Crystal structures of two hydrogen-bonded compounds of chloranilic acid–ethyleneurea (1/1) and chloranilic acid–hydantoin (1/2)

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The structures of the hydrogen-bonded 1:1 co-crystal of chloranilic acid (systematic name: 2,5-dichloro-3,6-dihydroxy-1,4-benzoquinone) with ethyleneurea (systematic name: imidazolidin-2-one), $C_6H_2Cl_2O_4 \cdot C_3H_6N_2O$, (I), and the 1:2 co-crystal of chloranilic acid with hydantoin (systematic name: imidazolidine-2,4-dione), $C_6H_2Cl_2O_4 \cdot 2C_3H_4N_2O_2$, (II), have been determined at 180 K. In the crystals of both compounds, the base molecules are in the lactam form and no acid–base interaction involving H-atom transfer is observed. The asymmetric unit of (I) consists of two independent half-molecules of chloranilic acid, with each of the acid molecules lying about an inversion centre, and one ethyleneurea molecule. The asymmetric unit of (II) consists of one half-molecule of chloranilic acid, which lies about an inversion centre, and one hydantoin molecule. In the crystal of (I), the acid and base molecules are linked via O—H⋯O and N—H⋯O hydrogen bonds, forming an undulating sheet structure parallel to the ab plane. In (II), the base molecules form an inversion dimer via a pair of N—H⋯O hydrogen bonds, and the base dimers are further linked through another N—H⋯O hydrogen bond into a layer structure parallel to ($\bar{1}$01). The acid molecule and the base molecule are linked via an O—H⋯O hydrogen bond.

1. Chemical context

Chloranilic acid, a dibasic acid with hydrogen-bond donor as well as acceptor groups, appears particularly attractive as a template for generating tightly bound self-assemblies with various organic bases, and also as a model compound for investigating hydrogen-transfer motions in O—H⋯N and N—H⋯O hydrogen-bonded systems (Zaman et al., 2004; Seliger et al., 2009; Asaji et al. 2010; Molčanov & Kojić-Prodić, 2010). In the present study, we have prepared two hydrogen-bonded compounds of chloranilic acid–ethyleneurea (1/1) and chloranilic acid–hydantoin (1/2) in order to extend our study on D—H⋯A hydrogen bonding (D = N, O, or C; A = N, O or Cl) in chloranilic acid–organic base systems (Gotoh & Ishida, 2017a,b, and references therein).

https://doi.org/10.1107/S205698901801561X **1727**
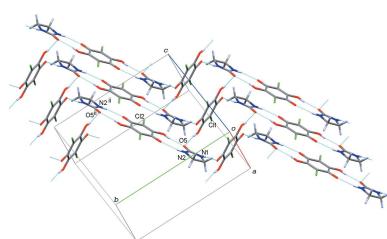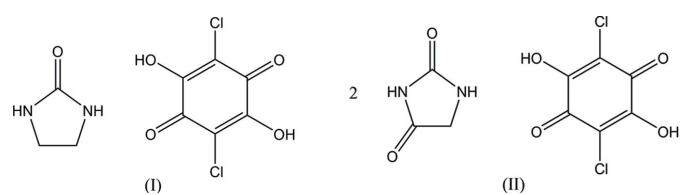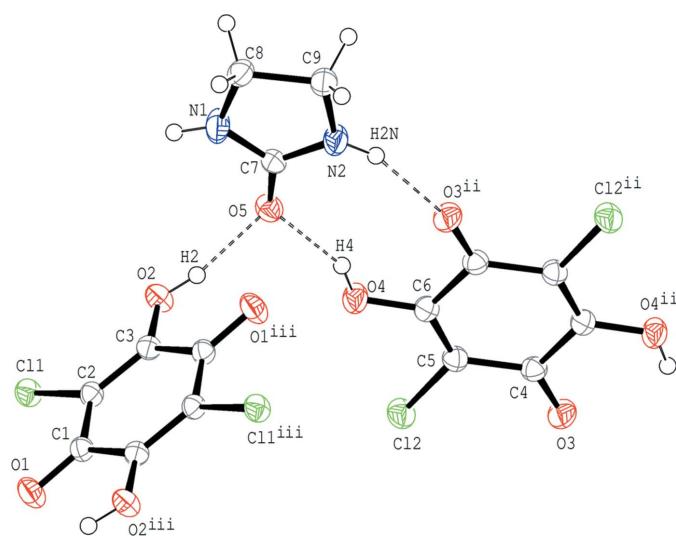
Figure 1
The molecular structure of compound (I), showing the atom-numbering scheme. Displacement ellipsoids of non-H atoms are drawn at the 50% probability level and H atoms are drawn as small spheres of arbitrary radii. O—H···O and N—H···O hydrogen bonds are shown by dashed lines. [Symmetry codes: (ii) $-x, -y + 1, -z + 1$; (iii) $-x + 1, -y, -z + 1$.]

2. Structural commentary

In compound (I), the base molecule is in the lactam form and no acid–base interaction involving H-atom transfer is observed (Fig. 1). In the asymmetric unit, there is one ethyleneurea molecule and two crystallographically independent half-molecules of chloranilic acid, with each of the acid molecules lying about an inversion centre. The O atom of ethyleneurea participates in two O—H···O hydrogen bonds as an acceptor for two O—H groups of chloranilic acid (O2—H2···O5 and O4—H4···O5; Table 1). The base ring (C7/N1/C8/C9/N2) is essentially planar and makes dihedral angles of 88.75 (6) and 3.27 (6)°, respectively, with the acid C1–C3/C1^{iii}–C3^{iii} and C4–C6/C4^{ii}–C6^{ii} rings [symmetry codes: (ii) $-x$, $-y + 1, -z + 1$; (iii) $-x + 1, -y, -z + 1$].

In compound (II), the base molecule is also in the lactam form and no acid–base interaction involving H-atom transfer is observed (Fig. 2). The chloranilic acid molecule is located on

Table 1
Hydrogen-bond geometry (Å, °) for (I).

$D—H···A$	$D—H$	$H···A$	$D···A$	$D—H···A$
O2—H2···O5	0.81 (2)	1.82 (2)	2.6090 (11)	164 (2)
O4—H4···O5	0.86 (2)	1.88 (2)	2.6635 (12)	151.0 (19)
N1—H1N···O1^i	0.85 (2)	2.06 (2)	2.9003 (15)	168 (2)
N2—H2N···O3^{ii}	0.85 (2)	2.06 (2)	2.8654 (15)	158.6 (17)

Symmetry codes: (i) $-x + 2, -y, -z + 1$; (ii) $-x, -y + 1, -z + 1$.

Table 2
Hydrogen-bond geometry (Å, °) for (II).

| $D—H···A$	$D—H$	$H···A$	$D···A$	$D—H···A$
O2—H2···O3	0.86 (2)	1.97 (2)	2.7917 (15)	160 (2)
N1—H1N···O3^i	0.91 (2)	2.00 (2)	2.8927 (13)	165 (2)
N2—H2N···O4^{ii}	0.91 (2)	1.85 (2)	2.7560 (14)	176 (2)

Symmetry codes: (i) $-x + 1, -y + 1, -z + 1$; (ii) $-x + \frac{1}{2}, y + \frac{1}{2}, -z + \frac{1}{2}$.

an inversion centre and the asymmetric unit consists of one hydantoin molecule and a half-molecule of chloranilic acid. The acid and base molecules are linked via an O—H···O hydrogen bond (O2—H2···O3; Table 2), forming a centrosymmetric 1:2 aggregate of the acid and the base. The 1:2 unit is approximately planar with a dihedral angle of 5.42 (5)° between the acid and base rings.

3. Supramolecular features

In the crystal of compound (I), the acid and base molecules are alternately arranged through O—H···O and N—H···O hydrogen bonds (O4—H4···O5, N1—H1N···O1^i, N2—H2H···O3^{ii}; symmetry codes as in Table 1), forming an undulating tape structure along [3$\overline{1}$0]. The tapes are stacked along the a axis via another O—H···O hydrogen bond (O2—H2—O5; Table 1) into a sheet structure parallel to the ab plane (Fig. 3).

In the crystal of (II), two adjacent base molecules, which are related by an inversion centre, form a dimer via a pair of N—H···O hydrogen bonds (N1—H1N···O3^i; symmetry code as

Figure 2
The molecular structure of compound (II), showing the atom-numbering scheme. Displacement ellipsoids of non-H atoms are drawn at the 50% probability level and H atoms are drawn as small spheres of arbitrary radii. O—H···O hydrogen bonds are shown by dashed lines. [Symmetry code: (iii) $-x + \frac{1}{2}, -y + \frac{5}{2}, -z + 1$.]
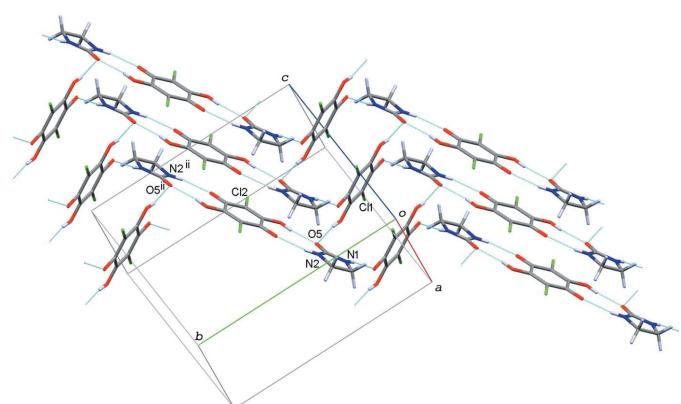
Figure 3
A partial packing diagram of compound (I), showing the undulating sheet structure formed by O—H···O and N—H···O hydrogen bonds (light-blue dotted lines). [Symmetry code: (ii) $-x, -y + 1, -z + 1$.]
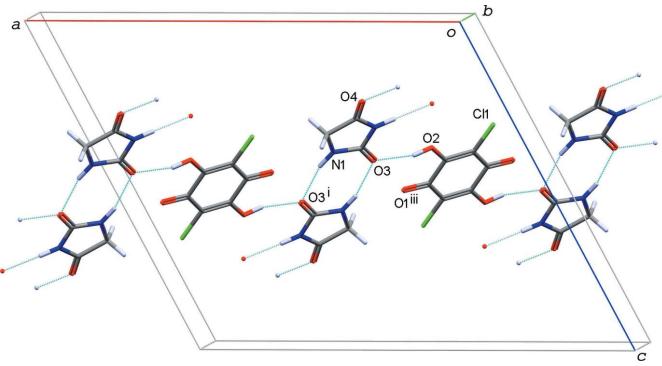
Figure 4
A partial packing diagram of compound (II) viewed approximately along the b axis, showing a hydrogen-bonded tape structure formed by acid molecules and pairs of base molecules. O—H···O and N—H···O hydrogen bonds are shown by light-blue dotted lines. [Symmetry codes: (i) $-x + 1$, $-y + 1$, $-z + 1$; (iii) $-x + \frac{1}{2}$, $-y + \frac{5}{2}$, $-z + 1$.]

Figure 5
A partial packing diagram of compound (II), showing hydrogen-bonding scheme in the layer formed by base molecules. N—H···O hydrogen bonds between the base molecules are shown by light-blue dotted lines, while O—H···O hydrogen bonds between the base and acid molecules are shown by red dotted lines. [Symmetry codes: (i) $-x + 1$, $-y + 1$, $-z + 1$; (ii) $-x + \frac{1}{2}$, $y + \frac{1}{2}$, $-z + \frac{1}{2}$; (iv) $-x + \frac{1}{2}$, $y - \frac{1}{2}$, $-z + \frac{1}{2}$.]

in Table 2), and the base dimer and the acid molecule are alternately linked through an O—H···O hydrogen bond (O2—H2···O3; Table 2), forming a flat tape structure along the a-axis direction (Fig. 4). The base dimers are assembled *via* another N—H···O hydrogen bond (N2—H2N···O4^{ii}; symmetry code as in Table 2), forming a layer parallel to ($\bar{1}$01) as shown in Fig. 5. The O—H···O hydrogen bond (O2—H2···O3; Table 2) formed between the acid and base molecules links the layers.

4. Database survey

A search of the Cambridge Structural Database (Version 5.39, last update August 2018; Groom *et al.*, 2016) for organic crystals of chloranilic acid with lactam-form base molecules gave ten hits. In the seven crystals of these compounds, O—H···O hydrogen bonds between the O—H group of chloranilic acid and the carbonyl group of base are observed

Table 3
Experimental details.

	(I)	(II)
Crystal data		
Chemical formula	$C_6H_2Cl_2O_4 \cdot C_3H_6N_2O$	$C_6H_2Cl_2O_4 \cdot 2C_3H_4N_2O_2$
M_r	295.08	409.14
Crystal system, space group	Monoclinic, $P2_1/c$	Monoclinic, $C2/c$
Temperature (K)	180	180
a, b, c (Å)	5.0180 (4), 14.6142 (10), 15.8882 (11)	19.5690 (8), 5.18661 (10), 16.6103 (3)
β (°)	105.563 (3)	117.965 (3)
V (Å^3)	1122.43 (15)	1489.03 (8)
Z	4	4
Radiation type	Mo $K\alpha$	Mo $K\alpha$
μ (mm^{-1})	0.59	0.49
Crystal size (mm)	$0.45 \times 0.29 \times 0.23$	$0.49 \times 0.33 \times 0.24$
Data collection		
Diffractometer	Rigaku R-AXIS RAPIDII	Rigaku R-AXIS RAPIDII
Absorption correction	Numerical (*NUMABS*; Higashi, 1999)	Numerical (*NUMABS*; Higashi, 1999)
T_{min}, T_{max}	0.716, 0.873	0.808, 0.888
No. of measured, independent and observed [$I > 2\sigma(I)$] reflections	21546, 3266, 3040	14622, 2181, 2029
R_{int}	0.057	0.072
$(\sin \theta/\lambda)_{max}$ (Å^{-1})	0.704	0.704
Refinement		
$R[F^2 > 2\sigma(F^2)]$, $wR(F^2)$, S	0.033, 0.092, 1.07	0.036, 0.100, 1.08
No. of reflections	3266	2181
No. of parameters	179	130
H-atom treatment	H atoms treated by a mixture of independent and constrained refinement	H atoms treated by a mixture of independent and constrained refinement
$\Delta\rho_{max}$, $\Delta\rho_{min}$ (e Å^{-3})	0.54, −0.31	0.44, −0.40

Computer programs: *RAPID-AUTO* (Rigaku, 2006), *SIR92* (Altomare *et al.*, 1993), *SHELXT2018* (Sheldrick, 2015a), *SHELXL2018* (Sheldrick, 2015b), *ORTEP-3* for Windows (Farrugia, 2012), *Mercury* (Macrae *et al.*, 2006), *CrystalStructure* (Rigaku, 2018) and *PLATON* (Spek, 2015).

[refcodes ACOJIO (Gotoh & Ishida, 2017a), AJAGIB (Luo & Palmore, 2002), HUFZUE (Jasinski *et al.*, 2010), ODIHIU, SADTIC, SADTOI and SADTUO (Gotoh & Ishida, 2011)]. In particular, the compounds of chloranilic acid with 2-pyridone (ACOJIO), gabapentin-lactum (HUFZUE), pyrrolidin-2-one (ODIHIU) and piperidin-2-one (SADTUO) show short O—H···O hydrogen bonds (O···O shorter than 2.5 Å). In the O—H···O hydrogen bond [O···O = 2.4484 (10) Å] of chloranilic acid–piperidin-2-one (1/2) (SADTUO), the H atom is disordered over two positions.

5. Synthesis and crystallization

Single crystals of compound (I) were obtained by slow evaporation from an acetonitrile solution (150 ml) of chloranilic acid (330 mg) with ethyleneurea (140 mg) at room temperature. Crystals of compound (II) were obtained by slow evaporation from an acetonitrile solution (250 ml) of chloranilic acid (350 mg) with hydantoin (340 mg) at room temperature.

6. Refinement

Crystal data, data collection and structure refinement details are summarized in Table 3. All H atoms in compounds (I) and (II) were found in difference Fourier maps. The O- and N-bound H atoms were freely refined. C-bound H atoms were positioned geometrically (C—H = 0.99 Å) and were treated as riding with $U_{iso}(H) = 1.2U_{eq}(C)$.

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Crystal structures of two hydrogen-bonded compounds of chloranilic acid–ethyleneurea (1/1) and chloranilic acid–hydantoin (1/2)

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Computing details

For both structures, data collection: *RAPID-AUTO* (Rigaku, 2006); cell refinement: *RAPID-AUTO* (Rigaku, 2006); data reduction: *RAPID-AUTO* (Rigaku, 2006). Program(s) used to solve structure: *SIR92* (Altomare *et al.*, 1993) for (I); *SHELXT2018* (Sheldrick, 2015*a*) for (II). For both structures, program(s) used to refine structure: *SHELXL2018* (Sheldrick, 2015*b*); molecular graphics: *ORTEP-3* for Windows (Farrugia, 2012) and *Mercury* (Macrae *et al.*, 2006); software used to prepare material for publication: *CrystalStructure* (Rigaku, 2018) and *PLATON* (Spek, 2015).

2,5-Dichloro-3,6-dihydroxy-1,4-benzoquinone–imidazolidin-2-one (1/1) (I)

Crystal data

$C_6H_2Cl_2O_4 \cdot C_3H_6N_2O$	$F(000) = 600.00$
$M_r = 295.08$	$D_x = 1.746$ Mg m^{-3}
Monoclinic, $P2_1/c$	Mo $K\alpha$ radiation, $\lambda = 0.71075$ Å
$a = 5.0180$ (4) Å	Cell parameters from 19204 reflections
$b = 14.6142$ (10) Å	$\theta = 3.0–30.1°$
$c = 15.8882$ (11) Å	$\mu = 0.59$ mm^{-1}
$\beta = 105.563$ (3)°	$T = 180$ K
$V = 1122.43$ (15) Å^3	Block, brown
$Z = 4$	$0.45 \times 0.29 \times 0.23$ mm

Data collection

Rigaku R-AXIS RAPIDII diffractometer	3266 independent reflections
	3040 reflections with $I > 2\sigma(I)$
Detector resolution: 10.000 pixels mm^{-1}	$R_{int} = 0.057$
ω scans	$\theta_{max} = 30.0°$, $\theta_{min} = 3.0°$
Absorption correction: numerical (NUMABS; Higashi, 1999)	$h = -7 \rightarrow 7$
	$k = -20 \rightarrow 19$
$T_{min} = 0.716$, $T_{max} = 0.873$	$l = -21 \rightarrow 22$
21546 measured reflections	

Refinement

Refinement on F^2	Primary atom site location: structure-invariant direct methods
$R[F^2 > 2\sigma(F^2)] = 0.033$	
$wR(F^2) = 0.092$	Secondary atom site location: difference Fourier map
$S = 1.07$	
3266 reflections	Hydrogen site location: mixed
179 parameters	H atoms treated by a mixture of independent and constrained refinement
0 restraints	

$w = 1/[\sigma^2(F_o^2) + (0.0573P)^2 + 0.2007P]$
  where $P = (F_o^2 + 2F_c^2)/3$
$(\Delta/\sigma)_{max} = 0.001$

$\Delta\rho_{max} = 0.54$ e Å^{-3}
$\Delta\rho_{min} = -0.31$ e Å^{-3}

Special details

Geometry. All esds (except the esd in the dihedral angle between two l.s. planes) are estimated using the full covariance matrix. The cell esds are taken into account individually in the estimation of esds in distances, angles and torsion angles; correlations between esds in cell parameters are only used when they are defined by crystal symmetry. An approximate (isotropic) treatment of cell esds is used for estimating esds involving l.s. planes.

Fractional atomic coordinates and isotropic or equivalent isotropic displacement parameters (Å^2)

	x	*y*	*z*	U_{iso}*/U_{eq}
Cl1	1.06653 (5)	0.00389 (2)	0.64704 (2)	0.02517 (9)
Cl2	0.28647 (6)	0.43040 (2)	0.69108 (2)	0.03101 (9)
O1	0.64834 (18)	−0.14266 (6)	0.60507 (6)	0.03198 (19)
O2	0.83758 (17)	0.15225 (5)	0.51887 (5)	0.02612 (17)
O3	−0.17821 (19)	0.55990 (6)	0.63245 (6)	0.03138 (19)
O4	0.43441 (18)	0.37722 (6)	0.52805 (6)	0.03039 (19)
O5	0.70481 (17)	0.29928 (5)	0.42370 (5)	0.02644 (17)
N1	0.8521 (2)	0.24152 (9)	0.30842 (7)	0.0372 (3)
N2	0.4933 (2)	0.33134 (8)	0.27881 (7)	0.0323 (2)
C1	0.5873 (2)	−0.07583 (7)	0.55745 (7)	0.02118 (19)
C2	0.7619 (2)	0.00483 (7)	0.56565 (7)	0.02058 (19)
C3	0.6846 (2)	0.07805 (7)	0.51226 (6)	0.02064 (19)
C4	−0.0902 (2)	0.53136 (7)	0.57314 (7)	0.0233 (2)
C5	0.1390 (2)	0.46653 (7)	0.58624 (7)	0.0238 (2)
C6	0.2263 (2)	0.43578 (7)	0.51801 (7)	0.0236 (2)
C7	0.6858 (2)	0.29121 (7)	0.34329 (7)	0.0224 (2)
C8	0.7865 (2)	0.24974 (8)	0.21442 (8)	0.0275 (2)
H8A	0.737465	0.189643	0.185683	0.033*
H8B	0.943169	0.276222	0.195803	0.033*
C9	0.5369 (3)	0.31466 (9)	0.19376 (8)	0.0325 (2)
H9A	0.578932	0.372186	0.166986	0.039*
H9B	0.372898	0.285493	0.153938	0.039*
H1N	0.989 (4)	0.2121 (14)	0.3405 (14)	0.059 (6)*
H2N	0.381 (4)	0.3687 (13)	0.2915 (12)	0.041 (5)*
H2	0.766 (4)	0.1933 (16)	0.4861 (14)	0.056 (6)*
H4	0.476 (4)	0.3613 (13)	0.4811 (13)	0.045 (5)*

Atomic displacement parameters (Å^2)

	U^{11}	U^{22}	U^{33}	U^{12}	U^{13}	U^{23}
Cl1	0.02251 (14)	0.02897 (15)	0.02081 (14)	−0.00049 (8)	0.00027 (10)	0.00183 (8)
Cl2	0.03685 (16)	0.03457 (16)	0.02165 (15)	0.00812 (10)	0.00789 (11)	0.00275 (9)
O1	0.0284 (4)	0.0277 (4)	0.0345 (4)	−0.0009 (3)	−0.0007 (3)	0.0114 (3)
O2	0.0273 (4)	0.0226 (4)	0.0252 (4)	−0.0031 (3)	0.0014 (3)	0.0039 (3)
O3	0.0355 (4)	0.0356 (4)	0.0262 (4)	0.0082 (3)	0.0138 (3)	−0.0009 (3)
O4	0.0333 (4)	0.0350 (4)	0.0243 (4)	0.0124 (3)	0.0100 (3)	0.0016 (3)

O5	0.0319 (4)	0.0244 (4)	0.0230 (4)	0.0017 (3)	0.0071 (3)	0.0006 (3)
N1	0.0378 (6)	0.0468 (6)	0.0272 (5)	0.0224 (5)	0.0093 (4)	0.0057 (4)
N2	0.0336 (5)	0.0411 (5)	0.0240 (5)	0.0163 (4)	0.0106 (4)	0.0045 (4)
C1	0.0207 (4)	0.0220 (4)	0.0204 (4)	0.0018 (3)	0.0047 (3)	0.0012 (3)
C2	0.0192 (4)	0.0236 (5)	0.0178 (4)	0.0012 (3)	0.0029 (3)	0.0002 (3)
C3	0.0215 (4)	0.0221 (4)	0.0181 (4)	0.0003 (3)	0.0049 (3)	−0.0006 (3)
C4	0.0253 (5)	0.0233 (5)	0.0225 (5)	−0.0001 (4)	0.0083 (4)	−0.0004 (4)
C5	0.0260 (5)	0.0245 (5)	0.0213 (5)	0.0019 (4)	0.0070 (4)	0.0010 (4)
C6	0.0244 (5)	0.0231 (5)	0.0240 (5)	0.0016 (3)	0.0077 (4)	0.0002 (3)
C7	0.0239 (4)	0.0191 (4)	0.0246 (5)	−0.0005 (3)	0.0070 (4)	0.0021 (3)
C8	0.0254 (5)	0.0315 (5)	0.0275 (5)	0.0034 (4)	0.0100 (4)	−0.0007 (4)
C9	0.0346 (6)	0.0416 (6)	0.0240 (5)	0.0125 (5)	0.0127 (4)	0.0075 (4)

Geometric parameters (Å, º)

Cl1—C2	1.7169 (10)	N2—C9	1.4464 (15)
Cl2—C5	1.7145 (11)	N2—H2N	0.849 (19)
O1—C1	1.2230 (13)	C1—C2	1.4537 (14)
O2—C3	1.3165 (12)	C1—C3[i]	1.5092 (14)
O2—H2	0.81 (2)	C2—C3	1.3560 (14)
O3—C4	1.2168 (13)	C4—C5	1.4612 (14)
O4—C6	1.3264 (12)	C4—C6[ii]	1.5045 (15)
O4—H4	0.86 (2)	C5—C6	1.3504 (15)
O5—C7	1.2609 (13)	C8—C9	1.5348 (15)
N1—C7	1.3338 (14)	C8—H8A	0.9900
N1—C8	1.4455 (15)	C8—H8B	0.9900
N1—H1N	0.85 (2)	C9—H9A	0.9900
N2—C7	1.3397 (14)	C9—H9B	0.9900
C3—O2—H2	114.1 (14)	C6—C5—Cl2	121.94 (8)
C6—O4—H4	115.8 (13)	C4—C5—Cl2	117.13 (8)
C7—N1—C8	112.92 (10)	O4—C6—C5	122.18 (10)
C7—N1—H1N	121.2 (15)	O4—C6—C4[ii]	117.46 (9)
C8—N1—H1N	125.7 (15)	C5—C6—C4[ii]	120.35 (9)
C7—N2—C9	112.46 (10)	O5—C7—N1	125.87 (10)
C7—N2—H2N	119.3 (12)	O5—C7—N2	125.24 (10)
C9—N2—H2N	127.5 (12)	N1—C7—N2	108.88 (10)
O1—C1—C2	123.17 (9)	N1—C8—C9	102.65 (9)
O1—C1—C3[i]	117.64 (9)	N1—C8—H8A	111.2
C2—C1—C3[i]	119.19 (8)	C9—C8—H8A	111.2
C3—C2—C1	121.28 (9)	N1—C8—H8B	111.2
C3—C2—Cl1	121.68 (8)	C9—C8—H8B	111.2
C1—C2—Cl1	117.03 (7)	H8A—C8—H8B	109.2
O2—C3—C2	122.48 (9)	N2—C9—C8	102.92 (9)
O2—C3—C1[i]	117.99 (9)	N2—C9—H9A	111.2
C2—C3—C1[i]	119.53 (9)	C8—C9—H9A	111.2
O3—C4—C5	123.19 (10)	N2—C9—H9B	111.2
O3—C4—C6[ii]	118.08 (10)	C8—C9—H9B	111.2

| C5—C4—C6ⁱⁱ | 118.73 (9) | H9A—C9—H9B | 109.1 |
| C6—C5—C4 | 120.92 (9) | | |

O1—C1—C2—C3	−179.04 (11)	C4—C5—C6—O4	179.73 (10)
C3ⁱ—C1—C2—C3	0.34 (16)	Cl2—C5—C6—O4	−1.31 (16)
O1—C1—C2—Cl1	−0.19 (14)	C4—C5—C6—C4ⁱⁱ	0.61 (17)
C3ⁱ—C1—C2—Cl1	179.19 (7)	Cl2—C5—C6—C4ⁱⁱ	179.57 (8)
C1—C2—C3—O2	179.32 (9)	C8—N1—C7—O5	−176.92 (10)
Cl1—C2—C3—O2	0.52 (15)	C8—N1—C7—N2	3.23 (15)
C1—C2—C3—C1ⁱ	−0.34 (16)	C9—N2—C7—O5	175.71 (11)
Cl1—C2—C3—C1ⁱ	−179.14 (7)	C9—N2—C7—N1	−4.43 (15)
O3—C4—C5—C6	178.94 (11)	C7—N1—C8—C9	−0.84 (14)
C6ⁱⁱ—C4—C5—C6	−0.60 (17)	C7—N2—C9—C8	3.73 (14)
O3—C4—C5—Cl2	−0.07 (15)	N1—C8—C9—N2	−1.64 (13)
C6ⁱⁱ—C4—C5—Cl2	−179.61 (8)		

Symmetry codes: (i) −x+1, −y, −z+1; (ii) −x, −y+1, −z+1.

Hydrogen-bond geometry (Å, º)

D—H···A	D—H	H···A	D···A	D—H···A
O2—H2···O5	0.81 (2)	1.82 (2)	2.6090 (11)	164 (2)
O4—H4···O5	0.86 (2)	1.88 (2)	2.6635 (12)	151.0 (19)
N1—H1N···O1ⁱⁱⁱ	0.85 (2)	2.06 (2)	2.9003 (15)	168 (2)
N2—H2N···O3ⁱⁱ	0.85 (2)	2.06 (2)	2.8654 (15)	158.6 (17)

Symmetry codes: (ii) −x, −y+1, −z+1; (iii) −x+2, −y, −z+1.

2,5-Dichloro-3,6-dihydroxy-1,4-benzoquinone–imidazolidine-2,4-dione (1/2) (II)

Crystal data
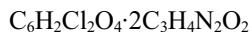
$C_6H_2Cl_2O_4·2C_3H_4N_2O_2$
$M_r = 409.14$
Monoclinic, $C2/c$
$a = 19.5690$ (8) Å
$b = 5.18661$ (10) Å
$c = 16.6103$ (3) Å
$\beta = 117.965$ (3)°
$V = 1489.03$ (8) Å³
$Z = 4$

$F(000) = 832.00$
$D_x = 1.825$ Mg m⁻³
Mo $K\alpha$ radiation, $\lambda = 0.71075$ Å
Cell parameters from 13670 reflections
$\theta = 3.3$–30.2°
$\mu = 0.49$ mm⁻¹
$T = 180$ K
Block, brown
$0.49 \times 0.33 \times 0.24$ mm

Data collection

Rigaku R-AXIS RAPIDII
  diffractometer
Detector resolution: 10.000 pixels mm⁻¹
ω scans
Absorption correction: numerical
  (NUMABS; Higashi, 1999)
$T_{min} = 0.808$, $T_{max} = 0.888$
14622 measured reflections

2181 independent reflections
2029 reflections with $I > 2\sigma(I)$
$R_{int} = 0.072$
$\theta_{max} = 30.0°$, $\theta_{min} = 4.1°$
$h = -27 \rightarrow 27$
$k = -7 \rightarrow 7$
$l = -22 \rightarrow 23$

Refinement

Refinement on F^2	Secondary atom site location: difference Fourier
$R[F^2 > 2\sigma(F^2)] = 0.036$	map
$wR(F^2) = 0.100$	Hydrogen site location: mixed
$S = 1.08$	H atoms treated by a mixture of independent
2181 reflections	and constrained refinement
130 parameters	$w = 1/[\sigma^2(F_o^2) + (0.0633P)^2 + 0.4572P]$
0 restraints	where $P = (F_o^2 + 2F_c^2)/3$
Primary atom site location: structure-invariant	$(\Delta/\sigma)_{max} < 0.001$
direct methods	$\Delta\rho_{max} = 0.44$ e Å^{-3}
	$\Delta\rho_{min} = -0.40$ e Å^{-3}

Special details

Geometry. All esds (except the esd in the dihedral angle between two l.s. planes) are estimated using the full covariance matrix. The cell esds are taken into account individually in the estimation of esds in distances, angles and torsion angles; correlations between esds in cell parameters are only used when they are defined by crystal symmetry. An approximate (isotropic) treatment of cell esds is used for estimating esds involving l.s. planes.

Refinement. Refinement was performed using all reflections. The weighted R-factor (wR) and goodness of fit (S) are based on F^2. R-factor (gt) are based on F. The threshold expression of $F^2 > 2.0$ sigma(F^2) is used only for calculating R-factor (gt).

Fractional atomic coordinates and isotropic or equivalent isotropic displacement parameters (Å^2)

	x	*y*	*z*	$U_{iso}*/U_{eq}$
Cl1	0.09683 (2)	1.02553 (6)	0.33978 (2)	0.02980 (12)
O1	0.11596 (5)	1.47273 (17)	0.45960 (6)	0.0313 (2)
O2	0.25729 (5)	0.82514 (16)	0.40485 (6)	0.02794 (19)
O3	0.39430 (5)	0.57413 (17)	0.43836 (6)	0.02824 (19)
O4	0.31705 (4)	−0.10296 (18)	0.23989 (6)	0.0296 (2)
N1	0.46359 (5)	0.26895 (19)	0.40528 (6)	0.0269 (2)
N2	0.33612 (5)	0.25929 (18)	0.32811 (6)	0.02336 (19)
C1	0.17655 (6)	1.3650 (2)	0.47556 (7)	0.0229 (2)
C2	0.18138 (6)	1.1410 (2)	0.42588 (7)	0.0228 (2)
C3	0.25018 (6)	1.0302 (2)	0.44779 (7)	0.0223 (2)
C4	0.39913 (6)	0.3869 (2)	0.39566 (7)	0.0226 (2)
C5	0.35871 (6)	0.0519 (2)	0.29613 (7)	0.0229 (2)
C6	0.44610 (6)	0.0494 (2)	0.34539 (8)	0.0255 (2)
H6A	0.466307	−0.112237	0.380266	0.031*
H6B	0.467588	0.070964	0.302420	0.031*
H1N	0.5127 (12)	0.308 (4)	0.4484 (15)	0.054 (5)*
H2	0.3043 (12)	0.772 (4)	0.4269 (14)	0.049 (5)*
H2N	0.2861 (11)	0.313 (4)	0.3055 (13)	0.045 (5)*

Atomic displacement parameters (Å^2)

	U^{11}	U^{22}	U^{33}	U^{12}	U^{13}	U^{23}
Cl1	0.02000 (16)	0.03480 (18)	0.02881 (17)	−0.00423 (8)	0.00662 (12)	−0.00241 (9)
O1	0.0206 (4)	0.0327 (4)	0.0358 (4)	0.0052 (3)	0.0091 (3)	−0.0012 (3)
O2	0.0237 (4)	0.0269 (4)	0.0314 (4)	0.0015 (3)	0.0114 (3)	−0.0036 (3)
O3	0.0219 (4)	0.0300 (4)	0.0288 (4)	0.0015 (3)	0.0085 (3)	−0.0058 (3)

O4	0.0198 (4)	0.0324 (4)	0.0321 (4)	−0.0038 (3)	0.0085 (3)	−0.0086 (3)
N1	0.0160 (4)	0.0312 (5)	0.0288 (4)	−0.0018 (3)	0.0067 (3)	−0.0071 (4)
N2	0.0158 (4)	0.0268 (4)	0.0239 (4)	0.0003 (3)	0.0063 (3)	−0.0025 (3)
C1	0.0191 (4)	0.0254 (5)	0.0226 (4)	0.0009 (3)	0.0084 (4)	0.0032 (4)
C2	0.0177 (4)	0.0257 (5)	0.0220 (4)	−0.0009 (3)	0.0069 (3)	0.0020 (4)
C3	0.0205 (4)	0.0232 (5)	0.0226 (4)	0.0002 (3)	0.0094 (4)	0.0021 (3)
C4	0.0180 (4)	0.0256 (5)	0.0221 (4)	−0.0010 (3)	0.0076 (3)	0.0003 (4)
C5	0.0173 (4)	0.0262 (5)	0.0239 (5)	−0.0002 (3)	0.0086 (4)	−0.0002 (4)
C6	0.0162 (4)	0.0276 (5)	0.0295 (5)	−0.0008 (3)	0.0079 (4)	−0.0049 (4)

Geometric parameters (Å, º)

Cl1—C2	1.7094 (10)	N2—C5	1.3620 (14)
O1—C1	1.2222 (12)	N2—C4	1.3857 (13)
O2—C3	1.3237 (13)	N2—H2N	0.912 (18)
O2—H2	0.86 (2)	C1—C2	1.4536 (15)
O3—C4	1.2318 (14)	C1—C3^{i}	1.5035 (14)
O4—C5	1.2117 (13)	C2—C3	1.3475 (14)
N1—C4	1.3425 (13)	C5—C6	1.5106 (14)
N1—C6	1.4436 (14)	C6—H6A	0.9900
N1—H1N	0.91 (2)	C6—H6B	0.9900
C3—O2—H2	112.8 (14)	O2—C3—C1^{i}	116.58 (9)
C4—N1—C6	111.82 (8)	C2—C3—C1^{i}	120.65 (9)
C4—N1—H1N	125.3 (13)	O3—C4—N1	127.75 (10)
C6—N1—H1N	122.5 (13)	O3—C4—N2	124.29 (9)
C5—N2—C4	111.40 (8)	N1—C4—N2	107.95 (9)
C5—N2—H2N	124.3 (12)	O4—C5—N2	126.87 (10)
C4—N2—H2N	124.2 (12)	O4—C5—C6	126.41 (10)
O1—C1—C2	123.86 (10)	N2—C5—C6	106.71 (9)
O1—C1—C3^{i}	117.46 (10)	N1—C6—C5	102.06 (8)
C2—C1—C3^{i}	118.67 (8)	N1—C6—H6A	111.4
C3—C2—C1	120.68 (9)	C5—C6—H6A	111.4
C3—C2—Cl1	121.94 (8)	N1—C6—H6B	111.4
C1—C2—Cl1	117.37 (7)	C5—C6—H6B	111.4
O2—C3—C2	122.77 (10)	H6A—C6—H6B	109.2
O1—C1—C2—C3	−179.33 (10)	C6—N1—C4—N2	−1.64 (12)
C3^{i}—C1—C2—C3	−0.32 (16)	C5—N2—C4—O3	−176.84 (11)
O1—C1—C2—Cl1	0.48 (15)	C5—N2—C4—N1	2.61 (12)
C3^{i}—C1—C2—Cl1	179.49 (7)	C4—N2—C5—O4	176.75 (11)
C1—C2—C3—O2	179.56 (9)	C4—N2—C5—C6	−2.45 (12)
Cl1—C2—C3—O2	−0.24 (15)	C4—N1—C6—C5	0.19 (12)
C1—C2—C3—C1^{i}	0.32 (16)	O4—C5—C6—N1	−177.85 (11)
Cl1—C2—C3—C1^{i}	−179.48 (7)	N2—C5—C6—N1	1.35 (11)
C6—N1—C4—O3	177.78 (11)		

Symmetry code: (i) −x+1/2, −y+5/2, −z+1.

Hydrogen-bond geometry (Å, º)

D—H···A	D—H	H···A	D···A	D—H···A
O2—H2···O3	0.86 (2)	1.97 (2)	2.7917 (15)	160 (2)
N1—H1N···O3[ii]	0.91 (2)	2.00 (2)	2.8927 (13)	165 (2)
N2—H2N···O4[iii]	0.91 (2)	1.85 (2)	2.7560 (14)	176 (2)

Symmetry codes: (ii) $-x+1$, $-y+1$, $-z+1$; (iii) $-x+1/2$, $y+1/2$, $-z+1/2$.