3-Mo3	141.56(10)
P1-O3-Mo1	141.91(14)	P1-O4-Mo2	137.07(13)
P2-O7-Mo2	138.42(15)	P2-O8-Mo1	136.97(13)
P2-O9-Cu1	136.25(14)	Cu1-O10-H10C	112.(3)
Cu1-O10-H10D	109.(3)	H10C-O10-H10D	107.(4)
P3-O12-Mo3	142.24(15)	P3-O13-Mo4	140.65(15)
P3-O14-Cu2	122.61(13)	P4-O15-Mo4	146.80(15)

P4-O18-Cu3	125.65(15)	P4-O19-Mo3	137.29(15)
Cu3-O20-H20C	116.(3)	Cu3-O20-H20D	119.(3)
H20C-O20-H20D	96.(4)	H90A-O90-H90B	108.(4)
C1-N1-C5	117.9(3)	C1-N1-Cu1	127.3(3)
C5-N1-Cu1	114.4(2)	C6-N2-C10	122.0(3)
C6-N2-Cu1	119.0(2)	C10-N2-Cu1	118.8(2)
C15-N3-C11	118.8(3)	C15-N3-Cu1	127.4(2)
C11-N3-Cu1	113.5(2)	C38-N4-C34	118.6(3)
C38-N4-Cu2	126.8(2)	C34-N4-Cu2	114.7(2)
C29-N5-C33	122.4(3)	C29-N5-Cu2	119.0(2)
C33-N5-Cu2	118.2(2)	C24-N6-C28	118.4(3)
C24-N6-Cu2	126.4(3)	C28-N6-Cu2	115.2(2)
C39-N7-C43	119.3(3)	C39-N7-Cu3	127.4(2)
C43-N7-Cu3	113.3(2)	C44-N8-C48	122.5(3)
C44-N8-Cu3	118.7(2)	C48-N8-Cu3	118.7(2)
C53-N9-C49	119.3(3)	C53-N9-Cu3	126.5(2)
C49-N9-Cu3	113.7(2)	N1-C1-C2	122.6(4)
N1-C1-H1	118.7	C2-C1-H1	118.7
C3-C2-C1	119.0(3)	C3-C2-H2	120.5
C1-C2-H2	120.5	C2-C3-C4	119.2(4)
C2-C3-H3	120.4	C4-C3-H3	120.4
C5-C4-C3	118.8(4)	C5-C4-H4	120.6

C3-C4-H4	120.6	N1-C5-C4	122.4(3)
N1-C5-C6	113.9(3)	C4-C5-C6	123.6(3)
N2-C6-C7	120.3(3)	N2-C6-C5	112.8(3)
C7-C6-C5	126.9(3)	C8-C7-C6	118.3(4)
C8-C7-H7	120.8	C6-C7-H7	120.8
C9-C8-C7	120.5(4)	C9-C8-H8	119.8
C7-C8-H8	119.8	C8-C9-C10	118.4(3)
C8-C9-H9	120.8	C10-C9-H9	120.8
N2-C10-C9	120.4(3)	N2-C10-C11	112.7(3)
C9-C10-C11	126.9(3)	N3-C11-C12	121.5(3)
N3-C11-C10	114.4(3)	C12-C11-C10	124.1(3)
C13-C12-C11	119.0(3)	C13-C12-H12	120.5
C11-C12-H12	120.5	C12-C13-C14	119.7(3)
C12-C13-H13	120.2	C14-C13-H13	120.2
C13-C14-C15	118.5(4)	C13-C14-H14	120.7
C15-C14-H14	120.7	N3-C15-C14	122.5(3)
N3-C15-H15	118.7	C14-C15-H15	118.7
C17-C16-P1	118.1(3)	C17-C16-H16A	107.8
P1-C16-H16A	107.8	C17-C16-H16B	107.8
P1-C16-H16B	107.8	H16A-C16-H16B	107.1
C18-C17-C22	118.8(3)	C18-C17-C16	121.5(3)
C22-C17-C16	119.8(3)	C17-C18-C19	122.0(3)

C17-C18-H18	119.0	C19-C18-H18	119.0
C20-C19-C18	119.0(3)	C20-C19-H19	120.5
C18-C19-H19	120.5	C21-C20-C19	119.8(3)
C21-C20-H20	120.1	C19-C20-H20	120.1
C20-C21-C22	121.3(3)	C20-C21-H21	119.3
C22-C21-H21	119.3	C21-C22-C17	119.1(3)
C21-C22-C23	117.0(3)	C17-C22-C23	123.9(3)
C22-C23-P2	113.8(2)	C22-C23-H23A	108.8
P2-C23-H23A	108.8	C22-C23-H23B	108.8
P2-C23-H23B	108.8	H23A-C23-H23B	107.7
N6-C24-C25	122.4(3)	N6-C24-H24	118.8
C25-C24-H24	118.8	C26-C25-C24	119.3(3)
C26-C25-H25	120.3	C24-C25-H25	120.3
C25-C26-C27	118.7(4)	C25-C26-H26	120.6
C27-C26-H26	120.6	C28-C27-C26	119.1(4)
C28-C27-H27	120.5	C26-C27-H27	120.5
N6-C28-C27	122.0(3)	N6-C28-C29	113.6(3)
C27-C28-C29	124.4(3)	N5-C29-C30	120.8(3)
N5-C29-C28	112.5(3)	C30-C29-C28	126.6(3)
C29-C30-C31	117.7(3)	C29-C30-H30	121.1
C31-C30-H30	121.1	C32-C31-C30	120.8(3)
C32-C31-H31	119.6	C30-C31-H31	119.6

C31-C32-C33	118.3(3)	C31-C32-H32	120.8
C33-C32-H32	120.8	N5-C33-C32	119.9(3)
N5-C33-C34	113.2(3)	C32-C33-C34	126.8(3)
N4-C34-C35	121.5(3)	N4-C34-C33	113.9(3)
C35-C34-C33	124.7(3)	C36-C35-C34	118.8(3)
C36-C35-H35	120.6	C34-C35-H35	120.6
C37-C36-C35	119.7(3)	C37-C36-H36	120.1
C35-C36-H36	120.1	C36-C37-C38	118.7(3)
C36-C37-H37	120.7	C38-C37-H37	120.7
N4-C38-C37	122.7(3)	N4-C38-H38	118.6
C37-C38-H38	118.6	N7-C39-C40	122.1(3)
N7-C39-H39	118.9	C40-C39-H39	118.9
C41-C40-C39	118.9(3)	C41-C40-H40	120.5
C39-C40-H40	120.5	C40-C41-C42	119.5(4)
C40-C41-H41	120.3	C42-C41-H41	120.3
C43-C42-C41	118.8(3)	C43-C42-H42	120.6
C41-C42-H42	120.6	N7-C43-C42	121.3(3)
N7-C43-C44	114.7(3)	C42-C43-C44	124.0(3)
N8-C44-C45	119.9(3)	N8-C44-C43	113.2(3)
C45-C44-C43	127.0(3)	C46-C45-C44	118.4(4)
C46-C45-H45	120.8	C44-C45-H45	120.8
C47-C46-C45	120.3(4)	C47-C46-H46	119.9

C45-C46-H46	119.9	C46-C47-C48	118.5(4)
C46-C47-H47	120.7	C48-C47-H47	120.7
N8-C48-C47	120.3(4)	N8-C48-C49	112.8(3)
C47-C48-C49	126.8(3)	N9-C49-C50	121.1(3)
N9-C49-C48	113.8(3)	C50-C49-C48	125.1(3)
C51-C50-C49	119.1(4)	C51-C50-H50	120.4
C49-C50-H50	120.4	C52-C51-C50	119.3(4)
C52-C51-H51	120.3	C50-C51-H51	120.3
C51-C52-C53	118.7(4)	C51-C52-H52	120.7
C53-C52-H52	120.7	N9-C53-C52	122.4(4)
N9-C53-H53	118.8	C52-C53-H53	118.8
C55-C54-P3	118.6(2)	C55-C54-H54A	107.7
P3-C54-H54A	107.7	C55-C54-H54B	107.7
P3-C54-H54B	107.7	H54A-C54-H54B	107.1
C56-C55-C60	117.6(3)	C56-C55-C54	121.7(3)
C60-C55-C54	120.6(3)	C57-C56-C55	121.8(3)
C57-C56-H56	119.1	C55-C56-H56	119.1
C56-C57-C58	119.9(3)	C56-C57-H57	120.1
C58-C57-H57	120.1	C59-C58-C57	119.5(3)
C59-C58-H58	120.2	C57-C58-H58	120.2
C58-C59-C60	121.0(3)	C58-C59-H59	119.5
C60-C59-H59	119.5	C59-C60-C55	120.1(3)

C59-C60-C61	116.9(3)	C55-C60-C61	122.9(3)
C60-C61-P4	110.4(2)	C60-C61-H61A	109.6
P4-C61-H61A	109.6	C60-C61-H61B	109.6
P4-C61-H61B	109.6	H61A-C61-H61B	108.1

Table S100. Torsion angles (°) for [{Cu(terpy)}$_2$(H_2O)(OH)$Mo_2F_4O_3$(1,2-$O_3PC_8H_8PO_3$)][{Cu(terpy)(H_2O)}$Mo_2F_3O_4$(1,2-$O_3PC_8H_8PO_3$)]·H_2O (**13**·H_2O).

O5-P1-O3-Mo1	160.4(2)	O4-P1-O3-Mo1	33.8(3)
C16-P1-O3-Mo1	-79.5(3)	O5-P1-O4-Mo2	-94.4(2)
O3-P1-O4-Mo2	30.5(3)	C16-P1-O4-Mo2	142.4(2)
O9-P2-O7-Mo2	-76.0(2)	O8-P2-O7-Mo2	50.9(3)
C23-P2-O7-Mo2	165.9(2)	O9-P2-O8-Mo1	136.31(19)
O7-P2-O8-Mo1	8.8(3)	C23-P2-O8-Mo1	-105.4(2)
O8-P2-O9-Cu1	-92.0(2)	O7-P2-O9-Cu1	35.1(3)
C23-P2-O9-Cu1	151.1(2)	O14-P3-O12-Mo3	166.4(2)
O13-P3-O12-Mo3	41.6(3)	C54-P3-O12-Mo3	-74.7(3)
O14-P3-O13-Mo4	-117.1(2)	O12-P3-O13-Mo4	6.5(3)
C54-P3-O13-Mo4	120.6(2)	O13-P3-O14-Cu2	-150.25(15)
O12-P3-O14-Cu2	85.34(18)	C54-P3-O14-Cu2	-29.5(2)
O18-P4-O15-Mo4	-88.1(3)	O19-P4-O15-Mo4	38.0(3)
C61-P4-O15-Mo4	155.6(3)	O19-P4-O18-Cu3	-88.76(19)
O15-P4-O18-Cu3	37.9(2)	C61-P4-O18-Cu3	152.25(17)
O18-P4-O19-Mo3	130.0(2)	O15-P4-O19-Mo3	4.4(3)
C61-P4-O19-Mo3	-111.2(2)	C5-N1-C1-C2	0.1(5)
Cu1-N1-C1-C2	-171.6(3)	N1-C1-C2-C3	-0.6(5)
C1-C2-C3-C4	0.5(5)	C2-C3-C4-C5	0.2(5)
C1-N1-C5-C4	0.6(5)	Cu1-N1-C5-C4	173.3(3)

C1-N1-C5-C6	-176.3(3)	Cu1-N1-C5-C6	-3.5(4)
C3-C4-C5-N1	-0.7(5)	C3-C4-C5-C6	175.8(3)
C10-N2-C6-C7	-2.1(5)	Cu1-N2-C6-C7	-177.0(2)
C10-N2-C6-C5	176.3(3)	Cu1-N2-C6-C5	1.4(4)
N1-C5-C6-N2	1.5(4)	C4-C5-C6-N2	-175.3(3)
N1-C5-C6-C7	179.8(3)	C4-C5-C6-C7	3.0(6)
N2-C6-C7-C8	2.0(5)	C5-C6-C7-C8	-176.2(3)
C6-C7-C8-C9	0.0(5)	C7-C8-C9-C10	-2.0(5)
C6-N2-C10-C9	0.1(5)	Cu1-N2-C10-C9	175.0(2)
C6-N2-C10-C11	-179.2(3)	Cu1-N2-C10-C11	-4.3(4)
C8-C9-C10-N2	2.0(5)	C8-C9-C10-C11	-178.9(3)
C15-N3-C11-C12	2.0(5)	Cu1-N3-C11-C12	-171.8(3)
C15-N3-C11-C10	-179.8(3)	Cu1-N3-C11-C10	6.4(3)
N2-C10-C11-N3	-1.7(4)	C9-C10-C11-N3	179.1(3)
N2-C10-C11-C12	176.5(3)	C9-C10-C11-C12	-2.7(6)
N3-C11-C12-C13	0.2(5)	C10-C11-C12-C13	-177.8(3)
C11-C12-C13-C14	-2.2(5)	C12-C13-C14-C15	1.9(5)
C11-N3-C15-C14	-2.3(5)	Cu1-N3-C15-C14	170.6(2)
C13-C14-C15-N3	0.3(5)	O5-P1-C16-C17	-58.9(3)
O4-P1-C16-C17	65.5(3)	O3-P1-C16-C17	-178.2(2)
P1-C16-C17-C18	14.1(4)	P1-C16-C17-C22	-165.4(3)
C22-C17-C18-C19	1.2(5)	C16-C17-C18-C19	-178.4(3)

C17-C18-C19-C20	0.3(5)	C18-C19-C20-C21	-1.1(5)
C19-C20-C21-C22	0.5(5)	C20-C21-C22-C17	0.9(5)
C20-C21-C22-C23	-178.6(3)	C18-C17-C22-C21	-1.7(5)
C16-C17-C22-C21	177.8(3)	C18-C17-C22-C23	177.8(3)
C16-C17-C22-C23	-2.7(5)	C21-C22-C23-P2	-77.1(4)
C17-C22-C23-P2	103.4(3)	O9-P2-C23-C22	-65.9(3)
O8-P2-C23-C22	173.8(2)	O7-P2-C23-C22	55.0(3)
C28-N6-C24-C25	-1.6(5)	Cu2-N6-C24-C25	179.0(3)
N6-C24-C25-C26	0.5(6)	C24-C25-C26-C27	0.3(6)
C25-C26-C27-C28	0.0(6)	C24-N6-C28-C27	1.9(5)
Cu2-N6-C28-C27	-178.6(3)	C24-N6-C28-C29	-177.1(3)
Cu2-N6-C28-C29	2.4(4)	C26-C27-C28-N6	-1.1(6)
C26-C27-C28-C29	177.8(3)	C33-N5-C29-C30	-0.3(5)
Cu2-N5-C29-C30	-172.7(3)	C33-N5-C29-C28	177.7(3)
Cu2-N5-C29-C28	5.4(4)	N6-C28-C29-N5	-4.9(4)
C27-C28-C29-N5	176.2(3)	N6-C28-C29-C30	173.0(3)
C27-C28-C29-C30	-5.9(6)	N5-C29-C30-C31	0.0(5)
C28-C29-C30-C31	-177.7(3)	C29-C30-C31-C32	0.6(5)
C30-C31-C32-C33	-0.9(5)	C29-N5-C33-C32	0.0(5)
Cu2-N5-C33-C32	172.4(3)	C29-N5-C33-C34	-177.7(3)
Cu2-N5-C33-C34	-5.3(4)	C31-C32-C33-N5	0.6(5)
C31-C32-C33-C34	177.9(3)	C38-N4-C34-C35	-0.2(5)

Cu2-N4-C34-C35	-179.1(3)	C38-N4-C34-C33	-179.9(3)
Cu2-N4-C34-C33	1.1(4)	N5-C33-C34-N4	2.5(4)
C32-C33-C34-N4	-175.0(3)	N5-C33-C34-C35	-177.2(3)
C32-C33-C34-C35	5.3(6)	N4-C34-C35-C36	-0.5(5)
C33-C34-C35-C36	179.2(3)	C34-C35-C36-C37	0.3(6)
C35-C36-C37-C38	0.6(6)	C34-N4-C38-C37	1.2(5)
Cu2-N4-C38-C37	179.9(3)	C36-C37-C38-N4	-1.4(6)
C43-N7-C39-C40	-0.8(5)	Cu3-N7-C39-C40	177.3(3)
N7-C39-C40-C41	1.8(5)	C39-C40-C41-C42	-1.3(5)
C40-C41-C42-C43	-0.3(5)	C39-N7-C43-C42	-0.9(5)
Cu3-N7-C43-C42	-179.2(3)	C39-N7-C43-C44	176.9(3)
Cu3-N7-C43-C44	-1.4(4)	C41-C42-C43-N7	1.4(5)
C41-C42-C43-C44	-176.2(3)	C48-N8-C44-C45	0.6(5)
Cu3-N8-C44-C45	177.7(3)	C48-N8-C44-C43	-179.1(3)
Cu3-N8-C44-C43	-2.1(4)	N7-C43-C44-N8	2.3(4)
C42-C43-C44-N8	-180.0(3)	N7-C43-C44-C45	-177.5(3)
C42-C43-C44-C45	0.3(6)	N8-C44-C45-C46	0.8(5)
C43-C44-C45-C46	-179.5(3)	C44-C45-C46-C47	-1.6(5)
C45-C46-C47-C48	1.0(6)	C44-N8-C48-C47	-1.2(5)
Cu3-N8-C48-C47	-178.2(3)	C44-N8-C48-C49	176.9(3)
Cu3-N8-C48-C49	-0.1(4)	C46-C47-C48-N8	0.4(5)
C46-C47-C48-C49	-177.5(3)	C53-N9-C49-C50	2.6(5)

Cu3-N9-C49-C50	-169.8(3)	C53-N9-C49-C48	-177.6(3)
Cu3-N9-C49-C48	9.9(4)	N8-C48-C49-N9	-6.6(4)
C47-C48-C49-N9	171.4(3)	N8-C48-C49-C50	173.1(3)
C47-C48-C49-C50	-8.9(6)	N9-C49-C50-C51	-1.1(5)
C48-C49-C50-C51	179.2(3)	C49-C50-C51-C52	-1.0(5)
C50-C51-C52-C53	1.7(5)	C49-N9-C53-C52	-2.0(5)
Cu3-N9-C53-C52	169.4(3)	C51-C52-C53-N9	-0.2(5)
O14-P3-C54-C55	-57.0(3)	O13-P3-C54-C55	66.1(3)
O12-P3-C54-C55	-175.3(3)	P3-C54-C55-C56	-12.4(5)
P3-C54-C55-C60	170.2(3)	C60-C55-C56-C57	0.7(5)
C54-C55-C56-C57	-176.8(3)	C55-C56-C57-C58	-0.7(6)
C56-C57-C58-C59	0.0(6)	C57-C58-C59-C60	0.6(6)
C58-C59-C60-C55	-0.5(5)	C58-C59-C60-C61	175.1(3)
C56-C55-C60-C59	-0.1(5)	C54-C55-C60-C59	177.5(3)
C56-C55-C60-C61	-175.4(3)	C54-C55-C60-C61	2.1(5)
C59-C60-C61-P4	-86.5(3)	C55-C60-C61-P4	89.0(4)
O18-P4-C61-C60	-66.9(3)	O19-P4-C61-C60	172.1(2)
O15-P4-C61-C60	51.4(3)		

Table S101. Anisotropic atomic displacement parameters ($Å^2$) for $[\{Cu(terpy)\}_2(H_2O)(OH)Mo_2F_4O_3(1,2\text{-}O_3PC_8H_8PO_3)][\{Cu(terpy)(H_2O)\}Mo_2F_3O_4(1,2\text{-}O_3PC_8H_8PO_3)]\cdot H_2O$ (**13**·H_2O).

The anisotropic atomic displacement factor exponent takes the form: $-2\pi^2[h^2 a^{*2} U_{11} + ... + 2 h k a^* b^* U_{12}]$

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
Mo1	0.01910(19)	0.01424(18)	0.01190(18)	0.00889(14)	0.00571(14)	0.01011(14)
Mo2	0.02128(19)	0.00847(17)	0.00677(17)	0.00242(13)	0.00004(14)	0.00526(14)
Mo3	0.01571(18)	0.00956(17)	0.00962(17)	0.00580(14)	0.00092(13)	-0.00273(13)
Mo4	0.01906(18)	0.00868(17)	0.00644(17)	0.00374(14)	-0.00107(13)	-0.00266(13)
Cu1	0.0110(2)	0.0075(2)	0.0076(2)	0.00236(18)	0.00023(18)	0.00173(18)
Cu2	0.0098(2)	0.0114(2)	0.0127(2)	0.0072(2)	0.00347(18)	0.00241(18)
Cu3	0.0110(2)	0.0068(2)	0.0073(2)	0.00121(18)	0.00136(18)	-0.00026(18)
P1	0.0087(5)	0.0083(5)	0.0082(5)	0.0027(4)	0.0001(4)	0.0005(4)
P2	0.0071(5)	0.0065(5)	0.0088(5)	0.0033(4)	0.0022(4)	0.0018(4)
P3	0.0101(5)	0.0089(5)	0.0093(5)	0.0044(4)	0.0033(4)	0.0030(4)
P4	0.0067(5)	0.0077(5)	0.0070(5)	0.0025(4)	0.0004(4)	0.0005(4)
F1	0.0111(11)	0.0112(11)	0.0091(11)	0.0050(9)	0.0025(8)	0.0028(8)
F2	0.0160(12)	0.0142(12)	0.0265(13)	0.0106(10)	-0.0010(10)	0.0018(9)
F3	0.0099(11)	0.0126(11)	0.0117(11)	0.0081(9)	0.0012(8)	0.0004(8)
F4	0.0307(14)	0.0119(12)	0.0233(13)	0.0071(10)	0.0009(10)	0.0027(10)
F5	0.0152(12)	0.0201(12)	0.0191(12)	0.0059(10)	0.0018(9)	0.0018(9)
F6	0.0186(12)	0.0249(13)	0.0182(12)	0.0101(11)	-0.0007(10)	0.0017(10)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
O1	0.0205(15)	0.0173(14)	0.0149(14)	0.0112(12)	0.0046(11)	0.0041(12)
O2	0.0053(12)	0.0096(13)	0.0137(13)	0.0065(11)	-0.0012(10)	0.0021(10)
O3	0.0132(13)	0.0076(13)	0.0098(13)	0.0040(11)	0.0018(10)	0.0008(10)
O4	0.0097(13)	0.0110(13)	0.0094(13)	0.0049(11)	0.0002(10)	-0.0023(10)
O5	0.0112(13)	0.0116(13)	0.0132(14)	0.0044(11)	0.0032(11)	0.0022(10)
O6	0.0153(14)	0.0121(13)	0.0106(13)	0.0063(11)	0.0009(11)	0.0028(11)
O7	0.0099(13)	0.0112(13)	0.0109(13)	0.0074(11)	0.0025(10)	0.0022(10)
O8	0.0071(13)	0.0106(13)	0.0144(14)	0.0096(11)	0.0033(10)	0.0025(10)
O9	0.0085(13)	0.0120(13)	0.0084(13)	0.0045(11)	-0.0010(10)	0.0021(10)
O10	0.0100(14)	0.0210(16)	0.0163(15)	0.0083(13)	0.0022(12)	0.0061(11)
O11	0.0142(14)	0.0148(14)	0.0153(14)	0.0090(12)	0.0032(11)	0.0015(11)
O12	0.0112(13)	0.0104(13)	0.0084(13)	0.0043(11)	0.0015(10)	0.0048(10)
O13	0.0179(14)	0.0142(14)	0.0102(13)	0.0067(11)	0.0050(11)	0.0081(11)
O14	0.0110(13)	0.0099(13)	0.0103(13)	0.0026(11)	0.0031(10)	0.0006(10)
O15	0.0111(13)	0.0110(13)	0.0129(14)	0.0073(11)	-0.0017(10)	-0.0016(10)
O16	0.0080(13)	0.0062(12)	0.0066(12)	-0.0011(10)	0.0045(10)	0.0012(10)
O17	0.0126(14)	0.0106(13)	0.0129(14)	0.0071(11)	0.0016(11)	0.0013(10)
O18	0.0097(13)	0.0092(13)	0.0080(13)	0.0010(10)	0.0010(10)	0.0016(10)
O19	0.0085(13)	0.0109(13)	0.0112(13)	0.0070(11)	0.0007(10)	0.0019(10)
O20	0.0098(14)	0.0202(16)	0.0126(14)	0.0041(13)	-0.0007(11)	0.0008(11)
O21	0.0079(13)	0.0141(14)	0.0126(14)	0.0044(11)	0.0007(10)	0.0026(10)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
O22	0.0039(12)	0.0053(12)	0.0047(12)	0.0002(10)	0.0012(9)	-0.0006(9)
O23	0.0065(13)	0.0219(15)	0.0125(14)	0.0128(12)	-0.0012(10)	-0.0041(11)
O90	0.0328(17)	0.0161(16)	0.0138(16)	0.0075(13)	0.0057(13)	0.0063(13)
N1	0.0069(16)	0.0123(16)	0.0099(16)	0.0056(13)	0.0005(12)	0.0006(12)
N2	0.0093(16)	0.0106(16)	0.0082(15)	0.0029(13)	0.0019(12)	0.0021(12)
N3	0.0096(16)	0.0096(16)	0.0099(16)	0.0029(13)	0.0025(12)	0.0019(12)
N4	0.0115(16)	0.0107(16)	0.0122(16)	0.0056(13)	0.0029(13)	0.0009(13)
N5	0.0102(16)	0.0086(16)	0.0063(15)	0.0011(13)	0.0006(12)	0.0013(12)
N6	0.0146(17)	0.0114(16)	0.0080(16)	0.0040(13)	-0.0001(13)	0.0014(13)
N7	0.0095(16)	0.0104(16)	0.0103(16)	0.0016(13)	0.0027(12)	-0.0003(12)
N8	0.0076(16)	0.0079(15)	0.0125(16)	0.0002(13)	0.0015(12)	0.0022(12)
N9	0.0100(16)	0.0076(15)	0.0140(17)	0.0017(13)	0.0037(13)	0.0024(12)
C1	0.014(2)	0.0106(19)	0.013(2)	0.0046(16)	0.0029(15)	0.0016(15)
C2	0.018(2)	0.012(2)	0.016(2)	0.0046(17)	0.0053(16)	0.0069(16)
C3	0.015(2)	0.022(2)	0.015(2)	0.0111(18)	0.0041(16)	0.0056(17)
C4	0.011(2)	0.018(2)	0.012(2)	0.0080(16)	0.0017(15)	0.0017(16)
C5	0.0070(18)	0.0137(19)	0.0097(19)	0.0062(15)	0.0025(14)	-0.0008(15)
C6	0.0085(19)	0.0144(19)	0.0104(19)	0.0046(16)	0.0034(15)	0.0013(15)
C7	0.011(2)	0.022(2)	0.0099(19)	0.0037(17)	-0.0009(15)	0.0016(16)
C8	0.016(2)	0.020(2)	0.008(2)	-0.0034(17)	-0.0028(16)	0.0009(17)
C9	0.015(2)	0.012(2)	0.015(2)	0.0009(16)	0.0048(16)	0.0029(16)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
C10	0.0069(19)	0.0108(19)	0.0126(19)	0.0026(15)	0.0037(14)	0.0007(14)
C11	0.0052(18)	0.0127(19)	0.0160(19)	0.0065(16)	0.0033(15)	0.0030(15)
C12	0.0072(19)	0.0096(19)	0.018(2)	0.0004(16)	0.0018(15)	0.0007(15)
C13	0.011(2)	0.011(2)	0.027(2)	0.0091(17)	0.0067(16)	0.0040(15)
C14	0.011(2)	0.017(2)	0.018(2)	0.0101(17)	0.0056(16)	0.0058(16)
C15	0.010(2)	0.014(2)	0.014(2)	0.0044(16)	0.0025(15)	0.0026(15)
C16	0.0104(18)	0.0100(19)	0.0118(19)	0.0035(16)	-0.0002(15)	0.0016(15)
C17	0.0114(19)	0.0077(18)	0.0072(18)	0.0006(15)	0.0007(15)	0.0002(14)
C18	0.0113(19)	0.0097(19)	0.012(2)	0.0027(16)	-0.0009(15)	0.0007(15)
C19	0.021(2)	0.0047(18)	0.0105(19)	0.0018(16)	0.0008(16)	0.0039(15)
C20	0.014(2)	0.0100(19)	0.0109(19)	0.0041(16)	0.0044(15)	-0.0019(15)
C21	0.012(2)	0.0091(19)	0.012(2)	0.0011(16)	0.0034(15)	0.0015(15)
C22	0.0132(19)	0.0049(18)	0.0026(17)	-0.0021(14)	0.0005(14)	-0.0002(14)
C23	0.0061(18)	0.0074(18)	0.0108(19)	0.0031(15)	0.0018(14)	-0.0010(14)
C24	0.012(2)	0.017(2)	0.015(2)	0.0064(17)	0.0040(16)	0.0019(16)
C25	0.020(2)	0.018(2)	0.017(2)	0.0126(18)	-0.0001(17)	-0.0023(17)
C26	0.023(2)	0.015(2)	0.020(2)	0.0119(18)	0.0004(17)	0.0040(17)
C27	0.014(2)	0.017(2)	0.019(2)	0.0099(18)	0.0008(16)	0.0004(16)
C28	0.0125(19)	0.0081(19)	0.0070(19)	0.0005(15)	-0.0024(15)	0.0010(15)
C29	0.0145(19)	0.0098(19)	0.0044(18)	0.0001(15)	0.0002(14)	0.0032(15)
C30	0.014(2)	0.0096(19)	0.0068(19)	-0.0013(15)	-0.0009(15)	0.0005(15)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
C31	0.0106(19)	0.014(2)	0.013(2)	0.0030(16)	0.0033(15)	0.0001(15)
C32	0.0124(19)	0.012(2)	0.0096(19)	0.0036(16)	0.0037(15)	-0.0015(15)
C33	0.0120(19)	0.0072(18)	0.0050(18)	-0.0007(15)	-0.0003(14)	-0.0001(14)
C34	0.015(2)	0.0092(19)	0.0065(18)	0.0008(15)	0.0020(15)	-0.0003(15)
C35	0.018(2)	0.013(2)	0.014(2)	0.0060(17)	0.0033(16)	-0.0018(16)
C36	0.029(2)	0.014(2)	0.013(2)	0.0077(17)	-0.0001(17)	0.0034(17)
C37	0.021(2)	0.016(2)	0.013(2)	0.0059(17)	-0.0018(16)	0.0076(17)
C38	0.012(2)	0.017(2)	0.011(2)	0.0055(16)	0.0000(15)	0.0021(16)
C39	0.0111(19)	0.0111(19)	0.0106(19)	0.0010(16)	0.0017(15)	-0.0006(15)
C40	0.015(2)	0.0100(19)	0.017(2)	0.0041(16)	0.0032(16)	-0.0031(16)
C41	0.012(2)	0.020(2)	0.016(2)	0.0107(18)	0.0035(16)	-0.0004(16)
C42	0.011(2)	0.022(2)	0.0098(19)	0.0052(17)	0.0011(15)	0.0000(16)
C43	0.0079(19)	0.0139(19)	0.0093(19)	0.0031(15)	0.0030(14)	0.0014(15)
C44	0.0055(18)	0.0137(19)	0.0112(19)	0.0010(15)	0.0019(14)	0.0029(15)
C45	0.014(2)	0.022(2)	0.011(2)	0.0035(17)	0.0023(16)	0.0019(17)
C46	0.017(2)	0.018(2)	0.012(2)	-0.0060(17)	-0.0015(17)	-0.0049(17)
C47	0.020(2)	0.014(2)	0.018(2)	0.0005(17)	0.0005(17)	-0.0002(17)
C48	0.010(2)	0.0113(19)	0.016(2)	0.0011(16)	0.0020(15)	0.0025(15)
C49	0.0053(18)	0.0110(19)	0.019(2)	0.0028(16)	0.0004(15)	0.0008(15)
C50	0.014(2)	0.011(2)	0.027(2)	0.0063(18)	-0.0002(17)	0.0021(16)
C51	0.019(2)	0.013(2)	0.033(3)	0.0116(19)	0.0023(19)	0.0020(17)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
C52	0.015(2)	0.020(2)	0.026(2)	0.0154(19)	0.0045(17)	0.0057(17)
C53	0.012(2)	0.013(2)	0.019(2)	0.0070(17)	0.0023(16)	0.0043(16)
C54	0.0105(18)	0.0097(19)	0.0093(19)	0.0042(16)	-0.0010(15)	-0.0008(14)
C55	0.0109(18)	0.0045(18)	0.0065(18)	-0.0012(15)	-0.0040(14)	0.0010(14)
C56	0.0095(19)	0.0110(19)	0.0106(19)	0.0011(16)	0.0007(15)	0.0001(15)
C57	0.021(2)	0.0083(19)	0.018(2)	0.0065(17)	-0.0009(17)	-0.0004(16)
C58	0.017(2)	0.009(2)	0.021(2)	0.0051(17)	-0.0045(17)	0.0026(16)
C59	0.012(2)	0.0086(19)	0.021(2)	0.0029(17)	-0.0005(16)	0.0025(15)
C60	0.0128(19)	0.0057(18)	0.0074(19)	-0.0032(15)	0.0003(15)	0.0002(14)
C61	0.0080(18)	0.0105(19)	0.0077(18)	0.0004(15)	0.0015(14)	0.0033(14)

Table S102. Hydrogen atomic coordinates and isotropic atomic displacement parameters ($Å^2$) for [{Cu(terpy)}$_2$(H_2O)(OH)$Mo_2F_4O_3$(1,2-$O_3PC_8H_8PO_3$)][{Cu(terpy)(H_2O)}$Mo_2F_3O_4$(1,2-$O_3PC_8H_8PO_3$)]·H_2O (**13**·H_2O).

	x/a	y/b	z/c	U(eq)
H10C	0.231(3)	0.369(2)	0.142(2)	0.018
H10D	0.308(4)	0.366(2)	0.1967(10)	0.018
H20C	0.531(4)	1.088(2)	0.7834(13)	0.018
H20D	0.515(4)	1.0649(17)	0.827(2)	0.018
H90A	0.937(5)	0.573(2)	0.5141(15)	0.024
H90B	0.967(5)	0.5436(15)	0.4493(17)	0.024
H1	0.4967	0.5413	0.1714	0.015
H2	0.3770	0.6109	0.1252	0.018
H3	0.2582	0.5592	0.0086	0.019
H4	0.2598	0.4375	-0.0588	0.016
H7	0.2693	0.3166	-0.1160	0.019
H8	0.2744	0.1935	-0.1552	0.022
H9	0.3822	0.1481	-0.0791	0.018
H12	0.4989	0.1195	0.0077	0.016
H13	0.6152	0.1021	0.1027	0.018
H14	0.7218	0.2015	0.2062	0.017
H15	0.6970	0.3153	0.2140	0.016
H16A	0.4034	0.5935	0.3746	0.013
H16B	0.4032	0.5285	0.3014	0.013

	x/a	y/b	z/c	U(eq)
H18	0.3422	0.6836	0.2823	0.014
H19	0.5095	0.7541	0.2498	0.015
H20	0.7635	0.7366	0.2504	0.014
H21	0.8475	0.6521	0.2865	0.014
H23A	0.6414	0.5502	0.3569	0.01
H23B	0.8080	0.5884	0.3617	0.01
H24	0.1972	0.6618	0.5957	0.017
H25	0.2983	0.5753	0.6226	0.021
H26	0.5329	0.5450	0.5911	0.022
H27	0.6603	0.6041	0.5330	0.02
H30	0.7795	0.6796	0.4892	0.014
H31	0.8603	0.7572	0.4386	0.016
H32	0.6998	0.8324	0.4198	0.014
H35	0.5106	0.8968	0.4033	0.018
H36	0.3101	0.9599	0.3977	0.022
H37	0.0924	0.9401	0.4429	0.02
H38	0.0812	0.8601	0.4943	0.016
H39	0.7397	0.9190	0.7968	0.015
H40	0.7382	0.8503	0.8613	0.018
H41	0.7872	0.9073	0.9833	0.018
H42	0.8467	1.0325	1.0383	0.018

	x/a	y/b	z/c	U(eq)
H45	0.9247	1.1548	1.0816	0.02
H46	0.9853	1.2774	1.1096	0.025
H47	0.9804	1.3173	1.0201	0.024
H50	0.9401	1.3415	0.9195	0.021
H51	0.9280	1.3510	0.8121	0.026
H52	0.8923	1.2468	0.7084	0.022
H53	0.8586	1.1362	0.7140	0.017
H54A	0.5033	0.8957	0.6046	0.012
H54B	0.5404	0.9486	0.6852	0.012
H56	0.4324	0.8032	0.7060	0.014
H57	0.5813	0.7293	0.7330	0.019
H58	0.8287	0.7273	0.7085	0.02
H59	0.9249	0.7997	0.6572	0.018
H61A	0.7378	0.9073	0.5946	0.012
H61B	0.8883	0.8720	0.5952	0.012

Table S103. Sample and crystal data for [{$Cu_3(bpy)_2$}$Mo_2F_2O_4(H_2O)_2$(1,3-$O_3PC_8H_8PO_3$)$_2$] (**14**).

Identification code	[{$Cu_3(bpy)_2$}$Mo_2F_2O_4(H_2O)_2$(1,3-$O_3PC_8H_8PO_3$)$_2$]	
Chemical formula	$C_{36}H_{36}Cu_3F_2Mo_2N_4O_{18}P_4$	
Formula weight	1357.07	
Temperature	90(2) K	
Wavelength	0.71073 Å	
Crystal size	0.104 x 0.116 x 0.191 mm	
Crystal system	Monoclinic	
Space group	P21/n	
Unit cell dimensions	a = 8.562(2) Å	α = 90°
	b = 19.809(5) Å	β = 103.447(3)°
	c = 13.219(3) Å	γ = 90°
Volume	2180.5(9) $Å^3$	
Z	2	
Density (calculated)	2.067 g/cm^3	
Absorption coefficient	2.236 mm^{-1}	
F(000)	1346	

Table S104. Data collection and structure refinement for [{$Cu_3(bpy)_2$}$Mo_2F_2O_4(H_2O)_2$(1,3-$O_3PC_8H_8PO_3)_2$] (**14**).

Theta range for data collection	2.06 to 26.02°	
Index ranges	-10<=h<=10, -24<=k<=24, -16<=l<=16	
Reflections collected	15384	
Independent reflections	4293 [R(int) = 0.0682]	
Coverage of independent reflections	99.9%	
Absorption correction	multi-scan	
Max. and min. transmission	0.8010 and 0.6750	
Structure solution technique	direct methods	
Structure solution program	SHELXL-2013 (Sheldrick, 2013)	
Refinement method	Full-matrix least-squares on F^2	
Refinement program	SHELXL-2013 (Sheldrick, 2013)	
Function minimized	$\Sigma\, w(F_o^2 - F_c^2)^2$	
Data / restraints / parameters	4293 / 101 / 320	
Goodness-of-fit on F^2	1.062	
Δ/σ_{max}	0.001	
Final R indices	3616 data; I>2σ(I)	R1 = 0.0485, wR2 = 0.1262
	all data	R1 = 0.0573, wR2 = 0.1302
Weighting scheme	$w=1/[\sigma^2(F_o^2)+(0.0551P)^2+16.0284P]$ where $P=(F_o^2+2F_c^2)/3$	
Largest diff. peak and hole	0.840 and -1.314 eÅ^{-3}	
R.M.S. deviation from mean	0.162 eÅ^{-3}	

Table S105. Atomic coordinates and equivalent isotropic atomic displacement parameters ($Å^2$) for [{$Cu_3(bpy)_2$}$Mo_2F_2O_4(H_2O)_2$(1,3-$O_3PC_8H_8PO_3$)$_2$] (**14**).

U(eq) is defined as one third of the trace of the orthogonalized U_{ij} tensor.

	x/a	**y/b**	**z/c**	**U(eq)**
Mo1	0.59126(6)	0.08461(3)	0.85379(4)	0.01533(15)
Cu1	0.5	0.5	0.5	0.0095(2)
Cu2	0.25524(8)	0.47869(3)	0.80027(5)	0.01162(17)
P1	0.31133(16)	0.39203(7)	0.60006(10)	0.0105(3)
P2	0.19712(16)	0.08524(7)	0.86128(10)	0.0100(3)
F1	0.7890(4)	0.04757(18)	0.8549(2)	0.0200(7)
O1	0.2739(4)	0.4099(2)	0.7026(3)	0.0133(8)
O2	0.1626(4)	0.4092(2)	0.5124(3)	0.0132(8)
O3	0.4635(4)	0.42138(19)	0.5799(3)	0.0123(8)
O4	0.3779(4)	0.1025(2)	0.8844(3)	0.0149(8)
O5	0.1693(4)	0.04957(19)	0.9566(3)	0.0114(8)
O6	0.1425(4)	0.04842(19)	0.7593(3)	0.0127(8)
O7	0.5590(5)	0.9748(2)	0.8987(3)	0.0148(8)
O8	0.4907(4)	0.05269(19)	0.7286(3)	0.0105(8)
O9	0.6335(5)	0.1644(2)	0.8264(3)	0.0189(9)
N1	0.1289(5)	0.4164(2)	0.8675(3)	0.0133(9)
N2	0.2983(5)	0.5213(2)	0.9412(3)	0.0134(9)

	x/a	y/b	z/c	U(eq)
C1	0.0342(7)	0.3675(3)	0.8180(5)	0.0193(12)
C2	0.9397(8)	0.3290(3)	0.8689(5)	0.0218(13)
C3	0.9490(8)	0.3412(3)	0.9722(5)	0.0232(13)
C4	0.0490(7)	0.3915(3)	0.0235(5)	0.0194(12)
C5	0.1362(7)	0.4287(3)	0.9683(4)	0.0148(11)
C6	0.2403(6)	0.4860(3)	0.0119(4)	0.0151(11)
C7	0.2754(7)	0.5045(3)	0.1158(4)	0.0184(12)
C8	0.3702(7)	0.5601(3)	0.1475(4)	0.0214(13)
C9	0.4261(7)	0.5965(3)	0.0749(5)	0.0198(12)
C10	0.3883(7)	0.5756(3)	0.9716(4)	0.0164(12)
C11	0.3306(7)	0.3015(3)	0.5984(5)	0.0164(11)
C12	0.1925(7)	0.2634(3)	0.6231(4)	0.0143(11)
C13	0.0455(7)	0.2600(3)	0.5505(5)	0.0203(13)
C14	0.9167(7)	0.2265(3)	0.5752(5)	0.0235(14)
C15	0.2052(6)	0.2315(3)	0.7191(4)	0.0127(11)
C16	0.0783(7)	0.1985(3)	0.7440(4)	0.0136(11)
C17	0.9317(7)	0.1967(3)	0.6711(5)	0.0186(12)
C18	0.0906(7)	0.1645(3)	0.8479(4)	0.0141(11)

Table S106. Bond lengths (Å) for [{$Cu_3(bpy)_2$}$Mo_2F_2O_4(H_2O)_2$(1,3-$O_3PC_8H_8PO_3)_2$] (**14**).

Mo1-O9	1.680(4)	Mo1-O8	1.794(3)
Mo1-F1	1.842(3)	Mo1-O4	1.992(4)
Mo1-O2	2.047(4)	Mo1-O7	2.289(4)
Cu1-O5	1.944(4)	Cu1-O5	1.944(4)
Cu1-O3	1.947(4)	Cu1-O3	1.947(4)
Cu2-O6	1.902(4)	Cu2-O1	1.909(4)
Cu2-N1	1.983(5)	Cu2-N2	1.999(5)
P1-O3	1.506(4)	P1-O1	1.506(4)
P1-O2	1.547(4)	P1-C11	1.802(6)
P2-O6	1.508(4)	P2-O5	1.511(4)
P2-O4	1.545(4)	P2-C18	1.804(6)
O2-Mo1	2.047(4)	O5-Cu1	1.944(4)
O6-Cu2	1.902(4)	O7-H7C	0.84(2)
O7-H7D	0.84(2)	N1-C1	1.334(8)
N1-C5	1.341(7)	N2-C10	1.330(7)
N2-C6	1.351(7)	C1-C2	1.393(9)
C1-H1	0.95	C2-C3	1.372(9)
C2-H2	0.95	C3-C4	1.384(9)
C3-H3	0.95	C4-C5	1.373(8)
C4-H4	0.95	C5-C6	1.475(8)
C6-C7	1.386(8)	C7-C8	1.374(9)

C7-H7	0.95	C8-C9	1.372(9)
C8-H8	0.95	C9-C10	1.391(8)
C9-H9	0.95	C10-H10	0.95
C11-C12	1.501(8)	C11-H11A	0.99
C11-H11B	0.99	C12-C13	1.395(8)
C12-C15	1.399(8)	C13-C14	1.389(9)
C13-H13	0.95	C14-C17	1.377(8)
C14-H14	0.95	C15-C16	1.372(8)
C15-H15	0.95	C16-C17	1.394(8)
C16-C18	1.510(8)	C17-H17	0.95
C18-H18A	0.99	C18-H18B	0.99

Table S107. Bond angles (°) for [{$Cu_3(bpy)_2$}$Mo_2F_2O_4(H_2O)_2$(1,3-$O_3PC_8H_8PO_3)_2$] (**14**).

O9-Mo1-O8	102.35(18)	O9-Mo1-F1	97.65(18)
O8-Mo1-F1	96.31(15)	O9-Mo1-O4	97.45(18)
O8-Mo1-O4	89.09(16)	F1-Mo1-O4	162.47(16)
O9-Mo1-O2	97.85(18)	O8-Mo1-O2	158.63(16)
F1-Mo1-O2	87.79(15)	O4-Mo1-O2	81.39(15)
O9-Mo1-O7	174.57(17)	O8-Mo1-O7	80.98(16)
F1-Mo1-O7	77.61(15)	O4-Mo1-O7	86.84(15)
O2-Mo1-O7	79.44(15)	O5-Cu1-O5	180.0
O5-Cu1-O3	91.10(16)	O5-Cu1-O3	88.90(16)
O5-Cu1-O3	88.90(16)	O5-Cu1-O3	91.10(16)
O3-Cu1-O3	180.0(2)	O6-Cu2-O1	96.87(17)
O6-Cu2-N1	171.76(18)	O1-Cu2-N1	89.95(18)
O6-Cu2-N2	94.38(18)	O1-Cu2-N2	154.79(17)
N1-Cu2-N2	81.17(19)	O3-P1-O1	116.3(2)
O3-P1-O2	112.2(2)	O1-P1-O2	108.2(2)
O3-P1-C11	107.3(2)	O1-P1-C11	106.5(3)
O2-P1-C11	105.7(2)	O6-P2-O5	116.3(2)
O6-P2-O4	111.9(2)	O5-P2-O4	106.5(2)
O6-P2-C18	106.8(2)	O5-P2-C18	108.3(2)
O4-P2-C18	106.7(2)	P1-O1-Cu2	147.7(3)
P1-O2-Mo1	141.1(2)	P1-O3-Cu1	131.2(2)

P2-O4-Mo1	147.6(3)	P2-O5-Cu1	139.0(2)
P2-O6-Cu2	130.1(2)	Mo1-O7-H7C	124.(4)
Mo1-O7-H7D	107.(4)	H7C-O7-H7D	109.(4)
C1-N1-C5	120.2(5)	C1-N1-Cu2	124.4(4)
C5-N1-Cu2	115.3(4)	C10-N2-C6	119.3(5)
C10-N2-Cu2	126.3(4)	C6-N2-Cu2	114.1(4)
N1-C1-C2	121.0(6)	N1-C1-H1	119.5
C2-C1-H1	119.5	C3-C2-C1	118.6(6)
C3-C2-H2	120.7	C1-C2-H2	120.7
C2-C3-C4	120.1(6)	C2-C3-H3	120.0
C4-C3-H3	120.0	C5-C4-C3	118.5(6)
C5-C4-H4	120.7	C3-C4-H4	120.7
N1-C5-C4	121.6(6)	N1-C5-C6	114.3(5)
C4-C5-C6	124.0(5)	N2-C6-C7	121.3(6)
N2-C6-C5	114.5(5)	C7-C6-C5	124.2(5)
C8-C7-C6	119.4(6)	C8-C7-H7	120.3
C6-C7-H7	120.3	C9-C8-C7	119.0(5)
C9-C8-H8	120.5	C7-C8-H8	120.5
C8-C9-C10	119.4(6)	C8-C9-H9	120.3
C10-C9-H9	120.3	N2-C10-C9	121.6(6)
N2-C10-H10	119.2	C9-C10-H10	119.2
C12-C11-P1	114.8(4)	C12-C11-H11A	108.6

P1-C11-H11A	108.6	C12-C11-H11B	108.6
P1-C11-H11B	108.6	H11A-C11-H11B	107.5
C13-C12-C15	118.0(5)	C13-C12-C11	120.3(5)
C15-C12-C11	121.7(5)	C14-C13-C12	120.0(5)
C14-C13-H13	120.0	C12-C13-H13	120.0
C17-C14-C13	120.8(5)	C17-C14-H14	119.6
C13-C14-H14	119.6	C16-C15-C12	122.3(5)
C16-C15-H15	118.9	C12-C15-H15	118.9
C15-C16-C17	118.8(5)	C15-C16-C18	122.6(5)
C17-C16-C18	118.6(5)	C14-C17-C16	120.1(6)
C14-C17-H17	119.9	C16-C17-H17	119.9
C16-C18-P2	113.8(4)	C16-C18-H18A	108.8
P2-C18-H18A	108.8	C16-C18-H18B	108.8
P2-C18-H18B	108.8	H18A-C18-H18B	107.7

Table S108. Anisotropic atomic displacement parameters ($Å^2$) for $[\{Cu_3(bpy)_2\}Mo_2F_2O_4(H_2O)_2(1,3\text{-}O_3PC_8H_8PO_3)_2]$ (**14**).

The anisotropic atomic displacement factor exponent takes the form: $-2\pi^2[h^2 a^{*2} U_{11} + ... + 2 h k a^* b^* U_{12}]$

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
Mo1	0.0201(3)	0.0162(3)	0.0103(3)	0.00119(19)	0.00481(19)	0.0009(2)
Cu1	0.0079(4)	0.0123(5)	0.0071(4)	0.0002(3)	-0.0006(3)	-0.0003(3)
Cu2	0.0125(3)	0.0152(4)	0.0058(3)	0.0003(3)	-0.0007(2)	-0.0011(3)
P1	0.0096(6)	0.0131(7)	0.0070(6)	0.0004(5)	-0.0018(5)	-0.0009(5)
P2	0.0091(6)	0.0130(7)	0.0063(6)	0.0007(5)	-0.0015(5)	-0.0001(5)
F1	0.0153(16)	0.0290(19)	0.0131(16)	-0.0009(14)	-0.0023(13)	0.0002(14)
O1	0.0137(19)	0.017(2)	0.0077(17)	-0.0016(15)	-0.0011(14)	-0.0025(16)
O2	0.0109(18)	0.019(2)	0.0077(18)	0.0015(15)	-0.0022(14)	0.0002(15)
O3	0.0117(18)	0.0139(19)	0.0095(18)	0.0034(15)	-0.0010(14)	0.0005(15)
O4	0.0100(18)	0.020(2)	0.0128(19)	0.0018(16)	-0.0015(14)	-0.0003(15)
O5	0.0101(18)	0.015(2)	0.0074(17)	0.0012(15)	-0.0015(14)	-0.0022(15)
O6	0.0123(18)	0.0146(19)	0.0088(17)	-0.0014(15)	-0.0023(14)	0.0025(15)
O7	0.0097(18)	0.020(2)	0.0123(19)	0.0035(16)	-0.0024(15)	0.0004(16)
O8	0.0075(17)	0.0167(19)	0.0048(16)	-0.0002(14)	-0.0035(13)	0.0009(15)
O9	0.017(2)	0.020(2)	0.018(2)	0.0019(17)	0.0000(16)	-0.0026(16)
N1	0.014(2)	0.017(2)	0.008(2)	0.0017(18)	0.0018(17)	0.0034(18)
N2	0.011(2)	0.018(2)	0.008(2)	0.0013(18)	-0.0036(17)	0.0041(18)
C1	0.021(3)	0.019(3)	0.018(3)	-0.004(2)	0.005(2)	-0.001(2)

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
C2	0.025(3)	0.013(3)	0.029(3)	-0.003(2)	0.009(3)	0.000(2)
C3	0.029(3)	0.017(3)	0.027(3)	0.007(3)	0.015(3)	0.003(2)
C4	0.026(3)	0.021(3)	0.014(3)	0.004(2)	0.008(2)	0.008(2)
C5	0.017(3)	0.018(3)	0.009(2)	0.003(2)	0.002(2)	0.008(2)
C6	0.011(3)	0.022(3)	0.011(2)	0.002(2)	0.000(2)	0.008(2)
C7	0.017(3)	0.030(3)	0.007(2)	0.003(2)	0.001(2)	0.005(2)
C8	0.014(3)	0.036(4)	0.011(3)	-0.004(2)	-0.003(2)	0.007(3)
C9	0.012(3)	0.030(3)	0.016(3)	-0.008(2)	-0.001(2)	0.001(2)
C10	0.013(3)	0.021(3)	0.013(3)	-0.002(2)	-0.002(2)	0.002(2)
C11	0.018(3)	0.013(3)	0.017(3)	0.001(2)	0.001(2)	0.003(2)
C12	0.018(3)	0.008(3)	0.015(3)	-0.002(2)	0.002(2)	0.000(2)
C13	0.027(3)	0.019(3)	0.011(3)	0.006(2)	-0.003(2)	-0.002(2)
C14	0.019(3)	0.023(3)	0.021(3)	0.006(3)	-0.010(2)	-0.004(3)
C15	0.012(3)	0.015(3)	0.010(2)	-0.001(2)	0.001(2)	0.003(2)
C16	0.015(3)	0.013(3)	0.011(3)	-0.001(2)	-0.001(2)	0.003(2)
C17	0.013(3)	0.018(3)	0.021(3)	0.006(2)	-0.004(2)	-0.003(2)
C18	0.013(3)	0.017(3)	0.011(3)	-0.002(2)	0.000(2)	-0.001(2)

Table S109. Hydrogen atomic coordinates and isotropic atomic displacement parameters ($Å^2$) for [{$Cu_3(bpy)_2$}$Mo_2F_2O_4(H_2O)_2$(1,3-$O_3PC_8H_8PO_3$)$_2$] (**14**).

	x/a	y/b	z/c	U(eq)
H7C	0.482(4)	-0.039(3)	0.923(4)	0.018
H7D	0.646(4)	-0.037(3)	0.939(4)	0.018
H1	0.0311	0.3587	0.7469	0.023
H2	-0.1297	0.2950	0.8328	0.026
H3	-0.1133	0.3152	1.0087	0.028
H4	0.0572	0.4001	1.0952	0.023
H7	0.2343	0.4790	1.1647	0.022
H8	0.3967	0.5732	1.2186	0.026
H9	0.4901	0.6356	1.0950	0.024
H10	0.4276	0.6008	0.9216	0.02
H11A	0.3425	0.2875	0.5287	0.02
H11B	0.4302	0.2884	0.6494	0.02
H13	0.0334	0.2807	0.4843	0.024
H14	-0.1829	0.2241	0.5254	0.028
H15	0.3051	0.2328	0.7688	0.015
H17	-0.1580	0.1748	0.6876	0.022
H18A	-0.0191	0.1566	0.8578	0.017
H18B	0.1459	0.1955	0.9037	0.017