Table S1. Crystal data for *trans*-[Ni(cyan-κN)$_2$(NH$_3$)$_4$].

determination	**1a**	**1b**	**1c**	**2a**	**2b**	**2c**	**3**	**4**	**5**
temperature, K	298	150	100	298	139	298	223	148	13
space group	Fmmm	Cmcm	Cmcm	Fmmm	Cmcm	Fmmm	Cmcm	Cmcm	Cmcm
ang. distortion param, deg	0	12.1	13.6	0	13.2	0	8.7	12.0	15.3
dihedral[a]	0	29.55(8)	33.28(8)	0	32.14(5)	0	21.61(4)	29.48(10)	35.18(1)
formula	$C_6H_{16}N_{10}NiO_6$								
fw	382.95								
crystal system	orthorhombic								
crystal color	purple/violet								
cryst size, mm	See note (i).			0.4 × 0.3 × 0.3			0.4×0.3×0.3	0.2×0.13×0.07	
a, Å	12.0455(12)	12.014(2)	11.998(4)	12.0551(6)	12.015(2)	12.0551(12)	12.0338(11)	12.006(2)	12.0323(15)
b, Å	7.2710(12)	7.219(2)	7.203(6)	7.2825(6)	7.226(2)	7.2846(12)	7.2535(10)	7.215(2)	7.2266(12)
c, Å	16.0707(15)	15.805(2)	15.736(5)	16.0779(7)	15.766(3)	16.0766(14)	15.9377(13)	15.809(3)	15.661(6)
V, Å^3	1407.5(3)	1370.7(5)	1359.9(13)	1411.5(2)	1368.7(6)	1411.8(3)	1391.2(3)	1369.4(5)	1361.8(6)
Z	4								
d_{calcd}, g/cm^3	1.807	1.856	1.871	1.802	1.859	1.802	1.829	1.858	1.868

μ (MoKα), cm^{-1}	14.32	14.71	14.82	14.28	14.73	14.28	14.49	14.72	$1.537 + 1.698 \cdot \lambda^{g}$
transmission, max., min.	0.915, 0.875			1.000, 0.988			1.000,0.874	0.962, 0.794	
radiation	MoKα ($\lambda_{<\alpha>}$ = 0.71073 Å)(graphite monochromated)								
2θ (min,max), deg	4.0-50.0	4.0-50.0	4.0-60.0	4.0-60.0	4.0-60.0	4.0-60.0	4.0-60.0	4.0-50.0	see text
scan method	ω-scan	ω-scan	ω-scan	ω-2θ scan	ω-θ scan	ω-2θ scan	ω-2θ scan	ω-scan	TOF
instrument	CAD-4								IPNS/SCD
no. unique data	370	666	1068	595	1067	597	1094	667	1164
no. data with $F \geq 2 \cdot \sigma(F)$	321	565	871	483	880	486	942	511	1164
no., type data in refinement	370 F_o^2	613 F_o^2	1032 F_o^2	544 F_o^2	1067 F_o^2	597 F_o^2	1094 F_o^2	667 F_o^2	1164 |F|
no. restraints	0	0	0	0	0	0	0	0	0
no. parameters refined	49	80	80	49	80	49	72	80	
weighting parms g_1, g_2[b]	0.0600,0.90	0.0492,1.00	0.0387,0.00	0.0505,0.25	0.0469,0.10	0.0500,0.16	0.0465,1.30	0.0442,0.00	
wR2[c]	0.1057	0.0943	0.0951	0.0951	0.0859	0.0827	0.0895	0.1020	0.091
R1[d] (for $F_o^2 \geq 2 \cdot \sigma(F_o^2)$)	0.0398	0.0371	0.0393	0.0382	0.0334	0.0301	0.0313	0.0434	0.069[h]
quality of fit[e]	1.076	1.096	1.069	1.087	0.997	1.075	1.120	1.033	2.86
restrained quality of fit[f]	(1.076)	(1.096)	(1.069)	(1.087)	(0.997)	(1.075)	(1.120)	(1.033)	(2.86)

mean, max $\lvert\Delta/\sigma\rvert$	<0.001, 0.002	<0.001, <0.001	<0.001, 0.001	<0.001, <0.001	0.001, 0.008	<0.001, 0.001	0.003, 0.017	0.002, 0.009	
max,min $\Delta\rho$, e/Å^3	0.47,-0.31	0.59,-0.36	0.98,-0.50	0.38,-0.27	0.42,-0.41	0.40,-0.24	0.81,-0.44	0.71,-0.48	

[a]Dihedral angle between the six-membered rings of the two cyanurate ligands. [b]$w^{-1} = [\sigma^2(F_o^2) + (g_1 \cdot P)^2 + g_2 \cdot P]$; $P = [\max(F_o^2;0) + 2F_{calc}^2] / 3$. [c] $wR2 = [\Sigma w(F_o^2 - F_c^2)^2 / \Sigma w(F_o^2)^2]^{1/2}$ [d]$R1 = \Sigma \; ||F_{obs}| - |F_{calc}|| \; / \Sigma \; |F_{obs}|$ [e]$q.o.f. = [\Sigma w(F_o^2 - F_c^2)^2 / (n_{obs} - n_{params})]^{1/2}$ [f]restrained q.o.f. = $[\{\Sigma w(F_o^2 - F_c^2)^2 + \Sigma w(Y_{target} - Y_{calc})^2\} / (n_{reflecns} + n_{restraints} - n_{params})]^{1/2}$. [g]for neutron data in the wavelength range 0.7 – 4.2 Å. [h]for $F_o^2 \geq 3 \cdot \sigma(F_o^2)$. [i]Crystal 1 was lost before its size could be measured.

Table S2. Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters (A^2 x 10^3) for 1a. T = 298 K.
U(eq) is defined as one third of the trace of the orthogonalized Uij tensor.

	x	y	z	U(eq)
Ni(1)	0	0	0	38(1)
N(2)	1189(4)	2053(8)	0	46(1)
N(1)	0	0	-1428(3)	39(2)
C(2)	-972(4)	0	-1857(3)	37(1)
O(2)	-1890(2)	0	-1516(2)	52(1)
N(3)	-944(3)	0	-2711(2)	36(1)
C(4)	0	0	-3179(4)	34(2)
O(4)	0	0	-3934(3)	47(1)

Table S3. Bond lengths [A] and angles [deg] for 1a. T = 298 K.

Ni(1)-N(2)	2.068(5)
Ni(1)-N(1)	2.295(5)
N(2)-H(20)	0.97(9)
N(2)-H(21)	0.72(6)
N(1)-C(2)	1.358(5)
C(2)-O(2)	1.234(6)
C(2)-N(3)	1.373(6)
N(3)-C(4)	1.363(5)
N(3)-H(3)	0.80(6)
C(4)-O(4)	1.213(8)
N(2)-Ni(1)-N(2)#1	180.0
N(2)-Ni(1)-N(2)#2	87.6(3)
N(2)-Ni(1)-N(1)	90.0
N(1)-Ni(1)-N(1)#1	180.0
Ni(1)-N(2)-H(20)	114(5)
Ni(1)-N(2)-H(21)	107(5)
H(20)-N(2)-H(21)	116(6)
C(2)-N(1)-C(2)#3	119.0(5)
C(2)-N(1)-Ni(1)	120.5(3)
O(2)-C(2)-N(1)	123.2(4)
O(2)-C(2)-N(3)	117.7(4)
N(1)-C(2)-N(3)	119.1(4)
C(4)-N(3)-C(2)	124.9(4)
C(4)-N(3)-H(3)	121(4)
C(2)-N(3)-H(3)	115(4)
O(4)-C(4)-N(3)	123.5(3)
N(3)#3-C(4)-N(3)	113.0(5)

Symmetry transformations used to generate equivalent atoms:
#1 -x,-y,-z #2 -x,y,z #3 -x,-y,z

Table S4. Anisotropic displacement parameters (A^2 x 10^3) for 1a. T = 298 K.
The anisotropic displacement factor exponent takes the form:
-2 pi^2 [h^2 a*^2 U11 + ... + 2 h k a* b* U12]

	U11	U22	U33	U23	U13	U12
Ni(1)	14(1)	74(1)	26(1)	0	0	0
N(2)	27(2)	77(4)	34(2)	0	0	-4(2)
N(1)	17(3)	80(4)	19(3)	0	0	0
C(2)	25(2)	66(3)	21(2)	0	-2(2)	0
O(2)	16(2)	117(3)	24(2)	0	0(1)	0
N(3)	18(2)	70(3)	19(2)	0	-2(2)	0
C(4)	29(3)	53(4)	19(4)	0	0	0
O(4)	33(3)	92(4)	17(2)	0	0	0

Table S5. Principal mean square atomic displacements U in A^2 for 1a. T = 298 K.

0.0737	0.0257	0.0136	Ni1
0.0772	0.0342	0.0264	N2
0.0798	0.0192	0.0171	N1
0.0655	0.0254	0.0206	C2
0.1167	0.0243	0.0162	O2
0.0698	0.0213	0.0164	N3
0.0527	0.0292	0.0191	C4
0.0916	0.0331	0.0170	O4

Table S6. Hydrogen coordinates (x 10^4) and isotropic displacement parameters (A^2 x 10^3) for 1a. T = 298 K.

	x	y	z	U(eq)
H(20)	880(7)	3290(13)	0	116(19)
H(21)	1570(4)	1850(9)	-330(3)	116(19)
H(3)	-1540(5)	0	-2930(4)	46(16)

Table S7. Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters (A^2 x 10^3) for 1b. T = 150 K. U(eq) is defined as one third of the trace of the orthogonalized Uij tensor.

	x	y	z	U(eq)
Ni(1)	5000	1778(1)	7500	14(1)
N(1)	5000	1836(6)	6064(2)	16(1)
C(2)	4027(3)	2054(4)	5637(2)	15(1)
O(2)	3099(2)	1845(4)	5972(1)	20(1)
N(3)	4050(2)	2543(4)	4790(2)	16(1)
C(4)	5000	2829(6)	4341(3)	15(1)
O(4)	5000	3300(5)	3597(2)	21(1)
N(4)	6178(4)	3874(7)	7500	22(1)
N(5)	6210(4)	-244(7)	7500	19(1)

Table S8. Bond lengths [A] and angles [deg] for 1b. T = 150 K.

Ni(1)-N(5)	2.060(4)
Ni(1)-N(4)	2.072(5)
Ni(1)-N(1)	2.270(4)
N(1)-C(2)	1.359(4)
C(2)-O(2)	1.242(4)
C(2)-N(3)	1.385(5)
N(3)-C(4)	1.360(4)
N(3)-H(3)	0.86(5)
C(4)-O(4)	1.224(6)
N(4)-H(41)	0.82(7)
N(4)-H(42)	0.79(11)
N(5)-H(51)	0.81(4)
N(5)-H(52)	0.89(8)
N(5)#1-Ni(1)-N(5)	89.8(3)
N(5)#1-Ni(1)-N(4)	178.2(2)
N(5)-Ni(1)-N(4)	92.0(2)
N(4)#1-Ni(1)-N(4)	86.2(3)
N(5)-Ni(1)-N(1)	90.75(8)
N(4)-Ni(1)-N(1)	89.23(8)
N(1)-Ni(1)-N(1)#2	177.9(2)
C(2)-N(1)-C(2)#3	118.8(4)
C(2)-N(1)-Ni(1)	119.9(2)
O(2)-C(2)-N(1)	123.1(3)
O(2)-C(2)-N(3)	117.4(3)
N(1)-C(2)-N(3)	119.4(3)
C(4)-N(3)-C(2)	124.1(3)
C(4)-N(3)-H(3)	119(3)
C(2)-N(3)-H(3)	117(3)
O(4)-C(4)-N(3)	122.9(2)
N(3)-C(4)-N(3)#3	114.1(4)
Ni(1)-N(4)-H(41)	112(5)
Ni(1)-N(4)-H(42)	115(8)
H(41)-N(4)-H(42)	114(6)
Ni(1)-N(5)-H(51)	114(3)
Ni(1)-N(5)-H(52)	111(5)
H(51)-N(5)-H(52)	107(4)

Symmetry transformations used to generate equivalent atoms:
#1 -x+1,y,-z+3/2 #2 x,y,-z+3/2 #3 -x+1,y,z

Table S9. Anisotropic displacement parameters (A^2 x 10^3) for 1b.
T = 150 K.
The anisotropic displacement factor exponent takes the form:
-2 pi^2 [h^2 a*^2 U11 + .. + 2 h k a* b* U12]

	U11	U22	U33	U23	U13	U12
Ni(1)	7(1)	22(1)	12(1)	0	0	0
N(1)	9(2)	28(2)	12(2)	0(2)	0	0
C(2)	13(2)	19(2)	13(2)	-1(1)	-2(1)	-1(1)
O(2)	9(1)	38(1)	14(1)	2(1)	1(1)	-1(1)
N(3)	9(1)	28(1)	11(2)	0(1)	-2(1)	1(1)
C(4)	12(2)	13(2)	19(3)	-1(2)	0	0
O(4)	22(2)	33(2)	9(2)	5(2)	0	0
N(4)	14(2)	29(2)	22(2)	0	0	-1(2)
N(5)	13(2)	27(2)	17(2)	0	0	2(2)

Table S10. Principal mean square atomic displacements U in A^2 for 1b. T = 150 K.

0.0220	0.0124	0.0073	Ni1
0.0283	0.0121	0.0087	N1
0.0195	0.0150	0.0108	C2
0.0382	0.0138	0.0086	O2
0.0275	0.0125	0.0075	N3
0.0190	0.0131	0.0123	C4
0.0342	0.0217	0.0079	O4
0.0293	0.0219	0.0135	N4
0.0270	0.0167	0.0125	N5

Table S11. Hydrogen coordinates (x 10^4) and isotropic displacement parameters (A^2 x 10^3) for 1b. T = 150 K.

	x	y	z	U(eq)
H(3)	3424(39)	2606(55)	4533(29)	42(13)
H(41)	6043(49)	4670(98)	7144(42)	119(29)
H(42)	6805(87)	3529(146)	7500	101(37)
H(51)	6620(31)	-208(62)	7904(23)	28(11)
H(52)	5899(60)	-1370(104)	7500	49(21)

Table S12. Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters (A^2 x 10^3) for 1c. T = 100 K. U(eq) is defined as one third of the trace of the orthogonalized Uij tensor.

	x	y	z	U(eq)
Ni(1)	5000	1679(1)	7500	10(1)
N(1)	5000	1754(4)	6064(2)	11(1)
C(2)	4019(2)	2001(3)	5641(1)	11(1)
O(2)	3098(1)	1757(2)	5969(1)	14(1)
N(3)	4052(2)	2561(3)	4793(1)	12(1)
C(4)	5000	2869(4)	4347(2)	11(1)
O(4)	5000	3412(4)	3607(2)	16(1)
N(4)	6180(2)	3781(4)	7500	14(1)
N(5)	6214(2)	-352(4)	7500	14(1)

Table S13. Bond lengths [A] and angles [deg] for 1c. T = 100 K.

Ni(1)-N(5)	2.065(3)
Ni(1)-N(4)	2.073(3)
Ni(1)-N(1)	2.260(3)
N(1)-C(2)	1.364(2)
C(2)-O(2)	1.232(3)
C(2)-N(3)	1.394(3)
N(3)-C(4)	1.356(2)
N(3)-H(3)	0.77(4)
C(4)-O(4)	1.228(4)
N(4)-H(41)	0.80(4)
N(4)-H(42)	0.86(6)
N(5)-H(51)	0.86(3)
N(5)-H(52)	0.85(6)
N(5)#1-Ni(1)-N(5)	89.7(2)
N(5)#1-Ni(1)-N(4)	178.21(13)
N(5)-Ni(1)-N(4)	92.04(12)
N(4)#1-Ni(1)-N(4)	86.2(2)
N(5)-Ni(1)-N(1)	90.97(5)
N(4)-Ni(1)-N(1)	89.00(5)
N(1)-Ni(1)-N(1)#2	177.27(15)
C(2)-N(1)-C(2)#3	119.2(3)
C(2)-N(1)-Ni(1)	119.46(14)
O(2)-C(2)-N(1)	123.3(2)
O(2)-C(2)-N(3)	117.9(2)
N(1)-C(2)-N(3)	118.8(2)
C(4)-N(3)-C(2)	124.5(2)
C(4)-N(3)-H(3)	119(3)
C(2)-N(3)-H(3)	116(3)
O(4)-C(4)-N(3)	122.92(14)
N(3)-C(4)-N(3)#3	114.1(3)
Ni(1)-N(4)-H(41)	118(3)
Ni(1)-N(4)-H(42)	116(4)
H(41)-N(4)-H(42)	105(4)
Ni(1)-N(5)-H(51)	104(3)
Ni(1)-N(5)-H(52)	110(4)
H(51)-N(5)-H(52)	117(3)

Symmetry transformations used to generate equivalent atoms:
#1 -x+1,y,-z+3/2 #2 x,y,-z+3/2 #3 -x+1,y,z

Table S14. Anisotropic displacement parameters (A^2 x 10^3) for 1c. T = 100 K.
The anisotropic displacement factor exponent takes the form:
-2 pi^2 [h^2 a*^2 U11 + ... + 2 h k a* b* U12]

	U11	U22	U33	U23	U13	U12
Ni(1)	5(1)	16(1)	9(1)	0	0	0
N(1)	7(1)	20(1)	7(1)	2(1)	0	0
C(2)	11(1)	14(1)	7(1)	-2(1)	0(1)	2(1)
O(2)	7(1)	26(1)	10(1)	1(1)	0(1)	-2(1)
N(3)	8(1)	20(1)	7(1)	1(1)	-2(1)	0(1)
C(4)	11(1)	11(1)	10(2)	0(1)	0	0
O(4)	16(1)	23(1)	8(1)	3(1)	0	0
N(4)	11(1)	17(1)	15(1)	0	0	0(1)
N(5)	10(1)	19(1)	12(1)	0	0	-1(1)

Table S15. Principal mean square atomic displacements U in A^2 for 1c. T = 100 K.

0.0156	0.0094	0.0053	Ni1
0.0202	0.0068	0.0067	N1
0.0153	0.0100	0.0067	C2
0.0267	0.0103	0.0064	O2
0.0201	0.0093	0.0059	N3
0.0114	0.0110	0.0103	C4
0.0240	0.0157	0.0071	O4
0.0169	0.0150	0.0105	N4
0.0190	0.0121	0.0099	N5

Table S16. Hydrogen coordinates (x 10^4) and isotropic displacement parameters (A^2 x 10^3) for 1c. T = 100 K.

	x	y	z	U(eq)
H(3)	3481(37)	2706(53)	4573(26)	56(13)
H(41)	6139(31)	4551(55)	7137(23)	52(13)
H(42)	6869(51)	3429(86)	7500	52(17)
H(51)	6621(27)	-68(56)	7928(20)	33(9)
H(52)	5913(50)	-1418(87)	7500	52(17)

Table S17. Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters (A^2 x 10^3) for 2a. T = 298 K. U(eq) is defined as one third of the trace of the orthogonalized Uij tensor.

	x	y	z	U(eq)
Ni(1)	0	0	0	35(1)
N(2)	1194(2)	2039(5)	0	42(1)
N(1)	0	0	-1426(2)	36(1)
C(2)	-970(2)	0	-1859(2)	32(1)
O(2)	-1890(2)	0	-1519(1)	49(1)
N(3)	-944(2)	0	-2716(1)	33(1)
C(4)	0	0	-3178(2)	30(1)
O(4)	0	0	-3935(2)	44(1)

Table S18. Bond lengths [A] and angles [deg] for 2a. T = 298 K.

Ni(1)-N(2)	2.068(3)
Ni(1)-N(1)	2.293(3)
N(2)-H(20)	0.74(6)
N(2)-H(21)	0.84(4)
N(1)-C(2)	1.361(3)
C(2)-O(2)	1.237(3)
C(2)-N(3)	1.378(4)
N(3)-C(4)	1.360(3)
N(3)-H(3)	0.86(4)
C(4)-O(4)	1.216(5)
C(4)-N(3)#1	1.360(3)
N(2)#2-Ni(1)-N(2)#3	91.8(2)
N(2)#2-Ni(1)-N(2)	180.0
N(2)-Ni(1)-N(1)	90.0
N(1)-Ni(1)-N(1)#2	180.0
Ni(1)-N(2)-H(20)	119(5)
Ni(1)-N(2)-H(21)	110(3)
H(20)-N(2)-H(21)	107(4)
C(2)-N(1)-C(2)#1	118.5(3)
C(2)-N(1)-Ni(1)	120.8(2)
O(2)-C(2)-N(1)	123.0(3)
O(2)-C(2)-N(3)	117.5(2)
N(1)-C(2)-N(3)	119.5(3)
C(4)-N(3)-C(2)	124.4(3)
C(4)-N(3)-H(3)	120(2)
C(2)-N(3)-H(3)	116(2)
O(4)-C(4)-N(3)	123.1(2)
N(3)#1-C(4)-N(3)	113.7(3)

Symmetry transformations used to generate equivalent atoms:
#1 -x,-y,z #2 -x,-y,-z #3 -x,y,z

Table S19. Anisotropic displacement parameters (A^2 x 10^3) for 2a. T = 298 K.
The anisotropic displacement factor exponent takes the form:
-2 pi^2 [h^2 a*^2 U11 + ... + 2 h k a* b* U12]

	U11	U22	U33	U23	U13	U12
Ni(1)	12(1)	68(1)	25(1)	0	0	0
N(2)	20(1)	73(2)	35(1)	0	0	-4(1)
N(1)	14(1)	76(3)	18(2)	0	0	0
C(2)	20(1)	55(2)	21(1)	0	-2(1)	0
O(2)	16(1)	109(2)	24(1)	0	-1(1)	0
N(3)	18(1)	62(2)	18(1)	0	-2(1)	0
C(4)	25(2)	45(2)	21(2)	0	0	0
O(4)	35(2)	82(2)	15(1)	0	0	0

Table S20. Principal mean square atomic displacements U in A^2 for 2a. T = 298 K.

0.0676	0.0249	0.0124	Ni1
0.0731	0.0349	0.0193	N2
0.0761	0.0179	0.0145	N1
0.0550	0.0230	0.0187	C2
0.1087	0.0239	0.0156	O2
0.0616	0.0202	0.0159	N3
0.0449	0.0247	0.0214	C4
0.0816	0.0347	0.0154	O4

Table S21. Hydrogen coordinates (x 10^4) and isotropic displacement parameters (A^2 x 10^3) for 2a. T = 298 K.

	x	y	z	U(eq)
H(20)	1014(53)	3012(89)	0	114(14)
H(21)	1618(30)	1894(58)	-409(23)	114(14)
H(3)	-1581(29)	0	-2960(21)	31(9)

Table S22. Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters (A^2 x 10^3) for 2b. T = 139 K.
U(eq) is defined as one third of the trace of the orthogonalized Uij tensor.

	x	y	z	U(eq)
Ni(1)	5000	1711(1)	7500	11(1)
N(1)	5000	1782(3)	6064(1)	13(1)
C(2)	4023(2)	2023(3)	5637(1)	12(1)
O(2)	3099(1)	1782(2)	5971(1)	16(1)
N(3)	4049(1)	2554(2)	4792(1)	13(1)
C(4)	5000	2863(4)	4342(2)	12(1)
O(4)	5000	3376(3)	3602(1)	17(1)
N(4)	6182(2)	3812(4)	7500	15(1)
N(5)	6212(2)	-318(4)	7500	15(1)

Table S23. Bond lengths [A] and angles [deg] for 2b. T = 139 K.

Ni(1)-N(5)	2.066(2)
Ni(1)-N(4)	2.079(2)
Ni(1)-N(1)	2.265(2)
N(1)-C(2)	1.364(2)
C(2)-O(2)	1.241(2)
C(2)-N(1)#1	1.364(2)
C(2)-N(3)	1.388(2)
N(3)-C(4)	1.362(2)
N(3)-H(3)	0.86(3)
C(4)-O(4)	1.224(3)
N(4)-H(41)	0.82(4)
N(4)-H(42)	0.91(6)
N(5)-H(51)	0.87(3)
N(5)-H(52)	0.83(6)
N(5)-Ni(1)-N(5)#2	89.60(15)
N(5)-Ni(1)-N(4)#2	178.27(11)
N(5)-Ni(1)-N(4)	92.12(10)
N(4)#2-Ni(1)-N(4)	86.15(15)
N(5)-Ni(1)-N(1)	90.93(4)
N(5)#2-Ni(1)-N(1)	90.92(4)
N(4)-Ni(1)-N(1)	89.05(5)
N(1)#3-Ni(1)-N(1)	177.39(12)
C(2)-N(1)-C(2)#4	118.8(2)
C(2)-N(1)-Ni(1)	119.71(12)
O(2)-C(2)-N(1)	122.93(17)
O(2)-C(2)-N(3)	117.78(17)
N(1)-C(2)-N(3)	119.29(18)
C(4)-N(3)-C(2)	124.32(18)
C(4)-N(3)-H(3)	117.9(18)
C(2)-N(3)-H(3)	117.7(18)
O(4)-C(4)-N(3)	123.04(12)
N(3)#4-C(4)-N(3)	113.9(2)
Ni(1)-N(4)-H(41)	116(2)
Ni(1)-N(4)-H(42)	116(4)
H(41)-N(4)-H(42)	105(3)
Ni(1)-N(5)-H(51)	108(2)
Ni(1)-N(5)-H(52)	116(4)
H(51)-N(5)-H(52)	111(3)

Symmetry transformations used to generate equivalent atoms:

#1 x,y,z #2 -x+1,y,-z+3/2 #3 x,y,-z+3/2 #4 -x+1,y,z

Table S24. Anisotropic displacement parameters (A^2 x 10^3) for 2b. T = 139 K.
The anisotropic displacement factor exponent takes the form:
-2 pi^2 [h^2 a*^2 U11 + ... + 2 h k a* b* U12]

	U11	U22	U33	U23	U13	U12
Ni(1)	7(1)	14(1)	11(1)	0	0	0
N(1)	8(1)	19(1)	12(1)	1(1)	0	0
C(2)	12(1)	15(1)	10(1)	0(1)	-1(1)	0(1)
O(2)	9(1)	29(1)	12(1)	3(1)	-1(1)	-1(1)
N(3)	9(1)	20(1)	11(1)	1(1)	-1(1)	1(1)
C(4)	14(1)	11(1)	12(1)	-1(1)	0	0
O(4)	17(1)	22(1)	11(1)	7(1)	0	0
N(4)	11(1)	18(1)	16(1)	0	0	-3(1)
N(5)	10(1)	18(1)	16(1)	0	0	0(1)

Table S25. Principal mean square atomic displacements U in A^2 for 2b. T = 139 K.

0.0144	0.0109	0.0065	Ni1
0.0190	0.0118	0.0078	N1
0.0146	0.0123	0.0095	C2
0.0291	0.0117	0.0083	O2
0.0205	0.0114	0.0082	N3
0.0136	0.0128	0.0108	C4
0.0253	0.0167	0.0083	O4
0.0185	0.0160	0.0098	N4
0.0184	0.0162	0.0100	N5

Table S26. Hydrogen coordinates (x 10^4) and isotropic displacement parameters (A^2 x 10^3) for 2b. T = 139 K.

	x	y	z	U(eq)
H(3)	3420(2)	2710(4)	4536(18)	27(7)
H(41)	6130(3)	4570(5)	7110(2)	58(11)
H(42)	6910(5)	3440(8)	7500	75(19)
H(51)	6660(2)	-110(5)	7928(17)	32(7)
H(52)	5990(5)	-1410(8)	7500	61(17)

Table S27. Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters (A^2 x 10^3) for 2c. T = 298 K. U(eq) is defined as one third of the trace of the orthogonalized Uij tensor.

	x	y	z	U(eq)
Ni(1)	0	0	0	35(1)
N(2)	1195(2)	2033(4)	0	42(1)
N(1)	0	0	-1429(2)	36(1)
C(2)	-971(2)	0	-1855(1)	33(1)
O(2)	-1891(1)	0	-1516(1)	50(1)
N(3)	-944(2)	0	-2716(1)	33(1)
C(4)	0	0	-3179(2)	30(1)
O(4)	0	0	-3935(1)	45(1)

Table S28. Bond lengths [A] and angles [deg] for 2c. T = 298 K.

Ni(1)-N(2)	2.066(2)
Ni(1)-N(1)	2.297(2)
N(2)-H(20)	0.82(5)
N(2)-H(21)	0.86(3)
N(1)-C(2)	1.356(2)
C(2)-O(2)	1.235(3)
C(2)-N(3)	1.385(3)
N(3)-C(4)	1.360(2)
N(3)-H(3)	0.82(3)
C(4)-O(4)	1.216(4)
N(2)#1-Ni(1)-N(2)#2	88.40(14)
N(2)#1-Ni(1)-N(2)	91.60(14)
N(2)#2-Ni(1)-N(2)	180.0
N(2)-Ni(1)-N(1)	90.0
N(1)#2-Ni(1)-N(1)	180.0
Ni(1)-N(2)-H(20)	119(4)
Ni(1)-N(2)-H(21)	110(2)
H(20)-N(2)-H(21)	104(3)
C(2)#3-N(1)-C(2)	119.4(2)
C(2)-N(1)-Ni(1)	120.32(12)
O(2)-C(2)-N(1)	123.51(19)
O(2)-C(2)-N(3)	117.50(18)
N(1)-C(2)-N(3)	118.99(19)
C(4)-N(3)-C(2)	124.48(18)
C(4)-N(3)-H(3)	120(2)
C(2)-N(3)-H(3)	115(2)
O(4)-C(4)-N(3)	123.14(12)
N(3)#3-C(4)-N(3)	113.7(2)

Symmetry transformations used to generate equivalent atoms:
#1 x,-y,-z #2 -x,-y,-z #3 -x,-y,z

Table S29. Anisotropic displacement parameters (A^2 x 10^3) for 2c. T = 298 K.
The anisotropic displacement factor exponent takes the form:
-2 pi^2 [h^2 a*^2 U11 + ... + 2 h k a* b* U12]

	U11	U22	U33	U23	U13	U12
Ni(1)	12(1)	68(1)	25(1)	0	0	0
N(2)	21(1)	73(2)	34(1)	0	0	-4(1)
N(1)	16(1)	73(2)	18(1)	0	0	0
C(2)	20(1)	62(1)	18(1)	0	-1(1)	0
O(2)	16(1)	112(2)	22(1)	0	0(1)	0
N(3)	19(1)	61(1)	17(1)	0	-3(1)	0
C(4)	25(1)	44(2)	20(1)	0	0	0
O(4)	35(1)	82(2)	16(1)	0	0	0

Table S30. Principal mean square atomic displacements U in A^2 for 2c. T = 298 K.

0.0679	0.0249	0.0121	Ni1
0.0732	0.0336	0.0203	N2
0.0734	0.0180	0.0158	N1
0.0616	0.0205	0.0175	C2
0.1117	0.0220	0.0162	O2
0.0612	0.0211	0.0155	N3
0.0441	0.0251	0.0201	C4
0.0821	0.0351	0.0163	O4

Table S31. Hydrogen coordinates (x 10^4) and isotropic displacement parameters (A^2 x 10^3) for 2c. T = 298 K.

	x	y	z	U(eq)
H(20)	1000(4)	3120(7)	0	101(10)
H(21)	1610(3)	1930(4)	-433(19)	101(10)
H(3)	-1550(3)	0	-2944(18)	33(7)

Table S32. Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters (A^2 x 10^3) for 3. T = 223 K. U(eq) is defined as one third of the trace of the orthogonalized Uij tensor.

	x	y	z	U(eq)
Ni(1)	5000	1964(1)	7500	20(1)
N(1)	5000	2006(3)	6070(1)	22(1)
C(2)	4027(1)	2164(3)	5641(1)	22(1)
O(2)	3104(1)	2002(2)	5976(1)	31(1)
N(3)	4053(1)	2520(2)	4785(1)	23(1)
C(4)	5000	2738(3)	4330(2)	22(1)
O(4)	5000	3078(3)	3578(1)	29(1)
N(4)	6186(2)	4037(3)	7500	26(1)
N(5)	6206(2)	-58(4)	7500	26(1)

Table S33. Bond lengths [A] and angles [deg] for 3. T = 223 K. N.b., atoms H(41D) and H(41G) have zero occupancy. They were used as aids in idealizing the ammine groups, and are symmetry relatives of H(41C) and H(41F), respectively. H(41C) and H(42C) belong to the orientation of the NH3 ligand found in the Cmcm structure, and H(41F) and H(42F) belong to the orientation found in the Fmmm structure.

Ni(1)-N(5)	2.063(2)
Ni(1)-N(4)	2.074(2)
Ni(1)-N(1)	2.280(2)
N(1)-C(2)	1.3603(19)
C(2)-O(2)	1.238(2)
C(2)-N(3)	1.388(2)
N(3)-C(4)	1.3601(19)
N(3)-H(3)	0.79(3)
C(4)-O(4)	1.224(3)
N(4)-H(41C)	0.8901
N(4)-H(42C)	0.8901
N(4)-H(41D)	0.8901
N(4)-H(41F)	0.8901
N(4)-H(42F)	0.8901
N(4)-H(41G)	0.8901
N(5)-H(52)	0.84(4)
N(5)-H(51)	0.85(2)
N(5)#1-Ni(1)-N(5)	89.41(14)
N(5)-Ni(1)-N(4)	91.78(9)
N(5)-Ni(1)-N(4)#1	178.80(10)
N(4)-Ni(1)-N(4)#1	87.02(13)
N(5)-Ni(1)-N(1)#2	90.55(4)
N(4)-Ni(1)-N(1)#2	89.44(5)
N(1)#2-Ni(1)-N(1)	178.44(12)
C(2)-N(1)-C(2)#3	118.7(2)
C(2)-N(1)-Ni(1)	120.25(10)
O(2)-C(2)-N(1)	123.14(16)
O(2)-C(2)-N(3)	117.49(15)
N(1)-C(2)-N(3)	119.37(16)
C(4)-N(3)-C(2)	124.33(16)
C(4)-N(3)-H(3)	117(2)
C(2)-N(3)-H(3)	118(2)
O(4)-C(4)-N(3)	123.07(11)

N(3)#3-C(4)-N(3)	113.9(2)
Ni(1)-N(4)-H(41C)	109.5
Ni(1)-N(4)-H(42C)	109.5
H(41C)-N(4)-H(42C)	109.5
Ni(1)-N(4)-H(41D)	109.5
H(41C)-N(4)-H(41D)	109.5
H(42C)-N(4)-H(41D)	109.5
Ni(1)-N(4)-H(41F)	109.5
Ni(1)-N(4)-H(42F)	109.4
H(41F)-N(4)-H(42F)	109.5
Ni(1)-N(4)-H(41G)	109.5
H(41F)-N(4)-H(41G)	109.5
H(42F)-N(4)-H(41G)	109.5
Ni(1)-N(5)-H(52)	112(2)
Ni(1)-N(5)-H(51)	110.3(17)
H(52)-N(5)-H(51)	109(2)

Symmetry transformations used to generate equivalent atoms:
#1 -x+1,y,-z+3/2 #2 x,y,-z+3/2 #3 -x+1,y,z

Table S34. Anisotropic displacement parameters (A^2 x 10^3) for 3. T = 223 K.
The anisotropic displacement factor exponent takes the form:
-2 pi^2 [h^2 a*^2 U11 + ... + 2 h k a* b* U12]

	U11	U22	U33	U23	U13	U12
Ni(1)	9(1)	32(1)	20(1)	0	0	0
N(1)	11(1)	40(1)	17(1)	0(1)	0	0
C(2)	15(1)	35(1)	16(1)	0(1)	-1(1)	0(1)
O(2)	12(1)	60(1)	20(1)	3(1)	-1(1)	-1(1)
N(3)	13(1)	40(1)	15(1)	2(1)	-2(1)	1(1)
C(4)	18(1)	28(1)	18(1)	0(1)	0	0
O(4)	26(1)	43(1)	18(1)	7(1)	0	0
N(4)	15(1)	39(1)	25(1)	0	0	-2(1)
N(5)	14(1)	37(1)	28(1)	0	0	3(1)

Table S35. Principal mean square atomic displacements U in A^2 for 3. T = 223 K.

0.0319	0.0200	0.0085	Ni1
0.0398	0.0168	0.0107	N1
0.0347	0.0162	0.0145	C2
0.0604	0.0196	0.0117	O2
0.0406	0.0160	0.0119	N3
0.0284	0.0184	0.0183	C4
0.0448	0.0262	0.0162	O4
0.0388	0.0251	0.0152	N4
0.0371	0.0275	0.0140	N5

Table S36. Hydrogen coordinates (x 10^4) and isotropic displacement parameters (A^2 x 10^3) for 3. T = 223 K. N.b., atoms H(41D) and H(41G) have zero occupancy. They were used as aids in idealizing the ammine groups, and are symmetry relatives of H(41C) and H(41F), respectively. H(41C) and H(42C) belong to the orientation of the NH3 ligand found in the Cmcm structure, and H(41F) and H(42F) belong to the orientation found in the Fmmm structure.

	x	y	z	U(eq)
H(3)	3480(3)	2570(3)	4539(18)	40(7)
H(41C)	6103	4732	7044	33
H(42C)	6862	3538	7500	33
H(41D)	6103	4732	7956	33
H(41F)	6609	3936	7044	33
H(42F)	5850	5130	7500	33
H(41G)	6609	3936	7956	33
H(52)	5930(3)	-1120(5)	7500	33
H(51)	6630(2)	60(4)	7927(15)	33

Table S37. Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters (A^2 x 10^3) for 4. T = 148 K.
U(eq) is defined as one third of the trace of the orthogonalized Uij tensor.

	x	y	z	U(eq)
Ni(1)	5000	1777(2)	7500	15(1)
N(1)	5000	1839(7)	6066(3)	14(1)
C(2)	4023(3)	2058(5)	5638(2)	15(1)
O(2)	3101(2)	1832(5)	5974(2)	21(1)
N(3)	4049(3)	2545(5)	4792(2)	15(1)
C(4)	5000	2835(8)	4340(4)	14(1)
O(4)	5000	3295(6)	3595(2)	22(1)
N(4)	6182(5)	3864(9)	7500	20(1)
N(5)	6202(4)	-262(9)	7500	20(1)

Table S38. Bond lengths [A] and angles [deg] for 4. T = 148 K.

Ni(1)-N(5)	2.061(6)
Ni(1)-N(4)	2.069(6)
Ni(1)-N(1)	2.268(4)
N(1)-C(2)	1.363(4)
C(2)-O(2)	1.239(5)
C(2)-N(3)	1.384(5)
N(3)-C(4)	1.363(4)
N(3)-H(3)	0.86(5)
C(4)-O(4)	1.223(7)
N(4)-H(41)	0.84(6)
N(4)-H(42)	0.64(11)
N(5)-H(51)	0.81(4)
N(5)-H(52)	0.87(9)
N(5)#1-Ni(1)-N(5)	88.9(4)
N(5)-Ni(1)-N(4)	92.3(2)
N(5)-Ni(1)-N(4)#1	178.8(3)
N(4)-Ni(1)-N(4)#1	86.6(4)
N(5)-Ni(1)-N(1)	90.81(10)
N(4)-Ni(1)-N(1)	89.18(10)
N(1)#2-Ni(1)-N(1)	177.7(3)
C(2)-N(1)-C(2)#3	118.7(5)
C(2)-N(1)-Ni(1)	119.9(2)
O(2)-C(2)-N(1)	122.8(4)
O(2)-C(2)-N(3)	117.9(4)
N(1)-C(2)-N(3)	119.3(4)
C(4)-N(3)-C(2)	124.4(4)
C(4)-N(3)-H(3)	115(3)
C(2)-N(3)-H(3)	121(3)
O(4)-C(4)-N(3)	123.1(2)
N(3)#3-C(4)-N(3)	113.8(5)
Ni(1)-N(4)-H(41)	111(4)
Ni(1)-N(4)-H(42)	119(10)
H(41)-N(4)-H(42)	111(8)
Ni(1)-N(5)-H(51)	106(4)
Ni(1)-N(5)-H(52)	116(6)
H(51)-N(5)-H(52)	109(5)

Symmetry transformations used to generate equivalent atoms:
#1 -x+1,y,-z+3/2 #2 x,y,-z+3/2 #3 -x+1,y,z

Table S39. Anisotropic displacement parameters (A^2 x 10^3) for 4. T = 148 K.
The anisotropic displacement factor exponent takes the form:
-2 pi^2 [h^2 a*^2 U11 + ... + 2 h k a* b* U12]

	U11	U22	U33	U23	U13	U12
Ni(1)	7(1)	25(1)	13(1)	0	0	0
N(1)	7(2)	25(2)	10(2)	-3(2)	0	0
C(2)	13(2)	21(2)	12(2)	-4(2)	1(2)	-2(2)
O(2)	9(1)	40(2)	15(2)	2(1)	-1(1)	0(1)
N(3)	9(2)	31(2)	6(2)	0(2)	-2(1)	0(2)
C(4)	13(3)	21(4)	9(3)	-3(2)	0	0
O(4)	16(2)	34(2)	15(2)	2(2)	0	0
N(4)	15(3)	26(3)	19(3)	0	0	3(2)
N(5)	10(3)	31(4)	18(3)	0	0	-4(2)

Table S40. Principal mean square atomic displacements U in A^2 for 4. T = 148 K.

0.0252	0.0133	0.0067	Ni1
0.0256	0.0090	0.0072	N1
0.0229	0.0126	0.0103	C2
0.0404	0.0147	0.0087	O2
0.0305	0.0103	0.0049	N3
0.0218	0.0130	0.0086	C4
0.0338	0.0159	0.0147	O4
0.0266	0.0185	0.0139	N4
0.0315	0.0179	0.0091	N5

Table S41. Hydrogen coordinates (x 10^4) and isotropic displacement parameters (A^2 x 10^3) for 4. T = 148 K.

	x	y	z	U(eq)
H(3)	3450(4)	2630(5)	4510(3)	21(12)
H(41)	6050(4)	4640(8)	7120(3)	49(18)
H(42)	6700(9)	3640(17)	7500	90(5)
H(51)	6570(4)	-90(7)	7920(3)	26(14)
H(52)	5960(7)	-1390(13)	7500	50(3)

Table S42. Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters (x 100) for crystal **5**. U(eq) is defined as one third of the trace of the orthogonalized Uij tensor. T = 11-16 K.

	x	y	z	U(eq)
Ni(1)	5000	1580(3)	7500	0.49
N(1)	5000	1666(2)	6060(2)	0.56
C(2)	4023(1)	1948(2)	5632(2)	0.41
O(2)	3097(1)	1670(2)	5967(2)	0.70
N(3)	4043(1)	2583(2)	4798(1)	0.62
C(4)	5000	2948(3)	4357(3)	0.43
O(4)	5000	3540(4)	3622(3)	0.89
N(4)	6178(1)	3699(2)	7500	0.74
N(5)	6213(2)	-461(2)	7500	0.67
H(3)	3293(3)	2817(4)	4520(3)	1.68
H(41)	6085(4)	4538(6)	8005(5)	3.76
H(42)	6924(4)	3196(8)	7500	4.71
H(51)	6691(4)	-241(6)	6972(5)	3.13
H(52)	5920(5)	-1762(7)	7500	4.37

Table S43. Anisotropic displacement parameters (x 100) for **5**. T = 11-16 K.

	U11	U22	U33	U12	U13	U23
Ni(1)	0.21(9)	0.42(10)	0.83(26)	0	0	0
N(1)	0.19(7)	0.80(8)	0.68(20)	0	0	0.16(7)
C(2)	0.27(7)	0.66(8)	0.30(22)	-0.01(5)	0.16(9)	0.14(6)
O(2)	0.33(8)	1.23(9)	0.54(25)	0.05(7)	-0.06(10)	0.09(8)
N(3)	0.49(5)	0.83(6)	0.53(15)	-0.01(4)	0.12(7)	-0.03(5)
C(4)	0.37(10)	0.48(11)	0.44(33)	0	0	0.18(9)
O(4)	1.03(13)	0.95(14)	0.71(36)	0	0	0.32(13)
N(4)	0.47(9)	0.87(9)	0.88(27)	-0.13(6)	0	0
N(5)	0.51(9)	0.94(9)	0.55(23)	0.25(6)	0	0
H(3)	1.11(17)	2.07(17)	1.9(5)	0.03(11)	-0.03(18)	-0.22(14)
H(41)	4.88(24)	3.37(24)	3.0(6)	-1.55(19)	0.26(30)	-1.26(23)
H(42)	1.24(26)	3.00(31)	9.9(11)	0.44(22)	0	0
H(51)	2.42(21)	5.10(26)	1.9(6)	0.70(17)	0.55(28)	0.11(22)
H(52)	2.82(28)	1.59(26)	8.7(10)	-0.10(22)	0	0

Table S44. Bond lengths (Å) and angles (deg) for **5**. T = 11-16 K.

Ni(1)-N(5)	2.0749(21)
Ni(1)-N(4)	2.0867(21)
Ni(1)-N(1)	2.2566(28)
N(1)-C(2)	1.3690(22)
C(2)-O(2)	1.2479(26)
C(2)-N(3)	1.3845(29)
N(3)-C(4)	1.3689(23)
N(3)-H(3)	1.015(4)
C(4)-O(4)	1.228(5)
N(4)-H(41)	1.003(6)
N(4)-H(42)	.968(6)
N(5)-H(51)	2.801(5)
N(5)-H(52)	1.004(6)
N(5)-Ni(1)-N(5)	89.39(13)
N(5)-Ni(1)-N(4)	92.51(7)
N(5)-Ni(1)-N(4)	178.10(10)
N(4)-Ni(1)-N(4)	85.59(12)
N(5)-Ni(1)-N(1)	91.12(4)
N(4)-Ni(1)-N(1)	88.84(4)
N(1)-Ni(1)-N(1)	176.8(14)
C(2)-N(1)-C(2)	118.43(26)
C(2)-N(1)-Ni(1)	119.57(13)
O(2)-C(2)-N(1)	122.46(24)
O(2)-C(2)-N(3)	117.78(19)
N(1)-C(2)-N(3)	119.76(17)
C(4)-N(3)-C(2)	123.66(18)
C(4)-N(3)-H(3)	119.93(34)
C(2)-N(3)-H(3)	116.38(31)
O(4)-C(4)-N(3)	122.62(30)
N(3)-C(4)-N(3)	114.63(29)
Ni(1)-N(4)-H(41)	111.59(27)
Ni(1)-N(4)-H(42)	111.28(30)
H(41)-N(4)-H(42)	109.1(5)
Ni(1)-N(5)-H(51)	106.58(25)
Ni(1)-N(5)-H(52)	114.8(4)
H(51)-N(5)-H(52)	110.13(32)