Acta Crystallographica Section C
Crystal Structure Communications
ISSN 0108-2701

Hydrogen-bonded structures of the isomeric compounds of phthalazine with 3-chloro-2-nitrobenzoic acid, 4-chloro-2-nitrobenzoic acid and 4-chloro-3-nitrobenzoic acid

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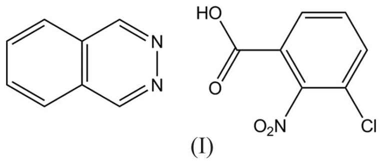
Received 28 September 2011
Accepted 26 October 2011
Online 31 October 2011

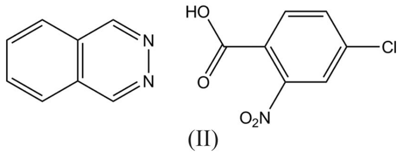
The structures of three isomeric compounds, $C_7H_4ClNO_4\cdot C_8H_6N_2$, of phthalazine with chloro- and nitro-substituted benzoic acid, namely, 3-chloro-2-nitrobenzoic acid–phthalazine (1/1), (I), 4-chloro-2-nitrobenzoic acid–phthalazine (1/1), (II), and 4-chloro-3-nitrobenzoic acid–phthalazine (1/1), (III), have been determined at 190 K. In the asymmetric unit of each compound, there are two crystallographically independent chloronitrobenzoic acid–phthalazine units, in each of which the two components are held together by a short hydrogen bond between an N atom of the base and a carboxyl O atom. In one hydrogen-bonded unit of (I) and in two units of (II), a weak $C-H\cdots O$ interaction is also observed between the two components. The $N\cdots O$ distances are 2.5715 (15) and 2.5397 (17) Å for (I), 2.5655 (13) and 2.6081 (13) Å for (II), and 2.613 (2) and 2.589 (2) Å for (III). In both hydrogen-bonded units of (I) and (II), the H atoms are each disordered over two positions with (N site):(O site) occupancies of 0.35 (3):0.65 (3) and 0.31 (3):0.69 (3) for (I), and 0.32 (3):0.68 (3) and 0.30 (3):0.70 (3) for (II). The H atoms in the hydrogen-bonded units of (III) are located at the O-atom sites.

Comment

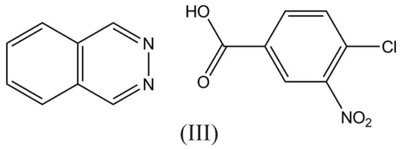
The hydrogen bonds formed between organic acids and organic bases vary from an $O-H\cdots N$ to an $O\cdots H-N$ type with increasing pK_a or proton affinities (PA) of the bases, and at an appropriate ΔpK_a [pK_a(base) − pK_a(acid)] the critical (inversion) point at which 50% proton transfer occurs, in the region of which a short strong hydrogen bond with a broad single minimum potential-energy curve for the H atom or a double-minimum potential is expected (Jerzykiewicz *et al.*, 1998; Kalenik *et al.*, 1989; Steiner *et al.*, 2001; Schmidtmann & Wilson, 2008; Gilli & Gilli, 2009). For the chloro- and nitro-substituted benzoic acid–pyridine derivative system, we have shown that the three isomeric compounds 3-chloro-2-nitrobenzoic acid–quinoline (1/1), (IV), 4-chloro-2-nitrobenzoic acid–quinoline (1/1), (V), and 5-chloro-2-nitrobenzoic acid–quinoline (1/1), (VI), are in a critical region and have a short double-well $N\cdots H\cdots O$ hydrogen bond between the pyridine N atom and the carboxyl group (Gotoh & Ishida, 2009). The (N site):(O site) occupancies of the H atom are 0.39 (3):0.61 (3), 0.47 (3):0.53 (3) and 0.65 (3):0.35 (3) for (IV), (V) and (VI), respectively, and the ΔpK_a values are 3.08, 2.93 and 3.04, respectively.



(I)



(II)



(III)

In this communication, we report the three isomeric compounds 3-chloro-2-nitrobenzoic acid–phthalazine (1/1), (I), 4-chloro-2-nitrobenzoic acid–phthalazine (1/1), (II), and 4-chloro-3-nitrobenzoic acid–phthalazine (1/1), (III), in order to extend our studies of the critical region and hydrogen bonding in the chloro- and nitro-substituted benzoic acid–pyridine derivative system (Gotoh & Ishida, 2007, 2009). Phthalazine is a weaker base (pK_a = 3.47) than quinoline (pK_a = 4.90), and the ΔpK_a values are 1.65, 1.50 and 0.12 for (I), (II) and (III), respectively.

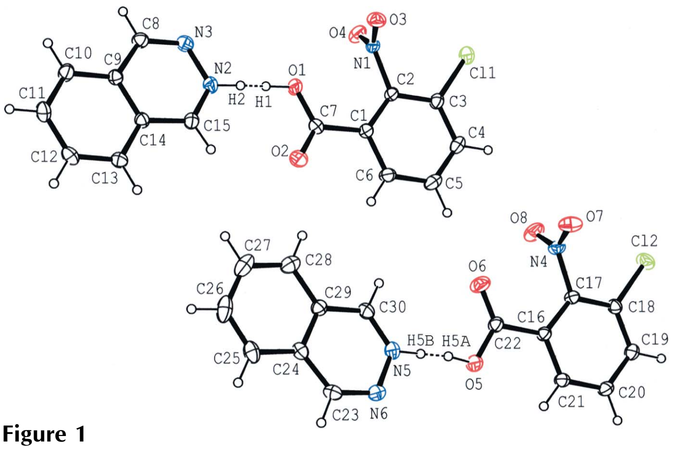


Figure 1
The asymmetric unit of (I), showing the atom-numbering scheme. Displacement ellipsoids are drawn at the 50% probability level. The disordered $O-H\cdots N/N-H\cdots O$ hydrogen bonds are indicated by dashed lines.

doi:10.1107/S0108270111044829

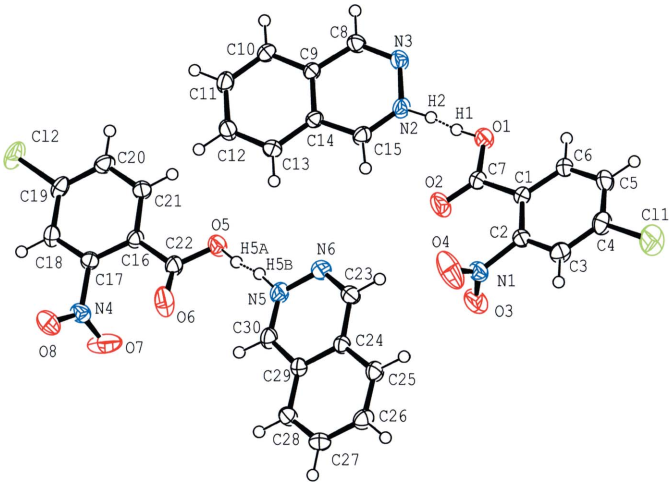


Figure 2
The asymmetric unit of (II), showing the atom-numbering scheme. Displacement ellipsoids are drawn at the 50% probability level. The disordered O—H···N/N—H···O hydrogen bonds are indicated by dashed lines.

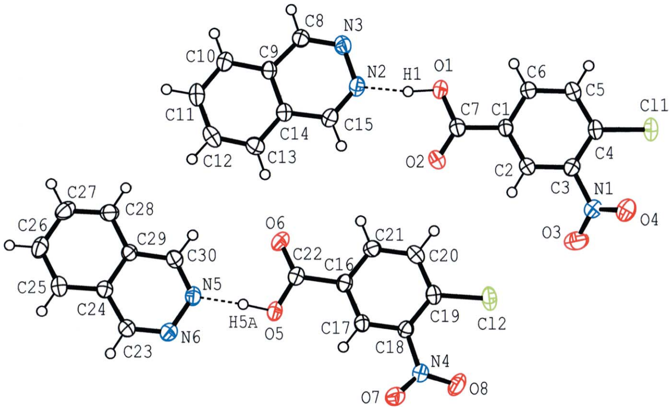


Figure 3
The asymmetric unit of (III), showing the atom-numbering scheme. Displacement ellipsoids are drawn at the 50% probability level. The O—H···N hydrogen bonds are indicated by dashed lines.

The molecular structures of compounds (I), (II) and (III) are shown in Figs. 1, 2 and 3, respectively. The asymmetric unit of each compound consists of two crystallographically independent chloronitrobenzoic acid–phthalazine units, where the two components are held together by a short hydrogen bond between one N atom of the base and the carboxyl group of the acid. The N···O distances are given in Tables 1–3. In the hydrogen bonds of (I) and (II), the H atoms were found to be disordered from difference Fourier maps (see supplementary figure), each of which shows two distinct peaks. The site occupancies refined to 0.35 (3):0.65 (3) (N2:O1) and 0.31 (3): 0.69 (3) (N5:O5) for (I), and 0.32 (3):0.68 (3) (N2:O1) and 0.30 (3):0.70 (3) (N5:O5) for (II). On the other hand, no disordered H atom was observed in (III), even though one of the O···N distances [2.589 (2) Å] is shorter than the O5···N5 distance [2.6081 (13) Å] in (I) and H atoms in the hydrogen bonds are located at an O-atom site (Table 3). In addition, a weak C—H···O interaction is observed in one hydrogen-bonded unit of (I) (C15—H15···O2; Table 1) and in each unit of (II) (C15—H15···O2 and C30—H30···O6; Table 2).

In the hydrogen-bonded units of (I) and (II), the benzene (C1–C6 or C16–C21) ring, the carboxyl (O1/C7/O2 or O5/C22/O6) plane and the phthalazine (N2/N3/C8–C15 or N5/N6/C23–C30) ring system are twisted slightly relative to each other. The dihedral angles between the C1–C6 and O1/C7/O2 planes, the C16–C21 and O5/C22/O6 planes, the O1/C7/O2 and N2/N3/C8–C15 planes, the O5/C22/O6 and N5/N6/C23–C30 planes, the C1–C6 and N2/N3/C8–C15 planes, and the C16–C21 and N5/N6/C23–C30 planes are 11.41 (14), 2.93 (15), 2.99 (14), 11.93 (15), 8.59 (5) and 9.02 (5)°, respectively, in (I), and 2.75 (17), 18.76 (17), 14.24 (16), 22.03 (16), 16.84 (5) and 9.56 (5)°, respectively, in (II). By contrast, in (III), these three planes are approximately coplanar with each other; the corresponding dihedral angles are 2.2 (2), 2.1 (3), 1.3 (2), 1.5 (3), 1.03 (8) and 0.66 (8)°, respectively. The planarity in both units of (III) may lead to a packing scheme accompanied by π–π stacking interactions in the crystal structure, as mentioned below.

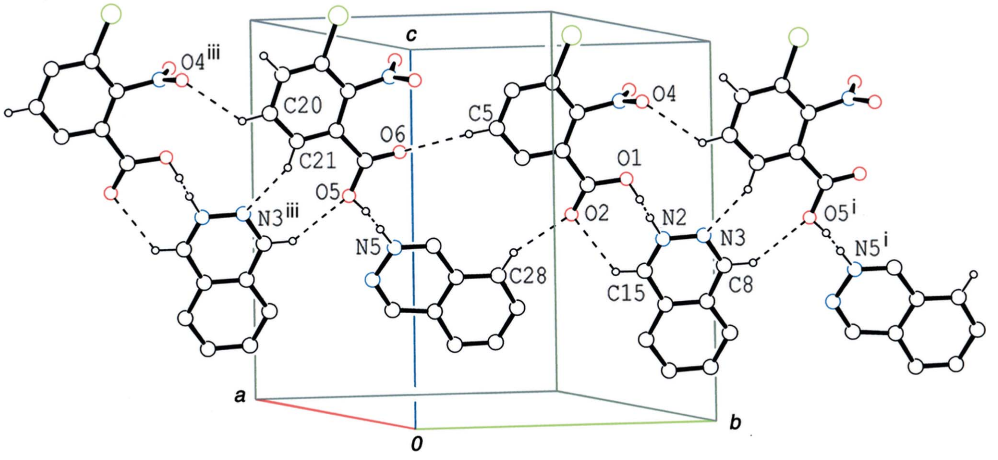


Figure 4
A packing diagram for (I), showing the hydrogen-bonded tape running along the $[1\overline{1}0]$ direction. H atoms not involved in the O···H···N, C—H···O and C—H···N hydrogen bonds (dashed lines) have been omitted. [Symmetry codes: (i) $x - 1, y + 1, z$; (iii) $x + 1, y - 1, z$.]

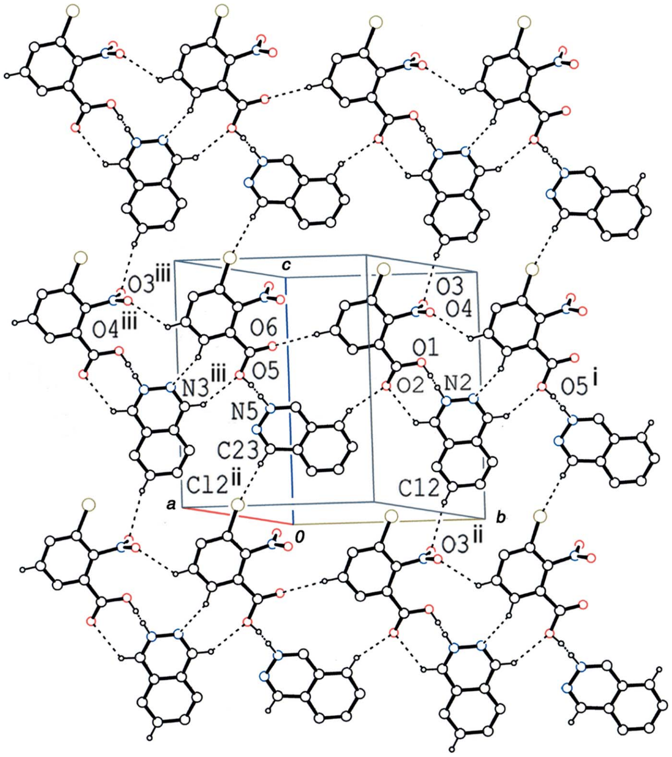


Figure 5
A packing diagram for (I), showing the hydrogen-bonded sheet structure parallel to the (110) plane. H atoms not involved in the O$\cdots$H$\cdots$N, C—H$\cdots$O, C—H$\cdots$N and C—H$\cdots$Cl hydrogen bonds (dashed lines) have been omitted. [Symmetry codes: (i) $x - 1, y + 1, z$; (ii) $x, y, z - 1$; (iii) $x + 1, y - 1, z$.]

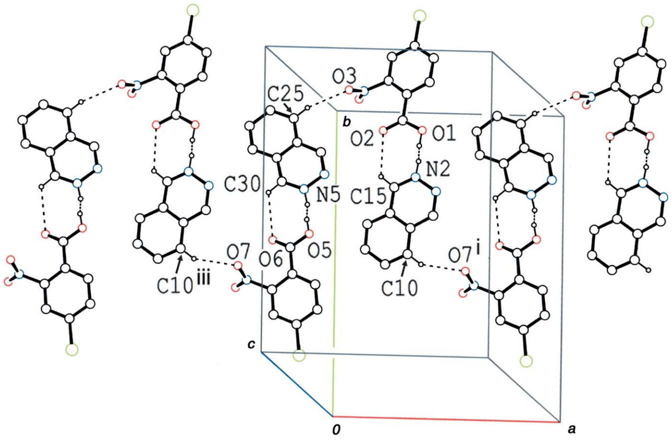


Figure 6
A packing diagram for (II), showing the hydrogen-bonded tape running along the *a* axis. Dashed lines indicate N$\cdots$H$\cdots$O and C—H$\cdots$O hydrogen bonds. H atoms not involved in the hydrogen bonds have been omitted. [Symmetry codes: (i) $x + 1, y, z$; (iii) $x - 1, y, z$.]

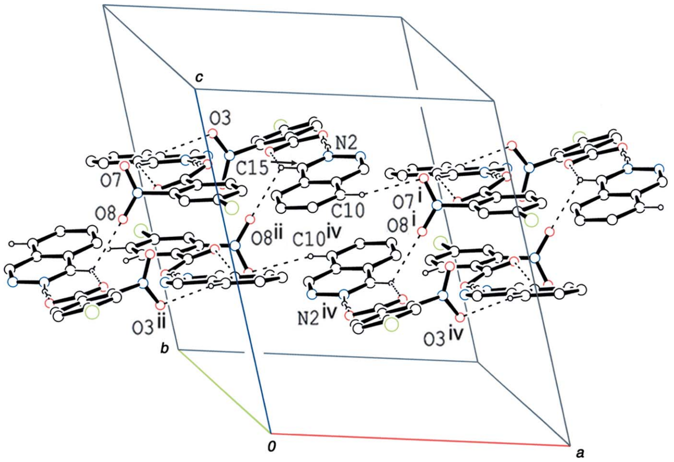


Figure 7
A packing diagram for (II), showing the double-tape structure. N$\cdots$H$\cdots$O and C—H$\cdots$O hydrogen bonds are indicated by dashed lines. H atoms not involved in the hydrogen bonds have been omitted. [Symmetry codes: (i) $x + 1, y, z$; (ii) $-x, -y + 1, -z + 1$; (iv) $-x + 1, -y + 1, -z + 1$.]

The packing diagrams for the three compounds are given in Figs. 4–9. In (I), the two crystallographically independent hydrogen-bonded units are arranged alternately *via* C—H$\cdots$O and C—H$\cdots$N hydrogen bonds (C5—H5$\cdots$O6, C8—H8$\cdots$O5^{i}, C20—H20$\cdots$O4^{iii}, C21—H21$\cdots$N3^{iii} and C28—H28$\cdots$O2; details and symmetry codes in Table 1), forming a tape structure along the [1$\overline{1}$0] direction (Fig. 4). These tapes are further linked *via* C—H$\cdots$O and C—H$\cdots$Cl hydrogen bonds (C12—H12$\cdots$O3^{ii} and C23—H23$\cdots$Cl2^{ii}; details and symmetry code in Table 1) into a sheet expanding parallel to the (110) plane (Fig. 5). Between these sheets, which are related to each other by an inversion centre, a C—H$\cdots$O interaction (C23—H23$\cdots$O3^{iv}; see Table 1 for symmetry code), and π–π stacking interactions between the C1–C6 and N2/N3/C8/C9/C14/C15 rings and the C16–C21 and C24–C29 rings, are observed. The centroid–centroid distances are in the range 3.5926 (9)–3.8215 (10) Å and the detailed geometries are given in Table 4.

In (II), there are fewer observable intermolecular hydrogen bonds than in (I), which may reflect the fact that it is a less

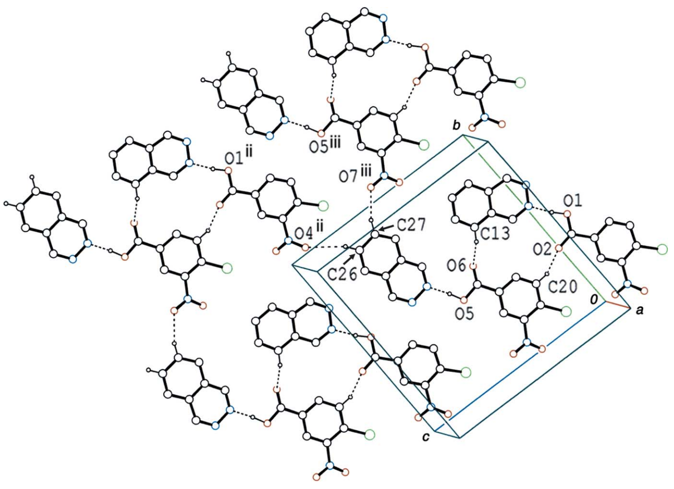


Figure 8
A packing diagram for (III), showing the hydrogen-bonded sheet structure parallel to the (101) plane. O—H$\cdots$N and C—H$\cdots$O hydrogen bonds are indicated by dashed lines. H atoms not involved in the hydrogen bonds have been omitted. [Symmetry codes: (ii) $x - 1, y + 1, z + 1$; (iii) $x, y + 1, z$.]

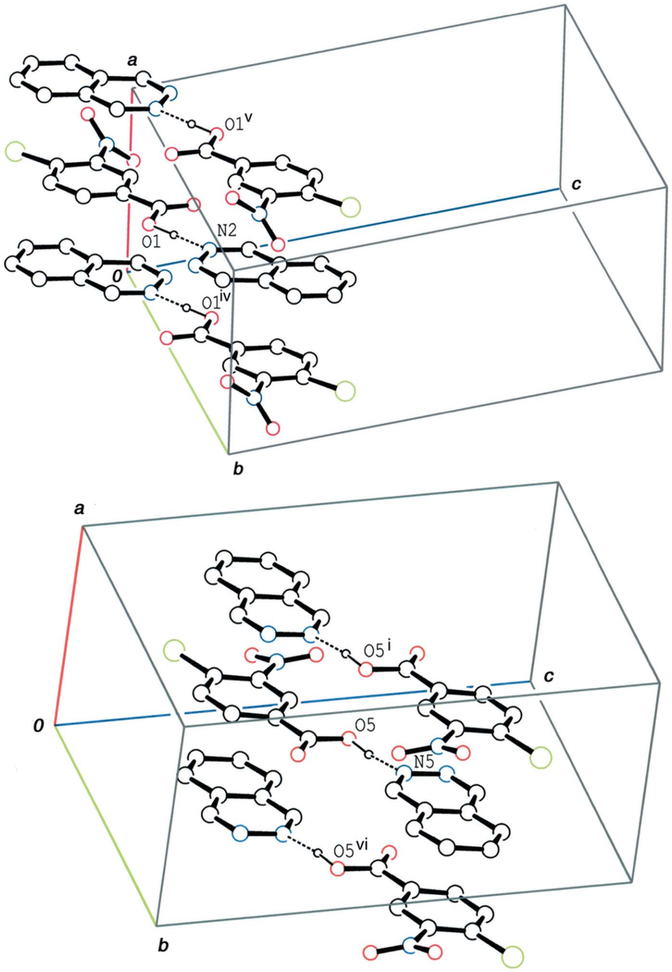


Figure 9
A packing diagram for (III), showing the π–π stacking structures of the two independent hydrogen-bonded units. Dashed lines indicate O—H$\cdots$N hydrogen bonds and H atoms not involved in the hydrogen bonds have been omitted. [Symmetry codes: (i) $-x + 1$, $-y + 1$, $-z + 1$; (iv) $-x + 1$, $-y + 1$, $-z$; (v) $-x + 2$, $-y + 1$, $-z$; (vi) $-x$, $-y + 1$, $-z + 1$.]

dense crystal than (I). The two independent hydrogen-bonded units are alternately linked in a head-to-tail manner *via* C—H$\cdots$O hydrogen bonds (C10—H10$\cdots$O7^{i} and C25—H25$\cdots$O3; details and symmetry code in Table 2), forming a tape structure along the *a* axis (Fig. 6). These tapes, related to each other by an inversion centre, are further linked by a weak C—H$\cdots$O interaction (C15—H15$\cdots$O8^{ii}; details and symmetry code in Table 2) and a π–π stacking interaction between the N2/N3/C8/C9/C14/C15 and C9–C14 rings (Table 5), forming a double-tape structure (Fig. 7). Between the double tapes, other π–π stacking interactions are also observed (Table 4). The centroid–centroid distances are in the range 3.6109 (7)–3.8139 (7) Å.

In (III), the two hydrogen-bonded units are linked by C—H$\cdots$O hydrogen bonds (C13—H13$\cdots$O6, C20—H20$\cdots$O2, C26—H26$\cdots$O4^{ii} and C27—H27$\cdots$O7^{iii}; details and symmetry codes in Table 3), forming a molecular sheet parallel to the (101) plane (Fig. 8). The arrangement of the two components in the sheet is similar to that in (I), but there are fewer hydrogen bonds than in (I). On the other hand, between these sheets, related to each other by an inversion centre, relatively short π–π interactions between the acid rings and the phthalazine ring systems are observed, with centroid–centroid distances of 3.5985 (12)–3.7248 (12) Å (Table 4), and each hydrogen-bonded unit is stacked in a column in an antiparallel manner along the *a* axis (Fig. 9), resulting in the most dense crystal of the three compounds.

We have thus shown the existence of short double-well N$\cdots$H$\cdots$O hydrogen bonds in the structures of (I) and (II). Therefore, the critical region of chloronitrobenzoic acid–phthalazine systems may be expected around $\Delta pK_a = 1.6$, which is much smaller than $\Delta pK_a = 2.93$–3.08 found in chloronitrobenzoic acid–quinoline systems.

Experimental

Crystals of all three compounds were obtained by slow evaporation from acetonitrile solutions of phthalazine with the corresponding chloro- and nitro-substituted benzoic acid in a 1:1 molar ratio at room temperature [acetonitrile solution (50 ml) of phthalazine (0.200 g) and 3-chloro-2-nitrobenzoic acid (0.310 g) for (I), acetonitrile solution (30 ml) of phthalazine (0.161 g) and 4-chloro-2-nitrobenzoic acid (0.249 g) for (II), and acetonitrile solution (90 ml) of phthalazine (0.096 g) and 4-chloro-3-nitrobenzoic acid (0.149 g) for (III)].

Compound (I)

Crystal data

$C_7H_{3.67}ClNO_4 \cdot C_8H_{6.33}N_2$
$M_r = 331.71$
Triclinic, $P\overline{1}$
$a = 9.7764$ (15) Å
$b = 12.0384$ (14) Å
$c = 12.8889$ (15) Å
$\alpha = 81.491$ (3)°
$\beta = 72.328$ (5)°
$\gamma = 88.390$ (5)°
$V = 1429.2$ (3) Å^3
$Z = 4$
Mo $K\alpha$ radiation
$\mu = 0.29$ mm^{-1}
$T = 190$ K
0.40 × 0.28 × 0.18 mm

Data collection

Rigaku R-AXIS RAPID II diffractometer
Absorption correction: numerical (*NUMABS*; Higashi, 1999)
$T_{min} = 0.874$, $T_{max} = 0.949$
31515 measured reflections
8271 independent reflections
6364 reflections with $I > 2\sigma(I)$
$R_{int} = 0.035$

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

D—H$\cdots$$A$	D—H	H$\cdots$$A$	$D\cdots A$	D—H$\cdots$$A$
O1—H1$\cdots$N2	0.86 (3)	1.71 (3)	2.5715 (15)	173 (3)
O5—H5$A$$\cdots$N5	0.84 (3)	1.71 (3)	2.5397 (17)	170 (4)
N2—H2$\cdots$O1	0.89 (4)	1.69 (4)	2.5715 (15)	172 (4)
N5—H5$B$$\cdots$O5	0.88 (5)	1.66 (5)	2.5397 (17)	172 (6)
C5—H5$\cdots$O6	0.95	2.56	3.4174 (17)	150
C8—H8$\cdots$O5^{i}	0.95	2.58	3.3560 (17)	140
C12—H12$\cdots$O3^{ii}	0.95	2.56	3.4609 (15)	158
C15—H15$\cdots$O2	0.95	2.59	3.2204 (17)	124
C20—H20$\cdots$O4^{iii}	0.95	2.52	3.2207 (17)	131
C21—H21$\cdots$N3^{iii}	0.95	2.43	3.2952 (16)	152
C23—H23$\cdots$Cl2^{ii}	0.95	2.81	3.4707 (14)	128
C23—H23$\cdots$O3^{iv}	0.95	2.56	3.3131 (16)	136
C28—H28$\cdots$O2	0.95	2.42	3.3222 (17)	158

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

Refinement

$R[F^2 > 2\sigma(F^2)] = 0.037$
$wR(F^2) = 0.112$
$S = 1.10$
8271 reflections
429 parameters
4 restraints
H atoms treated by a mixture of independent and constrained refinement
$\Delta\rho_{\max} = 0.30$ e Å^{-3}
$\Delta\rho_{\min} = -0.41$ e Å^{-3}

Compound (II)

Crystal data

$C_7H_{3.69}ClNO_4 \cdot C_8H_{6.31}N_2$
$M_r = 331.71$
Monoclinic, $P2_1/c$
$a = 12.2261$ (5) Å
$b = 16.9779$ (7) Å
$c = 14.4537$ (6) Å
$\beta = 106.8955$ (13)°
$V = 2870.7$ (2) Å^3
$Z = 8$
Mo $K\alpha$ radiation
$\mu = 0.29$ mm^{-1}
$T = 190$ K
0.35 × 0.35 × 0.30 mm

Data collection

Rigaku R-AXIS RAPID II diffractometer
Absorption correction: multi-scan (*ABSCOR*; Higashi, 1995)
$T_{\min} = 0.790$, $T_{\max} = 0.916$
60079 measured reflections
8318 independent reflections
6797 reflections with $I > 2\sigma(I)$
$R_{\rm int} = 0.030$

Refinement

$R[F^2 > 2\sigma(F^2)] = 0.042$
$wR(F^2) = 0.123$
$S = 1.07$
8318 reflections
429 parameters
4 restraints
H atoms treated by a mixture of independent and constrained refinement
$\Delta\rho_{\max} = 0.71$ e Å^{-3}
$\Delta\rho_{\min} = -0.41$ e Å^{-3}

Compound (III)

Crystal data

$C_7H_4ClNO_4 \cdot C_8H_6N_2$
$M_r = 331.71$
Triclinic, $P\overline{1}$
$a = 6.9197$ (5) Å
$b = 14.4263$ (12) Å
$c = 14.4750$ (9) Å
$\alpha = 81.704$ (2)°
$\beta = 79.0781$ (18)°
$\gamma = 85.647$ (2)°
$V = 1402.20$ (17) Å^3
$Z = 4$
Mo $K\alpha$ radiation
$\mu = 0.30$ mm^{-1}
$T = 190$ K
0.40 × 0.18 × 0.06 mm

Data collection

Rigaku R-AXIS RAPID II diffractometer
Absorption correction: numerical (*NUMABS*; Higashi, 1999)
$T_{\min} = 0.929$, $T_{\max} = 0.982$
22476 measured reflections
7849 independent reflections
4765 reflections with $I > 2\sigma(I)$
$R_{\rm int} = 0.056$

Refinement

$R[F^2 > 2\sigma(F^2)] = 0.057$
$wR(F^2) = 0.154$
$S = 0.99$
7849 reflections
423 parameters
H atoms treated by a mixture of independent and constrained refinement
$\Delta\rho_{\max} = 0.53$ e Å^{-3}
$\Delta\rho_{\min} = -0.48$ e Å^{-3}

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

D—H···A	D—H	H···A	D···A	D—H···A
O1—H1···N2	0.85 (2)	1.72 (2)	2.5655 (13)	175 (3)
O5—H5A···N5	0.86 (2)	1.75 (2)	2.6081 (13)	174 (2)
N2—H2···O1	0.88 (3)	1.70 (3)	2.5655 (13)	171 (6)
N5—H5B···O5	0.89 (3)	1.73 (3)	2.6081 (13)	170 (6)
C10—H10···O7$^{\rm i}$	0.95	2.56	3.4371 (18)	153
C15—H15···O2	0.95	2.52	3.1832 (16)	127
C15—H15···O8$^{\rm ii}$	0.95	2.54	3.1627 (17)	124
C25—H25···O3	0.95	2.47	3.3602 (19)	157
C30—H30···O6	0.95	2.57	3.2026 (16)	124

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

Table 3
Hydrogen-bond geometry (Å, °) for (III).

D—H···A	D—H	H···A	D···A	D—H···A
O1—H1···N2	1.01 (4)	1.61 (4)	2.613 (2)	171 (4)
O5—H5A···N5	1.02 (4)	1.58 (4)	2.589 (2)	172 (3)
C13—H13···O6	0.95	2.57	3.316 (3)	136
C20—H20···O2	0.95	2.52	3.203 (3)	129
C23—H23···O2$^{\rm i}$	0.95	2.51	3.220 (2)	132
C26—H26···O4$^{\rm ii}$	0.95	2.53	3.441 (3)	160
C27—H27···O7$^{\rm iii}$	0.95	2.46	3.340 (3)	155

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

Table 4
Geometries of π–π stacking interactions (Å, °) for compounds (I)–(III).

CgI···*CgJ*, α, *CgI*-Perp and *CgJ*-Perp are, respectively, the centroid–centroid distance between rings *I* and *J* (Å), the inter-ring dihedral angle (°), the perpendicular distance of *CgI* from ring *J* (Å), and the perpendicular distance of *CgJ* from ring *I* (Å). *Cg*1, *Cg*2, *Cg*3, *Cg*4, *Cg*5 and *Cg*6 are the centroids of the C1–C6, C16–C21, N2/N3/C8/C9/C14/C15, C9–C14, N5/N6/C23/C24/C29/C30 and C24–C29 rings, respectively.

	CgI···*CgJ*	α	*CgI*-Perp	*CgJ*-Perp
(I)				
*Cg*1···*Cg*3$^{\rm i}$	3.5926 (9)	9.40 (6)	3.3707 (5)	3.4833 (5)
*Cg*2···*Cg*6$^{\rm ii}$	3.8215 (9)	7.77 (6)	3.4644 (5)	3.5953 (6)
(II)				
*Cg*1···*Cg*3$^{\rm iii}$	3.6109 (7)	4.35 (6)	3.4477 (5)	3.4910 (5)
*Cg*1···*Cg*4$^{\rm iii}$	3.7346 (7)	3.20 (6)	3.4567 (5)	3.4135 (5)
*Cg*2···*Cg*5$^{\rm iv}$	3.8139 (7)	7.07 (6)	3.4937 (5)	3.4622 (5)
*Cg*2···*Cg*6$^{\rm iv}$	3.7051 (7)	6.55 (6)	3.4873 (5)	3.5827 (5)
*Cg*3···*Cg*4$^{\rm v}$	3.6714 (7)	1.58 (6)	3.4791 (5)	3.4485 (5)
(III)				
*Cg*1···*Cg*3$^{\rm vi}$	3.7248 (12)	0.78 (10)	3.5065 (4)	3.5066 (4)
*Cg*1···*Cg*3$^{\rm vii}$	3.5985 (12)	0.78 (10)	3.44488 (4)	3.4722 (4)
*Cg*1···*Cg*4$^{\rm vi}$	3.6921 (13)	1.29 (10)	3.4319 (4)	3.4669 (4)
*Cg*1···*Cg*4$^{\rm vii}$	3.6698 (13)	1.29 (10)	3.4485 (4)	3.4336 (4)
*Cg*2···*Cg*5$^{\rm viii}$	3.6536 (12)	0.89 (10)	3.3383 (4)	3.3225 (5)
*Cg*2···*Cg*5$^{\rm v}$	3.7211 (12)	0.89 (10)	3.4475 (5)	3.4475 (5)
*Cg*2···*Cg*6$^{\rm viii}$	3.6812 (13)	0.60 (10)	3.4475 (5)	3.4475 (5)
*Cg*2···*Cg*6$^{\rm v}$	3.6555 (13)	0.60 (10)	3.4475 (5)	3.4475 (5)

Symmetry codes: (i) $-x+1, -y+2, -z+1$; (ii) $-x+2, -y+1, -z+1$; (iii) $-x+1, y+\frac{1}{2}, -z+\frac{3}{2}$; (iv) $-x, y-\frac{1}{2}, -z+\frac{3}{2}$; (v) $-x+1, -y+1, -z+1$; (vi) $-x+1, -y+1, -z+1$; (vii) $-x+2, -y+1, -z$; (viii) $-x, -y+1, -z+1$.

For all three compounds, all H atoms were found in difference Fourier maps. H atoms in (I) and (II) which are involved in N···H···O hydrogen bonds were found to be disordered over two positions in difference Fourier maps. Since the site-occupation factors and isotropic displacement parameters were strongly correlated, the positional parameters and occupation factors for these atoms were refined with $U_{\rm iso}({\rm H}) = 1.5U_{\rm eq}({\rm N,O})$ and with bond-length restraints of O—H = 0.84 (2) Å and N—H = 0.88 (2) Å. Atoms H1 and H5*A* in (III) were refined isotropically. The refined distances are given in Tables 1–3. The remaining H atoms of all compounds were positioned geometrically (C—H = 0.95 Å) and treated as riding, with U_{iso}(H) = 1.2U_{eq}(C).

For all compounds, data collection and cell refinement: *PROCESS-AUTO* (Rigaku/MSC, 2004); data reduction: *CrystalStructure* (Rigaku/MSC, 2004); program(s) used to solve structure: *SHELXS97* (Sheldrick, 2008); program(s) used to refine structure: *SHELXL97* (Sheldrick, 2008); molecular graphics: *ORTEP-3* (Farrugia, 1997) and *WinGX* (Farrugia, 1999); software used to prepare material for publication: *CrystalStructure* and *PLATON* (Spek, 2009).

This work was supported by a Grant-in-Aid for Scientific Research (C) (No. 22550013) from the Japan Society for the Promotion of Science.

Supplementary data for this paper are available from the IUCr electronic archives (Reference: FG3232). Services for accessing these data are described at the back of the journal.

References

Farrugia, L. J. (1997). *J. Appl. Cryst.* **30**, 565.

Farrugia, L. J. (1999). *J. Appl. Cryst.* **32**, 837–838.

Gilli, G. & Gilli, P. (2009). *The Nature of the Hydrogen Bond*. Oxford University Press.

Gotoh, K. & Ishida, H. (2007). *Acta Cryst.* E**63**, o4500.

Gotoh, K. & Ishida, H. (2009). *Acta Cryst.* C**65**, o534–o538.

Higashi, T. (1995). *ABSCOR*. Rigaku Corporation, Tokyo, Japan.

Higashi, T. (1999). *NUMABS*. Rigaku Corporation, Tokyo, Japan.

Jerzykiewicz, L. B., Malarski, Z., Sobczyk, L., Lis, T. & Grech, E. (1998). *J. Mol. Struct.* **440**, 175–185.

Kalenik, J., Majerz, I., Sobczyk, L., Grech, E. & Habeeb, M. M. (1989). *J. Chem. Soc. Faraday Trans. 1*, **85**, 3187–3193.

Rigaku/MSC (2004). *PROCESS-AUTO* and *CrystalStructure*. Rigaku/MSC Inc., The Woodlands, Texas, USA.

Schmidtmann, M. & Wilson, C. C. (2008). *CrystEngComm*, **10**, 177–183.

Sheldrick, G. M. (2008). *Acta Cryst.* A**64**, 112–122.

Spek, A. L. (2009). *Acta Cryst.* D**65**, 148–155.

Steiner, T., Majerz, I. & Wilson, C. C. (2001). *Angew. Chem. Int. Ed.* **40**, 2651–2654.