Table S-1. ^1H NMR data for urea ligands and their complexes with the $[(en_2)_2Co]^{3+}$ fragment. NMR's were recorded in DMSO-$^2H_6$. All shifts are in ppm vs TMS and, unless noted, refer to a single hydrogen. The subscripts in this table refer to the carbon or nitrogen in Figure S-1 to which the hydrogen(s) is bonded. In addition, all complexes exhibit broad or multiple signals in the range of 4.0 - 6.0 and 2.0 - 4.0 ppm arising from the NH_2 groups and the CH_2 linkages of the ethylenediamine ligands, respectively.

Ligands	H_1	H_2	H_3	H_4	H_6	H_7	H_9	H_{11}	H_{12}	H_{13}
1	8.5 (d)	7.8 (t)	7.3 (m)	7.3 (m)	4.3 (d-2H))	6.6(t)	5.7 (s-2H)			
3a (CH_3)	8.52 (d)	7.76 t)	7.26 (t)	7.34 (m)	4.43 (d-2H)	6.74 (t)	8.69 (s)	7.34 (m-2H)	7.03 (d-2H)	2.21* (s-3H)
3b (H)	8.52 (d)	7.74 (t)	7.23 (m)	7.35 (d)	4.46 (d-2H)	6.82 (t)	8.82 (s)	7.47 (d 2H)	7.23 (m-2H)	6.90 (t)
3c (Br)	8.47 (d)	7.11 (t)	7.23 (m)	7.35 (m)	4.37 (d-2H)	6.77 (t)	8.90 (s)	7.35 (m-2H)	7.35 (m-2H)	
3d (3-Cl))	8.49 (d)	7.74 (t)	7.24 (t)	7.34 (m)	4.39 (d-2H)	6.80 (t)	8.92 (s)	7.34 (m-2H)	7.34 (m)	7.34 (m)
3e (CF_3)	8.53 (d)	7.79 (t)	7.28(t)	7.36 (d)	4.46 (d-2H)	6.92 (t)	9.23 (s)	7.60 (m-2H)	7.60 (m-2H)	
3f (NO_2)	8.55 (d)	7.79 (t)	7.30 (t)	7.38 (d)	4.50 (d-2H)	7.11 (t)	9.62 (s)	7.68 (d-2H)	8.17 (d-2H)	
Complexes						**H_{en}**				
2	8.45 (d)	8.03 (t)	7.46 (m)	7.46 (m)	4.63 (q-2H)	9.61 (s)	5.10 (s-2H)			
3a (CH_3)	8.53 (d)	8.07 (t)	7.60 (m)	7.60 (m)	4.85 (q-2H)	9.19 (s)	8.04 (s)	7.34 (d-2H)	7.01 (d-2H)	
3b (H)	8.53 (d)	8.09 (t)	7.60 (m)	7.60 (m)	4.86 (q-2H)	9.10 (s)	8.14 (s)	7.45 (d-2H)	7.21 (t-2H)	6.88(t)
3c (Br)	8.53 (d)	8.08 (t)	7.61 (m)	7.61 (m)	4.85 (q-2H)	8.99 (s)	8.29 (s)	7.46 (d-2H)	7.38 (d-2H)	
3d (3-Cl)	8.53 (d)	8.08 (t)	7.61 (m)	7.61 (m)	4.85 (q-2H)	8.90 (s)	8.35 (s)	6.90 (d) 7.74 (s)	7.22 (t)	7.36 (d)
3e (CF_3)	8.55 (d)	8.10 (t)	7.63 (m)	7.60 (m)	4.85 (q-2H)	8.82 (s)	8.57 (s)	7.69 (d-2H)	7.60 (m-2H)	
3f (NO_2)	8.55 (d)	8.10 (t)	7.63 (m)	7.63 (m)	4.85 (q-2H)	8.58 (s)	8.95 (s)	8.15 (d-2H)	7.73 (d-2H)	
4	8.57 (d)	8.12 (t)	7.64 (t)	7.75 (d)	4.3 (t-2H)					

*Hydrogens on X.

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Table S-2. ^{13}C NMR data for urea ligands and their complexes with the $[(en_2)_2Co]^{3+}$ fragment in DMSO-$^2H_6$. All shifts are in ppm vs TMS. See Figure S-1 for the numbering scheme. In addition to the urea ligand resonances, all of the complexes show 4 singlets in the region from 50 to 43 ppm due to the ethylenediamine ligands of the $[(en)_2Co]^{3+}$ fragment.

Ligands	C_1	C_2	C_3	C_4	C_5	C_6	C_8	C_{10}	$C_{11/11'}$	$C_{12/12'}$	C_{13}	C_{14}
1	148.7	120.9	136.7	122.0	158.8	44.9	159.8					
3a (CH_3)	148.7	122.0	136.7	121.1	155.4	44.6	159.0	137.9	129.1	117.8	129.8	20.3
3b (H)	148.7	121.1	136.7	121.1	155.3	44.7	158.9	140.5	128.7	122.0	117.7	
3c (Br)	148.7	122.0	136.7	121.1	155.0	44.5	158.6	139.9	131.3	119.5	112.3	
3d (3-Cl)[a]	148.7	122.1	136.7	121.1	155.0	44.6	158.7	139.0	131.4	131.4	119.5	
3e (CF_3)	148.7	122.1	136.7	121.1	154.8	44.5	158.5	144.1	119.5 125.9	112.4 117.2	122.3	120.8 (q)
3f (NO_2)	148.8	122.2	136.7	121.2	154.5	44.2	158.2	147.1	125.1	116.9	140.5	
Complexes												
2	148.4	125.5	140.4	122.6	165.4	57.4	167.4					
3a (CH_3)	150.0	124.0	139.6	121.5	162.5	56.7	167.0	138.7	128.5	120.0	129.8	20.4
3b (H)	151.1	124.0	139.6	121.5	162.4	56.7	166.9	141.3	128.1	119.9	121.1	
3c (Br)	150.0	124.0	139.6	121.6	161.9	56.5	166.8	140.7	130.7	121.5	112.4	
3d (3-Cl)[a]	151.1	125.1	140.7	121.5	162.7	57.6	167.8	143.9	130.7	133.6	118.7	
3e (CF_3)	149.9	123.9	139.5	121.4	161.4	56.4	166.6	144.9	125.6 125.2	120.1 118.8	126.4	120.5 (q)
3f (NO_2)	150.2	124.1	139.8	121.6	160.9	56.3	166.5	140.1	124.6	118.3	148.1	
4	150.8	125.2	140.6	122.9	165.4	49.2						

[a]Assignments of the 3-chloro ligand and complex should be considered as tentative. We have not performed the necessary 2-D experiment needed to unambiguously assign the phenyl group resonances.

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Table S-3. Values of k_{obsd} for the acid hydrolysis of 3.

X = -CH_3	T = 55°C	X = H	T = 55°C
$[H^+]$, M	k_{obsd}, s^{-1}	$[H^+]$, M	k_{obsd}, s^{-1}
0.99	0.00264	0.98	0.00473
0.99	0.00265	0.98	0.00451
0.99	0.00277	0.98	0.00437
0.80	0.00246	0.75	0.00396
0.80	0.00236	0.75	0.00390
0.80	0.00269	0.75	0.00442
0.60	0.00208	0.50	0.00322
0.60	0.00220	0.50	0.00325
0.60	0.00205	0.50	0.00328
0.50	0.00170	0.25	0.00204
0.50	0.00204	0.25	0.00208
0.50	0.00206	0.25	0.00209
0.25	0.00130	0.10	0.000941
0.25	0.00131	0.10	0.00103
0.25	0.00128	0.10	0.000975
0.10	0.000647	0.05	0.000515
0.10	0.000626	0.05	0.000557
0.10	0.000583	0.05	0.000485

X = -Br	T = 55°C	X = CF_3	T = 55°C
$[H^+]$, M	k_{obsd}, s^{-1}	$[H^+]$, M	k_{obsd}, s^{-1}
1.00	0.00588	0.99	0.0111
1.00	0.00603	0.99	0.0109
1.00	0.00603	0.99	0.0121
0.87	0.00564	0.80	0.00961
0.87	0.00559	0.80	0.0108
0.87	0.00544	0.80	0.00996
0.77	0.00529	0.60	0.00768
0.77	0.00511	0.60	0.00761
0.77	0.00512	0.60	0.00826
0.51	0.00394	0.50	0.00671
0.51	0.00439	0.50	0.00689

X = -Br	T = 55°C
$[H^+]$, M	k_{obsd}, s^{-1}
0.51	0.00389
0.26	0.00232
0.26	0.00231
0.26	0.00231
0.10	0.00102
0.10	0.00105
0.10	0.00105
0.05	0.000541
0.05	0.000511
0.05	0.000516

X = NO_2	T = 55°C
$[H^+]$, M	k_{obsd}, s^{-1}
0.87	0.0134
0.87	0.0133
0.87	0.0140
0.77	0.0120
0.77	0.0118
0.77	0.0116
0.51	0.00873
0.51	0.00866
0.51	0.00898
0.26	0.00474
0.26	0.00476
0.26	0.00481
0.10	0.00200
0.10	0.00198
0.10	0.00199
0.05	0.00100
0.05	0.000998
0.05	0.000992

X = CF_3	T = 55°C
$[H^+]$, M	k_{obsd}, s^{-1}
0.50	0.00667
0.25	0.00371
0.25	0.00368
0.25	0.00381
0.10	0.00150
0.10	0.00164
0.10	0.00164

X = NO_2	T = 45°C
$[H^+]$, M	k_{obsd}, s^{-1}
1.02	0.00535
0.87	0.00471
0.77	0.00424
0.51	0.00300
0.26	0.00154
0.10	0.000637
0.05	0.000321

X = NO_2	T = 35°C
$[H^+]$, M	k_{obsd}, s^{-1}
1.02	0.00161
0.87	0.00140
0.77	0.00125
0.51	0.000896
0.26	0.000445
0.10	0.000192
0.05	0.0000947

X = NO_2	T = 25°C
$[H^+]$, M	k_{obsd}, s^{-1}
1.02	0.000438
0.87	0.000371
0.77	0.000335
0.51	0.000229
0.26	0.000121
0.10	0.0000947

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Table S-4. Crystal Data for $[Co(C_2H_8N_2)_2(C_7H_8N_2O)](ClO_4)_2$.

formula	$C_{11}H_{24}Cl_2CoN_7O_9$
fw	528.19
system	monoclinic
space group	$P2_1/c$
a, Å	9.743(1)
b, Å	13.924(3)
c, Å	15.006(4)
β,°	97.07(1)
V, Å^3	2020.2(6)
Z	4
T,°C	21(1)
ρ_{calc}, g cm^{-3}	1.736
μ(Mo Kα), cm^{-1}	17.27
crystal size, mm	0.28 x 0.18 x 0.28

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Table S-5. Data Collection and Refinement for $[Co(C_2H_8N_2)_2(C_7H_8N_2O)](ClO_4)_2$.

diffractometer	Philips PW 1100/20
radiation	Mo Kα, graphite monochrometer
λ of radiation, Å	0.70930 (α_1)
scan mode	θ-2θ
θ scan width,°	0.90 + 0.346tanθ
θ scan speed,° min^{-1}	1.0
background	stationary counts of 10s at each side of every scan
2θ range,°	4 - 50
unique reflections	4665
observed reflections[a]	3454
variables	271
final R_F[b]	0.037[c]
final R_{wF}[d]	0.051[c]
final GOF[e]	1.32[c]

a Reflections with $I > 3\sigma(I)$

b $R_F = \Sigma| |F_o| - |F_c| |/|F_o|$

c Observed reflections only

d $R_{wF} = [\Sigma w(|F_o| - |F_c|)^2 / \Sigma w|F_o|^2]^{1/2}$

e GOF = $[\Sigma w(|F_o| - |F_c|)^2 /$ (no. obs - no. vars)$]^{1/2}$

Table S-6. Final Temperature Factors for the Non-Hydrogen Atoms of $[Co(C_2H_8N_2)_2(C_7H_8N_2O)](ClO_4)_2$.[a]

Atom	U/U_{11}	U_{22}	U_{33}	U_{12}	U_{13}	U_{23}
Co	0.0238(2)	0.0279(2)	0.0313(2)	0.0020(1)	0.0060(1)	0.0005(1)
N(1)	0.029(1)	0.029(1)	0.035(1)	0.000(1)	0.005(1)	0.005(1)
N(2)	0.031(1)	0.026(1)	0.046(1)	0.000(1)	0.015(1)	0.000(1)
N(3)	0.028(1)	0.047(1)	0.040(1)	0.000(1)	0.010(1)	0.005(1)
N(4)	0.034(1)	0.036(1)	0.038(1)	0.003(1)	0.004(1)	0.001(1)
N(5)	0.033(1)	0.042(1)	0.041(1)	0.006(1)	0.001(1)	-0.006(1)
N(6)	0.034(1)	0.042(1)	0.035(1)	-0.001(1)	0.006(1)	-0.006(1)
C(1)	0.042(2)	0.029(1)	0.052(2)	0.002(1)	0.004(1)	0.006(1)
C(2)	0.049(2)	0.035(2)	0.062(2)	-0.008(1)	-0.004(2)	0.015(2)
C(3)	0.039(2)	0.054(2)	0.068(2)	-0.014(2)	0.011(2)	0.016(2)
C(4)	0.034(2)	0.048(2)	0.068(2)	-0.003(1)	0.018(2)	0.008(2)
C(5)	0.026(1)	0.035(1)	0.037(1)	0.000(1)	0.004(1)	0.002(1)
C(6)	0.028(1)	0.031(1)	0.049(2)	0.002(1)	0.010(1)	0.002(1)
C(7)	0.044(2)	0.031(2)	0.084(3)	-0.004(1)	0.027(2)	-0.003(2)
C(8)	0.048(2)	0.108(3)	0.039(2)	-0.007(2)	0.005(2)	0.020(2)
C(9)	0.050(2)	0.086(3)	0.042(2)	-0.010(2)	-0.001(2)	0.020(2)
C(10)	0.052(2)	0.055(2)	0.057(2)	0.022(2)	-0.014(2)	-0.002(2)
C(11)	0.043(2)	0.057(2)	0.052(2)	0.009(2)	-0.007(2)	-0.005(2)
O(1A)	0.043(1)					
O(1B)	0.047(1)					
N(7)	0.062(2)	0.025(1)	0.121(3)	-0.002(1)	0.042(2)	-0.009(2)
Cl(1)	0.0461(4)	0.0401(4)	0.0391(4)	0.0040(3)	0.0089(3)	0.0001(3)
Cl(2)	0.0342(4)	0.0401(4)	0.0459(3)	-0.0049(3)	0.0090(3)	-0.0014(3)
O(11)	0.211(5)	0.053(2)	0.061(2)	0.029(2)	0.018(2)	0.016(1)
O(12)	0.090(2)	0.063(2)	0.071(2)	0.026(2)	0.035(2)	-0.001(1)
O(13)	0.132(3)	0.094(2)	0.051(2)	0.024(2)	-0.015(2)	-0.014(2)
O(14)	0.066(2)	0.106(3)	0.250(6)	-0.021(2)	0.068(3)	-0.059(3)
O(21)	0.059(2)	0.035(1)	0.069(2)	-0.003(1)	0.017(1)	-0.005(1)
O(22)	0.098(2)	0.055(2)	0.075(2)	-0.024(2)	0.036(2)	-0.027(1)
O(23)	0.032(1)	0.121(3)	0.129(3)	-0.014(2)	0.006(2)	-0.003(2)
O(24)	0.076(2)	0.079(2)	0.070(2)	-0.003(2)	0.001(2)	0.033(2)

a. Anisotropic temperature factors ($Å^2$) in the form: $-2\pi^2(\underline{U}_{11}\underline{h}^2\underline{a}^{*2} + ... + 2\underline{U}_{12}\underline{hka}^*\underline{b}^* + ...)$

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Table S-7. Final Fractional Coordinates and Isotropic Temperature Factors for the Hydrogen Atoms of $[Co(C_2H_8N_2)_2(C_7H_8N_2O)](ClO_4)_2$.[a]

Atom	*x/a*	*y/b*	*z/c*	$B(Å^2)$
H(N7A)	0.185	0.714	0.829	6.1
H(N7B)	0.072	0.633	0.850	6.1
H(N3A)	0.517	0.429	0.719	3.6
H(N3B)	0.432	0.508	0.723	3.6
H(N4A)	0.114	0.365	0.669	3.5
H(N4B)	0.212	0.303	0.641	3.5
H(N5A)	0.488	0.271	0.728	3.7
H(N5B)	0.382	0.213	0.751	3.7
H(N6A)	0.392	0.390	0.933	3.5
H(N6B)	0.464	0.458	0.889	3.5
H(1)	0.235	0.171	0.834	3.9
H(2)	0.048	0.103	0.889	4.6
H(3)	-0.129	0.200	0.929	5.1
H(4)	-0.117	0.366	0.904	4.6
H(6A)	0.070	0.502	0.888	3.3
H(6B)	0.004	0.485	0.790	3.3
H(8A)	0.428	0.384	0.588	6.3
H(8B)	0.416	0.495	0.580	6.3
H(9A)	0.196	0.492	0.609	5.7
H(9B)	0.205	0.413	0.538	5.7
H(10A)	0.592	0.203	0.849	5.3
H(10B)	0.457	0.209	0.894	5.3
H(11A)	0.594	0.328	0.958	4.9
H(11B)	0.629	0.363	0.865	4.9

a. Coordinates for H(N7A) and H(N7B) were from a difference map, the remainder were calculated geometrically.

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Table S-8. Final Fractional Coordinates for the Non-Hydrogen Atoms of $[Co(C_2H_8N_2)_2(C_7H_8N_2O)](ClO_4)_2$.

Co	0.31428(4)	0.37619(3)	0.77719(2)
N(1)	0.1716(2)	0.3069(2)	0.8341(2)
N(2)	0.2081(2)	0.4885(2)	0.8032(2)
N(3)	0.4345(2)	0.4497(2)	0.7080(2)
N(4)	0.1987(2)	0.3587(2)	0.6614(2)
N(5)	0.4327(3)	0.2610(2)	0.7667(2)
N(6)	0.4370(3)	0.3999(2)	0.8881(2)
C(1)	0.1620(3)	0.2108(2)	0.8481(2)
C(2)	0.0515(4)	0.1700(2)	0.8815(2)
C(3)	-0.0534(3)	0.2270(3)	0.9039(3)
C(4)	-0.0463(3)	0.3247(2)	0.8893(2)
C(5)	0.0663(3)	0.3622(2)	0.8537(2)
C(6)	0.0772(3)	0.4669(2)	0.8347(2)
C(7)	0.2497(3)	0.5801(2)	0.8090(3)
C(8)	0.3885(4)	0.4399(3)	0.6104(2)
C(9)	0.2360(4)	0.4328(3)	0.5976(2)
C(10)	0.5138(4)	0.2416(3)	0.8557(2)
C(11)	0.5592(3)	0.3363(3)	0.8962(2)
O(1A)[a]	0.3796(5)	0.6033(3)	0.8099(3)
O(1B)[a]	0.3539(5)	0.6088(3)	0.7697(4)
N(7)	0.1671(3)	0.6479(2)	0.8383(3)
Cl(1)	0.25275(8)	0.39817(5)	0.09532(5)
Cl(2)	0.75701(7)	0.09262(5)	0.11039(5)
O(11)	0.2448(5)	0.4744(2)	0.0348(2)
O(12)	0.3324(3)	0.3216(2)	0.0647(2)
O(13)	0.3059(4)	0.4301(3)	0.1818(2)
O(14)	0.1220(4)	0.3583(3)	0.1017(4)
O(21)	0.7035(3)	-0.0031(2)	0.1105(2)
O(22)	0.7174(3)	0.1430(2)	0.1868(2)
O(23)	0.9025(3)	0.0902(3)	0.1131(3)
O(24)	0.6986(3)	0.1407(2)	0.0312(2)

a. Occupancy 0.50