Table S1. Crystallographic data.[a]

Compound	$[Co(HCONH_2)_2(HCO_2)_2]_x$
Formula	$C_4H_8CoN_2O_6$
fw	239.05
Color, habit	pink, prism
Crystal size, mm	0.25 x 0.30 x 0.35
Crystal system	monoclinic
Space group	*C*2/*c* (No. 15)
a, Å	12.772(1)
b, Å	8.354(1)
c, Å	8.243(1)
β, deg	93.19(1)
V, Å^3	878.2(2)
Z	4
ρ_{calc}, g/cm^3	1.808
F(000)	484
Radiation	Mo
μ, cm^{-1}	19.57
Transmission factors (relative)	0.78-1.00
Scan type	ω–2θ
Scan range, deg in ω	1.21 + 0.35 tan θ
Scan speed, deg/min	16 (up to 9 scans)
Data collected	$+h, \pm k, \pm l$
$2\theta_{max}$, deg	75
Crystal decay, %	negligible
Total reflections	2508
Unique reflections	2423
R_{merge}	0.027
Reflections with $I \geq 3\sigma(I)$	1354
No. of variables	84
R	0.030
R_w	0.031
gof	1.74
Max Δ/σ (final cycle)	0.002
Residual density, e/Å^3	-0.38, +0.26

[a] Temperature 294 K, Rigaku AFC6S diffractometer, Mo K_α radiation (λ = 0.71069 Å), graphite monochromator, takeoff angle 6.0°, aperture 6.0 x 6.0 mm at a distance of 285 mm from the crystal, stationary background counts at each end of the scan (scan/background time ratio 2:1), $\sigma^2(F^2) = [S^2(C + 4B)]/Lp^2$ (S = scan rate, C = scan count, B = normalized background count), function minimized $\Sigma w(|F_o|-|F_c|)^2$ where $w = 4F_o^2/\sigma^2(F_o^2)$, $R = \Sigma||F_o|-|F_c||/\Sigma|F_o|$, $R_w = (\Sigma w(|F_o|-|F_c|)^2/\Sigma w|F_o|^2)^{1/2}$, and gof = $[\Sigma w(|F_o|-|F_c|)^2/(m-n)]^{1/2}$. Values given for R, R_w, and gof are based on those reflections with $I \geq 3\sigma(I)$.

Table S2. Atomic coordinates and B_{eq}

atom	x	y	z	B_{eq}	occ.
Co(1)	0.2500	0.2500	0.5000	1.786(6)	
O(1)	0.2053(1)	0.4736(2)	0.4087(2)	2.75(3)	
O(2)	0.2112(1)	0.6668(2)	0.2275(2)	2.51(3)	
O(3)	0.0942(1)	0.1638(2)	0.4635(2)	3.92(4)	
N(1)	-0.0732(4)	0.1966(7)	0.5257(8)	5.4(1)	0.52
N(1a)	-0.0075(10)	0.359(1)	0.507(2)	5.3(3)	0.19
N(1b)	-0.074(1)	0.257(3)	0.438(2)	5.5(4)	0.17
N(1c)	-0.012(2)	0.250(3)	0.621(3)	5.3(4)	0.12
C(1)	0.2356(2)	0.5322(2)	0.2810(2)	2.40(3)	
C(2)	0.0169(2)	0.2377(4)	0.5031(4)	4.86(7)	

$$B_{eq} = \frac{8}{3}\pi^2(U_{11}(aa^*)^2 + U_{22}(bb^*)^2 + U_{33}(cc^*)^2 + 2U_{12}aa^*bb^*\cos\gamma + 2U_{13}aa^*cc^*\cos\beta + 2U_{23}bb^*cc^*\cos\alpha)$$

Table S3. Bond Lengths(Å)*

atom	atom	distance	atom	atom	distance
Co(1)	O(1)	2.082(1)	Co(1)	O(2)^b	2.085(1)
Co(1)	O(3)	2.122(2)	O(1)	C(1)	1.242(2)
O(2)	C(1)	1.242(2)	O(3)	C(2)	1.224(3)
N(1)	C(2)	1.225(6)	N(1a)	C(2)	1.06(1)
N(1b)	C(2)	1.26(1)	N(1c)	C(2)	1.06(2)

Table S4. Bond Angles(°)*

atom	atom	atom	angle	atom	atom	atom	angle
O(1)	Co(1)	O(1)^a	180.0	O(1)	Co(1)	O(2)^b	92.74(5)
O(1)	Co(1)	O(2)^c	87.26(5)	O(1)	Co(1)	O(3)	90.97(7)
O(1)	Co(1)	O(3)^a	89.03(7)	O(2)^b	Co(1)	O(2)^c	180.0
O(2)^b	Co(1)	O(3)	91.57(6)	O(2)^b	Co(1)	O(3)^a	88.43(6)
O(3)	Co(1)	O(3)^a	180.0	Co(1)	O(1)	C(1)	124.6(1)
Co(1)^d	O(2)	C(1)	123.6(1)	Co(1)	O(3)	C(2)	123.7(2)
O(1)	C(1)	O(2)	125.1(2)	O(3)	C(2)	N(1)	132.6(4)
O(3)	C(2)	N(1a)	137.1(8)	O(3)	C(2)	N(1b)	133.3(8)
O(3)	C(2)	N(1c)	128(1)				

*Superscripts refer to symmetry operations: (a) 1/2-x, 1/2-y, 1-z

(b) 1/2-x, -1/2+y, 1/2-z (c) x, 1-y, 1/2+z (d) x, 1-y, -1/2+z

Table S5. Hydrogen atom coordinates and B_{iso}

atom	x	y	z	B_{iso}	occ.
H(1)	0.2818	0.4677	0.2182	2.9	
H(2)	0.0295	0.3511	0.5200	5.8	0.52
H(2a)	-0.0365	0.1661	0.5404	5.8	0.19
H(2b)	0.0277	0.2926	0.6063	5.8	0.17
H(3)	-0.1275	0.2779	0.5378	6.5	0.52
H(3a)	-0.0808	0.3856	0.5251	6.4	0.19
H(3b)	-0.1241	0.3228	0.4882	6.1	0.17
H(4)	-0.0908	0.0828	0.5312	6.5	0.52
H(4a)	0.0435	0.4457	0.4941	6.4	0.19
H(4b)	-0.0920	0.2137	0.3324	6.1	0.17

Table S6. Anisotropic Displacement Parameters

atom	U_{11}	U_{22}	U_{33}	U_{12}	U_{13}	U_{23}
Co(1)	0.0284(2)	0.0175(1)	0.0226(1)	0.0006(2)	0.00716(10)	-0.0002(2)
O(1)	0.0487(8)	0.0261(6)	0.0309(6)	0.0070(6)	0.0128(6)	0.0080(5)
O(2)	0.0451(8)	0.0237(6)	0.0274(6)	0.0035(6)	0.0108(5)	0.0043(5)
O(3)	0.0290(8)	0.0450(9)	0.076(1)	-0.0030(7)	0.0111(7)	-0.0178(9)
N(1)	0.030(2)	0.062(3)	0.115(5)	-0.009(2)	0.025(3)	-0.043(4)
N(1a)	0.035(6)	0.046(7)	0.12(1)	0.008(6)	0.014(6)	0.007(7)
N(1b)	0.023(5)	0.13(2)	0.055(8)	0.019(10)	0.003(6)	-0.04(1)
C(1)	0.039(1)	0.0250(8)	0.0277(8)	0.0051(8)	0.0103(7)	0.0021(7)
C(2)	0.047(1)	0.066(2)	0.073(2)	0.014(2)	0.015(1)	0.005(2)

The general temperature factor expression:

$$\exp(-2\pi^2(a^{*2}U_{11}h^2 + b^{*2}U_{22}k^2 + c^{*2}U_{33}l^2 + 2a^*b^*U_{12}hk + 2a^*c^*U_{13}hl + 2b^*c^*U_{23}kl))$$

Table S7. Bond Lengths(Å) Involving Hydrogen

atom	atom	distance	atom	atom	distance
N(1)	H(3)	0.98	N(1)	H(4)	0.98
N(1a)	H(3a)	0.98	N(1a)	H(4a)	0.98
N(1b)	H(3b)	0.96	N(1b)	H(4b)	0.96
C(1)	H(1)	0.97	C(2)	H(2a)	0.97
C(2)	H(2)	0.97	C(2)	H(2b)	0.97

Table S8. Bond Angles(°) Involving Hydrogen

atom	atom	atom	angle	atom	atom	atom	angle
C(2)	N(1)	H(3)	119.8	C(2)	N(1)	H(4)	120.0
H(3)	N(1)	H(4)	120.2	C(2)	N(1a)	H(3a)	120.2
C(2)	N(1a)	H(4a)	120.0	H(3a)	N(1a)	H(4a)	119.8
C(2)	N(1b)	H(3b)	120.5	C(2)	N(1b)	H(4b)	120.5
H(3b)	N(1b)	H(4b)	118.8	O(1)	C(1)	H(1)	117.4
O(2)	C(1)	H(1)	117.5	O(3)	C(2)	H(2a)	111.4
O(3)	C(2)	H(2)	113.6	O(3)	C(2)	H(2b)	113.3
N(1)	C(2)	H(2)	113.7	N(1a)	C(2)	H(2a)	111.5
N(1b)	C(2)	H(2b)	113.3				

Table S9. Torsion Angles(°)

atom	atom	atom	atom	angle	atom	atom	atom	atom	angle
Co(1)	O(1)	C(1)	O(2)	-178.3(1)	Co(1)	O(3)	C(2)	N(1)	-161.1(5)
Co(1)	O(3)	C(2)	N(1a)	35(1)	Co(1)	O(3)	C(2)	N(1b)	141(1)
Co(1)	O(3)	C(2)	N(1c)	-78(1)	O(1)	Co(1)	O(3)	C(2)	-46.3(2)
O(1)	Co(1)	O(3)^a	C(2)^a	-133.7(2)	O(3)	Co(1)	O(1)	C(1)	-114.2(2)
O(3)	Co(1)	O(1)^a	C(1)^a	-65.8(2)					

Table S10. Non-bonded Contacts out to 3.60 Å

atom	atom	distance	ADC	atom	atom	distance	ADC
O(1)	N(1a)	3.00(1)	56603	O(1)	N(1b)	3.12(2)	56603
O(1)	N(1)	3.292(6)	56603	O(1)	N(1c)	3.38(2)	56603
O(1)	N(1)	3.479(6)	5	O(2)	O(3)	2.934(2)	56404
O(2)	N(1c)	2.98(2)	56603	O(2)	N(1)	2.991(5)	56603
O(2)	O(3)	3.015(2)	6	O(2)	N(1c)	3.02(2)	56404
O(2)	C(2)	3.115(3)	56404	O(2)	C(1)	3.128(2)	6
O(2)	N(1b)	3.25(2)	5	O(2)	N(1a)	3.25(1)	56404
O(2)	N(1b)	3.41(1)	56603	O(2)	N(1a)	3.50(1)	56603
O(2)	N(1)	3.596(6)	5	O(3)	N(1)	3.024(6)	55603
O(3)	N(1b)	3.39(1)	2	N(1)	N(1c)	3.09(2)	55602
N(1)	C(1)	3.372(6)	44505	N(1)	C(1)	3.513(6)	56603
N(1a)	N(1a)	2.36(2)	56603	N(1a)	N(1c)	3.20(3)	55602
N(1a)	C(2)	3.37(1)	56603	N(1a)	N(1b)	3.40(3)	56603
N(1a)	N(1c)	3.44(3)	56603	N(1a)	C(1)	3.59(1)	56603
N(1b)	C(1)	3.28(2)	44505	N(1b)	C(1)	3.52(2)	2
N(1c)	N(1c)	2.14(4)	55602	N(1c)	C(2)	3.11(2)	55602
N(1c)	C(1)	3.52(2)	56603	Co(1)	H(4b)	3.33	2
Co(1)	H(4b)	3.33	8	O(1)	H(3a)	2.08	56603
O(1)	H(3b)	2.19	56603	O(1)	H(3)	2.35	56603
O(1)	H(4)	2.89	5	O(2)	H(3)	2.31	56603
O(2)	H(2b)	2.52	56404	O(2)	H(1)	2.55	6
O(2)	H(4b)	2.64	5	O(2)	H(3b)	2.65	56603
O(2)	H(3a)	2.74	56603	O(2)	H(2)	2.81	56404
O(2)	H(4a)	2.95	56404	O(3)	H(4)	2.06	55603

Table S10. Non-bonded Contacts out to 3.60 Å (continued)

atom	atom	distance	ADC	atom	atom	distance	ADC
O(3)	H(4b)	2.47	2	O(3)	H(2a)	2.85	55603
N(1)	H(1)	2.86	45508	N(1a)	H(4a)	1.69	56603
N(1a)	H(3a)	2.43	56603	N(1a)	H(2)	2.44	56603
N(1b)	H(4a)	2.57	56603	N(1c)	H(2b)	2.30	55602
N(1c)	H(4a)	2.73	56603	N(1c)	H(2a)	2.91	55602
C(1)	H(3)	2.62	56603	C(1)	H(4b)	2.69	5
C(1)	H(3a)	2.70	56603	C(1)	H(3b)	2.72	56603
C(1)	H(4)	2.97	5	C(2)	H(4a)	2.76	56603
C(2)	H(4)	2.86	55603	C(2)	H(4b)	2.98	2
H(1)	H(4)	2.34	55408	H(2)	H(4a)	1.94	56603
H(2)	H(3a)	2.33	56603	H(2)	H(2)	2.62	56603
H(2b)	H(4a)	2.49	56603	H(2b)	H(2b)	2.51	55602
H(3)	H(4a)	2.57	56603	H(3a)	H(4a)	1.50	56603
H(3b)	H(4a)	2.19	56603	H(4a)	H(4a)	1.44	56603

The ADC (atom designator code) specifies the position of an atom in a crystal. The 5-digit number shown in the table is a composite of three one-digit numbers and one two-digit number: TA (first digit) + TB (second digit) + TC (third digit) + SN (last two digits). TA, TB and TC are the crystal lattice translation digits along cell edges a, b and c. A translation digit of 5 indicates the origin unit cell. If TA = 4, this indicates a translation of one unit cell length along the a-axis in the negative direction. Each translation digit can range in value from 1 to 9 and thus ±4 lattice translations from the origin (TA=5, TB=5, TC=5) can be represented.

The SN, or symmetry operator number, refers to the number of the symmetry operator used to generate the coordinates of the target atom. A list of symmetry operators relevant to this structure are given below.

For a given intermolecular contact, the first atom (origin atom) is located in the origin unit cell and its position can be generated using the identity operator (SN=1). Thus, the ADC for an origin atom is always 55501. The position of the second atom (target atom) can be generated using the ADC and the coordinates of the atom in the parameter table. For example, an ADC of 47502 refers to the target atom moved through symmetry operator two, then translated -1 cell translations along the a axis, +2 cell translations along the b axis, and 0 cell translations along the c axis.

An ADC of 1 indicates an intermolecular contact between two fragments (eg. cation and anion) that reside in the same asymmetric unit.

Symmetry Operators:

(1)	x,	y,	z	(2)	-x,	y,	1/2-z
(3)	-x,	-y,	-z	(4)	x,	-y,	1/2+z
(5)	1/2+x,	1/2+y,	z	(6)	1/2-x,	1/2+y,	1/2-z
(7)	1/2-x,	1/2-y,	-z	(8)	1/2+x,	1/2-y,	1/2+z

Table S11. Hydrogen Bonds and C-H...O/N Interactions*

A	H	B	A-H	H...B	A...B	A-H...B
C(2)	H(2)	N(1a)a	0.97	2.44	3.369(14)	159
C(2)	H(2b)	N(1c)b	0.97	2.30	3.11(2)	140
C(2)	H(2b)	O(2)c	0.97	2.52	3.115(3)	120
N(1)	H(3)	O(2)a	0.98	2.31	2.992(5)	126
N(1)	H(3)	O(1)a	0.98	2.36	3.292(6)	160
N(1a)	H(3a)	O(1)a	0.98	2.08	3.001(13)	157
N(1b)	H(3b)	O(1)a	0.98	2.19	3.12(2)	164
N(1)	H(4)	O(3)a	0.98	2.06	3.025(6)	168
N(1a)	H(4a)	N(1a)a	0.98	1.70	2.36(2)	122
N(1a)	H(4a)	O(1)	0.98	2.23	3.034(13)	138
N(1b)	H(4b)	O(3)d	0.98	2.47	3.394(15)	162

*Symmetry operations: (a) -x, 1-y, 1-z (b) -x, y, 3/2-z

(c) x, 1-y, 1/2+z (d) -x, y, 1/2-z