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Crystal structure and Hirshfeld surface analysis of (*E*)-2-cyano-*N*′-(3,4,5-trimethoxybenzylidene)-acetohydrazide

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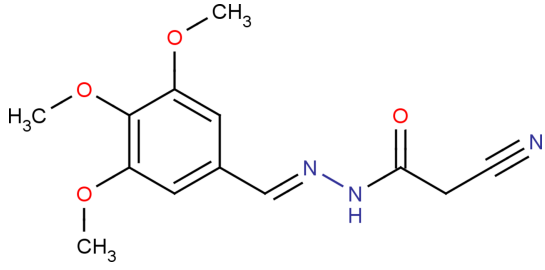
In the title compound, $C_{13}H_{15}N_3O_4$, the 2-cyano-*N*′-methylideneacetohydrazide moiety and the trimethoxy phenol ring form a dihedral angle of 13.8 (1)°. Intermolecular N—H···O and C—H···O hydrogen bonds are observed. The intermolecular interactions were quantified and analysed using Hirshfeld surface analysis, revealing that H···H interactions contribute most to the crystal packing (38.3%).

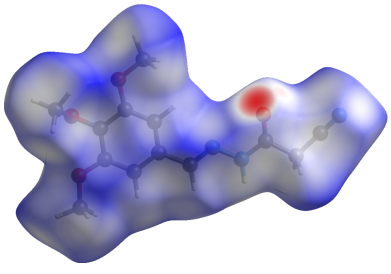
1. Chemical context

Hydrazones have been found to show various biological properties, including anticonvulsant (Angelova *et al.*, 2016), antifungal (Ozdemir *et al.*, 2008) and antitumoral (Parlar *et al.*, 2018). In the present work, the synthesis, structural and computational studies of (*E*)-2-cyano-*N*′-(3,4,5-trimethoxybenzylidene)acetohydrazide, (I), are reported.



2. Structural commentary

The molecular structure of (I) is displayed in Fig. 1. The phenyl ring (C1–C6) is planar with a maximum deviation of 0.008 (2) Å for atom C6 and its attached methoxy atoms O1, C11, O2, C12, O3 and C13 deviate by 0.023 (2), −0.169 (4), 0.133 (2), −1.089 (3), 0.010 (2) and 0.056 (3) Å, respectively. The 2-cyano-*N*′-methylideneacetohydrazide moiety (C7/N1/N2/C8/O4/C9/C10/N3) is nearly planar with a maximum deviation of 0.280 (3) Å for atom N3. This moiety forms a dihedral angle of 13.8 (1)° with the trimethoxy phenyl ring.



3. Supramolecular features

In the crystal, molecules associate pairwise by C9—H9*B*···O2^{i} and C13—H13*A*···O4^{i} hydrogen bonds (Table 1) into inversion dimers with an R_2^2 (22) graph-set motifs (Etter *et al.*, 1990), as shown in Fig. 2. The molecules are further linked into

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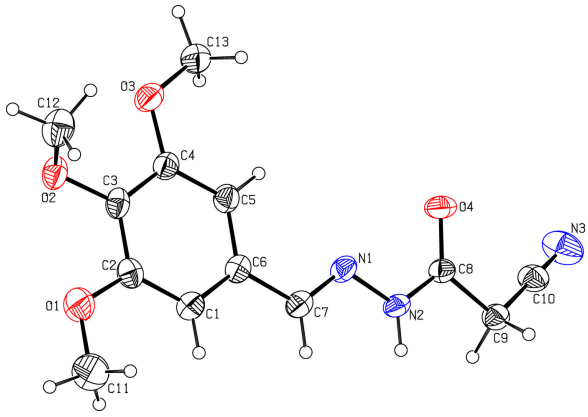

Figure 1
A view of the molecular structure of compound (I), showing the atom labelling. Displacement ellipsoids are drawn at the 30% probability level.

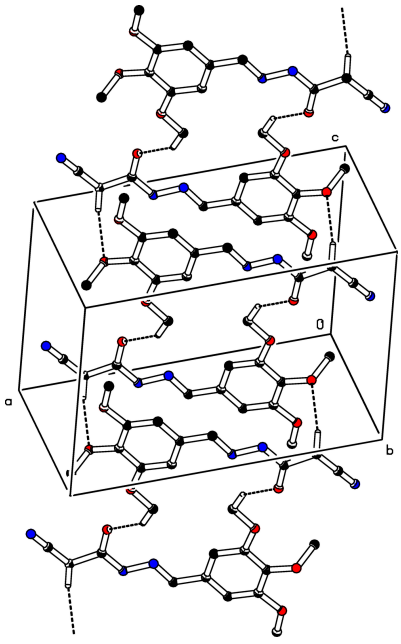

Figure 2
A view of the dimeric arrangement through O—H···O hydrogen bonds

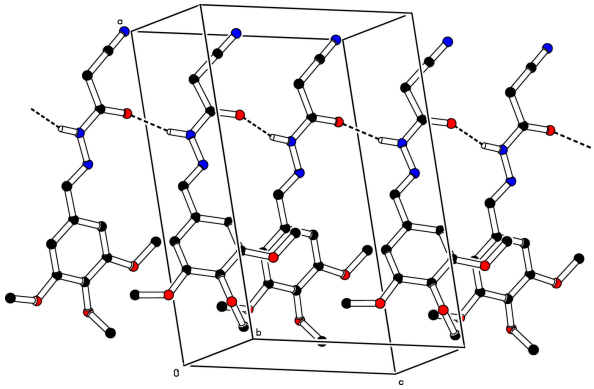

Figure 3
The crystal packing of the title compound (I) viewed along the *b* axis. The N—H···O and O—H···O intermolecular hydrogen bonds are shown as dashed lines. For clarity, H atoms not involved in hydrogen bonds have been omitted.

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

D—H···*A*	*D*—H	H···*A*	*D*···*A*	*D*—H···*A*
C9—H9*B*···O2^{i}	0.97	2.50	3.448 (3)	165
C13—H13*A*···O4^{ii}	0.96	2.51	3.350 (3)	146
N2—H2···O4^{iii}	0.86	1.98	2.802 (3)	160

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

a $C(4)$ chain motif by N2—H2···O4^{iii} hydrogen bonds running parallel to [0$\bar{1}$0] (Table 1; Fig. 3).

4. Hirshfeld surface analysis

To further characterize the intermolecular interactions, we carried out a Hirshfeld surface (HS) analysis (Spackman & Jayatilaka, 2009) using *Crystal Explorer 21* (Spackman *et al.*, 2021) The HS mapped over d_{norm} is illustrated in Fig. 4 where the deep-red spot occurs at O4 and this oxygen atom is responsible for intermolecular N—H···O and C—H···O hydrogen bonds.

The associated two-dimensional fingerprint plots (McKinnon *et al.*, 2007) provide quantitative information about the non-covalent interactions in the crystal packing in terms of the percentage contribution of the interatomic contacts (Spackman & McKinnon, 2002). The overall two-dimensional fingerprint plot is shown in Fig. 5*a*. The HS analysis reveals that H···H and H···O/O···H contacts are the main contributors to the crystal packing, followed by H···N/N···H, H···C/C···H, O···C/C···O and C···N/N···C contacts; see Fig. 5*b*–*g*. The HS analysis confirms the importance of H-atom contacts in the crystal (Hathwar *et al.*, 2015).

5. Synthesis and crystallization

The title compound (I) was synthesized by condensing 2-cyano acetohydrazide in methanol with 3,4,5-trimethoxybenzaldehyde in a 1:1 ratio following an established protocol (Shaik *et al.*, 2019). The progress of the reaction was moni-

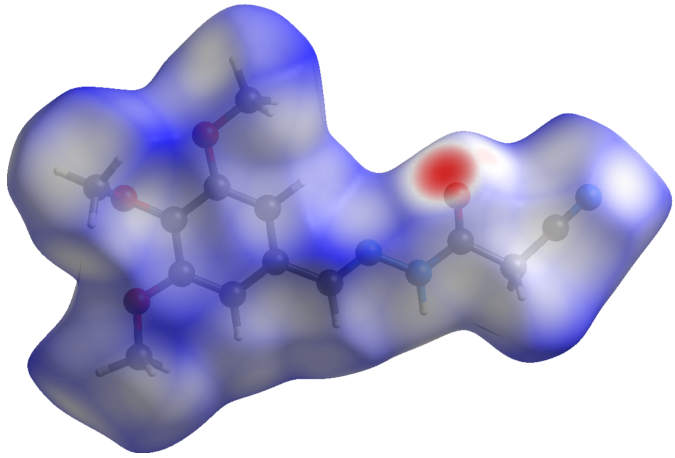

Figure 4
A view of the Hirshfeld surface mapped over d_{norm}.

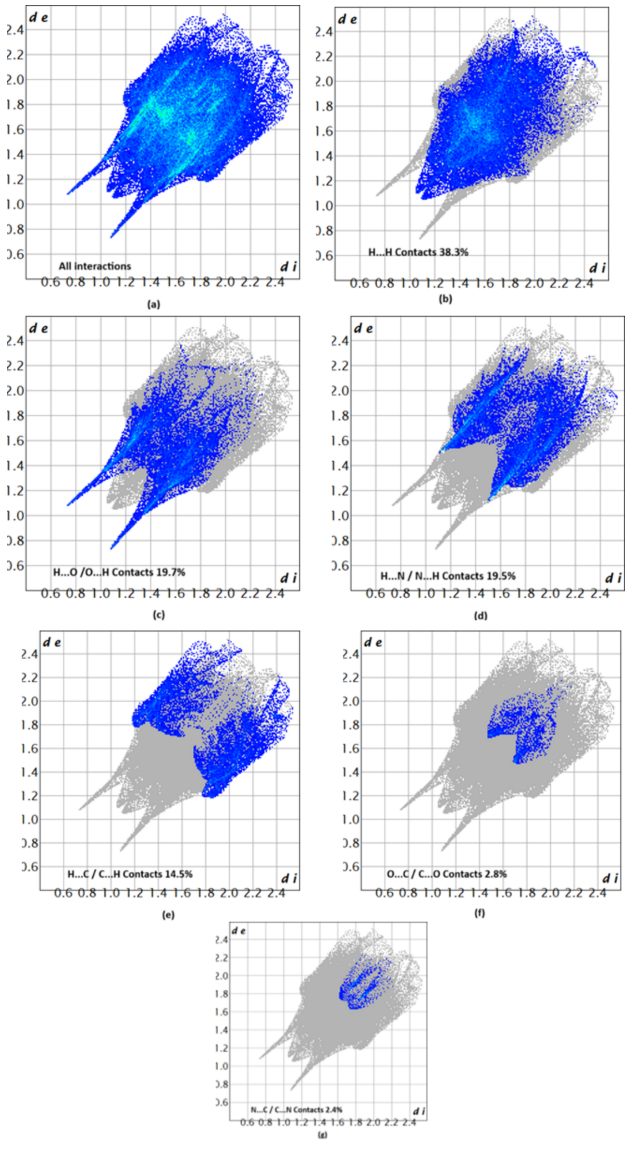

Figure 5
Two-dimensional fingerprint plots for compound (I), showing (*a*) all interactions, and delineated into (*b*) H···H, (*c*) H···O/O···H, (*d*) H···N/N···H, (*e*)H···C/C···H, (*f*) O···C/C···O and (*g*) N···C/C···N interactions. The d_i and d_e values are the closest internal and external distances (in Å) from given points on the Hirshfeld surface.

tored by thin layer chromatography (TLC). After completion of the reaction, methanol was removed under vacuum. The solid product was collected, washed, and recrystallized from methanol to obtain crystals of (I).

6. Refinement

Crystal data, data collection and structure refinement details are summarized in Table 2. All H atoms were placed in idealized positions and allowed to ride on their parent atoms: N—H = 0.86 and C—H = 0.93–0.97 Å, with U_{iso}(H) = 1.5U_{eq}(C) for methyl H atoms and U_{iso}(H) = 1.2U_{eq}(C, N) for all other H atoms.

Table 2
Experimental details.

Crystal data	
Chemical formula	$C_{13}H_{15}N_3O_4$
M_r	277.28
Crystal system, space group	Monoclinic, $P2_1/c$
Temperature (K)	298
a, b, c (Å)	13.9944 (8), 11.0371 (7), 9.0560 (5)
β (°)	99.936 (2)
V (Å^3)	1377.79 (14)
Z	4
Radiation type	Mo $K\alpha$
μ (mm^{-1})	0.10
Crystal size (mm)	0.16 × 0.12 × 0.08
Data collection	
Diffractometer	Bruker APEXII CCD
Absorption correction	Multi-scan (*SADABS*; Krause *et al.*, 2015)
T_{min}, T_{max}	0.624, 0.745
No. of measured, independent and observed [$I > 2\sigma(I)$] reflections	23917, 2823, 1540
R_{int}	0.072
(sin $\theta/\lambda)_{max}$ (Å^{-1})	0.625
Refinement	
$R[F^2 > 2\sigma(F^2)]$, $wR(F^2)$, S	0.053, 0.156, 1.05
No. of reflections	2823
No. of parameters	181
H-atom treatment	H-atom parameters constrained
$\Delta\rho_{max}$, $\Delta\rho_{min}$ (e Å^{-3})	0.15, −0.23

Computer programs: *APEX3* and *SAINT* (Bruker, 2017), *SHELXT2018/2* (Sheldrick, 2015*a*), *SHELXL2019/2* (Sheldrick, 2015*b*), *ORTEP-3 for Windows* (Farrugia, 2012) and *PLATON* (Spek, 2020).

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Computing details

(*E*)-2-Cyano-*N*′-(3,4,5-trimethoxybenzylidene)acetohydrazide

Crystal data

$C_{13}H_{15}N_3O_4$
$M_r = 277.28$
Monoclinic, $P2_1/c$
$a = 13.9944$ (8) Å
$b = 11.0371$ (7) Å
$c = 9.0560$ (5) Å
$\beta = 99.936$ (2)°
$V = 1377.79$ (14) Å^3
$Z = 4$
$F(000) = 584$
$D_x = 1.337$ Mg m^{-3}
Mo $K\alpha$ radiation, $\lambda = 0.71073$ Å
Cell parameters from 4885 reflections
$\theta = 2.4$–22.9°
$\mu = 0.10$ mm^{-1}
$T = 298$ K
Block, brown
0.16 × 0.12 × 0.08 mm

Data collection

Bruker APEXII CCD diffractometer
Radiation source: i-mu-s microfocus source
φ and ω scans
Absorption correction: multi-scan (SADABS; Krause *et al.*, 2015)
$T_{min} = 0.624$, $T_{max} = 0.745$
23917 measured reflections
2823 independent reflections
1540 reflections with $I > 2\sigma(I)$
$R_{int} = 0.072$
$\theta_{max} = 26.4°$, $\theta_{min} = 2.4°$
$h = -17\rightarrow15$
$k = -13\rightarrow13$
$l = -11\rightarrow8$

Refinement

Refinement on F^2
Least-squares matrix: full
$R[F^2 > 2\sigma(F^2)] = 0.053$
$wR(F^2) = 0.156$
$S = 1.05$
2823 reflections
181 parameters
0 restraints
Hydrogen site location: inferred from neighbouring sites
H-atom parameters constrained
$w = 1/[\sigma^2(F_o^2) + (0.0585P)^2 + 0.5215P]$ where $P = (F_o^2 + 2F_c^2)/3$
$(\Delta/\sigma)_{max} < 0.001$
$\Delta\rho_{max} = 0.15$ e Å^{-3}
$\Delta\rho_{min} = -0.23$ 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 (Å²)

	x	*y*	*z*	U_{iso}*/U_{eq}
O1	0.17230 (13)	0.43960 (19)	−0.1731 (2)	0.0708 (6)
O2	0.15221 (12)	0.52233 (16)	0.0935 (2)	0.0574 (5)
O3	0.29283 (12)	0.49755 (17)	0.3328 (2)	0.0586 (5)
O4	0.73449 (12)	0.30054 (17)	0.34803 (19)	0.0589 (5)
C7	0.51086 (18)	0.3277 (2)	0.0177 (3)	0.0473 (6)
H7	0.522223	0.308137	−0.077620	0.057*
N1	0.57857 (14)	0.31831 (18)	0.1302 (2)	0.0471 (5)
N3	0.98039 (19)	0.2939 (3)	0.3950 (3)	0.0824 (9)
C1	0.33918 (18)	0.3770 (2)	−0.0817 (3)	0.0502 (7)
H1	0.347981	0.349974	−0.175678	0.060*
C2	0.25037 (18)	0.4247 (2)	−0.0625 (3)	0.0496 (7)
C3	0.23722 (17)	0.4649 (2)	0.0780 (3)	0.0463 (6)
C4	0.31260 (18)	0.4556 (2)	0.1992 (3)	0.0458 (6)
C5	0.40109 (18)	0.4078 (2)	0.1801 (3)	0.0485 (7)
H5	0.451361	0.401270	0.261518	0.058*
C6	0.41465 (17)	0.3697 (2)	0.0388 (3)	0.0456 (6)
N2	0.66924 (14)	0.28820 (18)	0.1029 (2)	0.0446 (5)
H2	0.678178	0.272351	0.013346	0.054*
C8	0.74341 (17)	0.2840 (2)	0.2180 (3)	0.0423 (6)
C9	0.84027 (16)	0.2566 (2)	0.1732 (3)	0.0470 (6)
H9A	0.841502	0.172809	0.141281	0.056*
H9B	0.848985	0.307709	0.089430	0.056*
C10	0.9189 (2)	0.2776 (3)	0.2976 (3)	0.0533 (7)
C11	0.1728 (2)	0.3876 (3)	−0.3140 (4)	0.0847 (10)
H11A	0.112939	0.406117	−0.379339	0.127*
H11B	0.226071	0.419600	−0.355726	0.127*
H11C	0.179561	0.301281	−0.303628	0.127*
C12	0.0851 (2)	0.4491 (3)	0.1573 (4)	0.0719 (9)
H12A	0.028134	0.495629	0.164137	0.108*
H12B	0.067467	0.379559	0.094810	0.108*
H12C	0.114932	0.422970	0.255613	0.108*
C13	0.3689 (2)	0.4940 (3)	0.4573 (3)	0.0677 (8)
H13A	0.346330	0.525646	0.543898	0.102*
H13B	0.389969	0.411839	0.476062	0.102*
H13C	0.422169	0.542265	0.436720	0.102*

Atomic displacement parameters (Å²)

	U^{11}	U^{22}	U^{33}	U^{12}	U^{13}	U^{23}
O1	0.0544 (12)	0.0881 (15)	0.0646 (13)	0.0106 (10)	−0.0047 (10)	−0.0052 (11)
O2	0.0438 (10)	0.0538 (11)	0.0771 (13)	0.0103 (9)	0.0176 (9)	0.0081 (9)
O3	0.0502 (11)	0.0717 (13)	0.0553 (12)	0.0109 (9)	0.0128 (9)	−0.0084 (9)
O4	0.0594 (12)	0.0831 (14)	0.0376 (10)	0.0069 (10)	0.0184 (9)	0.0000 (9)
C7	0.0466 (15)	0.0463 (15)	0.0517 (16)	0.0016 (12)	0.0157 (13)	−0.0031 (12)
N1	0.0398 (12)	0.0527 (13)	0.0522 (13)	0.0056 (10)	0.0179 (11)	−0.0005 (10)

N3	0.0639 (17)	0.116 (2)	0.0637 (17)	−0.0227 (16)	0.0017 (14)	0.0100 (16)
C1	0.0499 (16)	0.0494 (15)	0.0522 (16)	0.0017 (12)	0.0110 (14)	−0.0042 (12)
C2	0.0438 (15)	0.0492 (15)	0.0534 (17)	0.0003 (12)	0.0020 (13)	0.0048 (13)
C3	0.0364 (14)	0.0400 (14)	0.0635 (18)	0.0024 (11)	0.0110 (13)	0.0062 (12)
C4	0.0460 (15)	0.0413 (14)	0.0521 (16)	0.0025 (11)	0.0138 (13)	0.0018 (12)
C5	0.0419 (15)	0.0508 (16)	0.0527 (16)	0.0048 (12)	0.0081 (12)	0.0020 (12)
C6	0.0432 (15)	0.0416 (14)	0.0532 (16)	0.0016 (11)	0.0118 (13)	−0.0011 (12)
N2	0.0410 (12)	0.0567 (13)	0.0396 (12)	0.0047 (10)	0.0162 (10)	−0.0017 (10)
C8	0.0432 (15)	0.0455 (14)	0.0407 (15)	0.0005 (11)	0.0141 (12)	0.0024 (11)
C9	0.0421 (15)	0.0551 (16)	0.0447 (15)	0.0009 (12)	0.0105 (12)	0.0021 (12)
C10	0.0477 (17)	0.0650 (18)	0.0497 (17)	−0.0052 (14)	0.0156 (14)	0.0077 (14)
C11	0.076 (2)	0.099 (3)	0.070 (2)	0.0001 (19)	−0.0110 (18)	−0.021 (2)
C12	0.0469 (17)	0.071 (2)	0.101 (2)	−0.0049 (15)	0.0221 (17)	0.0050 (18)
C13	0.072 (2)	0.078 (2)	0.0522 (18)	0.0183 (17)	0.0077 (16)	−0.0050 (15)

Geometric parameters (Å, º)

O1—C2	1.359 (3)	C5—C6	1.390 (3)
O1—C11	1.401 (3)	C5—H5	0.9300
O2—C3	1.376 (3)	N2—C8	1.339 (3)
O2—C12	1.434 (3)	N2—H2	0.8600
O3—C4	1.367 (3)	C8—C9	1.511 (3)
O3—C13	1.412 (3)	C9—C10	1.451 (4)
O4—C8	1.219 (3)	C9—H9A	0.9700
C7—N1	1.270 (3)	C9—H9B	0.9700
C7—C6	1.467 (3)	C11—H11A	0.9600
C7—H7	0.9300	C11—H11B	0.9600
N1—N2	1.375 (3)	C11—H11C	0.9600
N3—C10	1.136 (3)	C12—H12A	0.9600
C1—C6	1.384 (3)	C12—H12B	0.9600
C1—C2	1.388 (3)	C12—H12C	0.9600
C1—H1	0.9300	C13—H13A	0.9600
C2—C3	1.389 (4)	C13—H13B	0.9600
C3—C4	1.389 (3)	C13—H13C	0.9600
C4—C5	1.384 (3)		
C2—O1—C11	119.5 (2)	O4—C8—N2	123.5 (2)
C3—O2—C12	114.76 (19)	O4—C8—C9	122.4 (2)
C4—O3—C13	117.24 (19)	N2—C8—C9	114.1 (2)
N1—C7—C6	119.7 (2)	C10—C9—C8	110.8 (2)
N1—C7—H7	120.1	C10—C9—H9A	109.5
C6—C7—H7	120.1	C8—C9—H9A	109.5
C7—N1—N2	117.3 (2)	C10—C9—H9B	109.5
C6—C1—C2	120.1 (2)	C8—C9—H9B	109.5
C6—C1—H1	120.0	H9A—C9—H9B	108.1
C2—C1—H1	120.0	N3—C10—C9	179.9 (4)
O1—C2—C1	125.2 (2)	O1—C11—H11A	109.5
O1—C2—C3	114.8 (2)	O1—C11—H11B	109.5

C1—C2—C3	119.9 (2)	H11A—C11—H11B	109.5
O2—C3—C4	120.4 (2)	O1—C11—H11C	109.5
O2—C3—C2	119.6 (2)	H11A—C11—H11C	109.5
C4—C3—C2	119.8 (2)	H11B—C11—H11C	109.5
O3—C4—C5	124.2 (2)	O2—C12—H12A	109.5
O3—C4—C3	115.6 (2)	O2—C12—H12B	109.5
C5—C4—C3	120.2 (2)	H12A—C12—H12B	109.5
C4—C5—C6	119.8 (2)	O2—C12—H12C	109.5
C4—C5—H5	120.1	H12A—C12—H12C	109.5
C6—C5—H5	120.1	H12B—C12—H12C	109.5
C1—C6—C5	120.1 (2)	O3—C13—H13A	109.5
C1—C6—C7	120.5 (2)	O3—C13—H13B	109.5
C5—C6—C7	119.3 (2)	H13A—C13—H13B	109.5
C8—N2—N1	118.96 (19)	O3—C13—H13C	109.5
C8—N2—H2	120.5	H13A—C13—H13C	109.5
N1—N2—H2	120.5	H13B—C13—H13C	109.5

C6—C7—N1—N2	−174.8 (2)	O2—C3—C4—C5	−174.0 (2)
C11—O1—C2—C1	10.5 (4)	C2—C3—C4—C5	0.8 (4)
C11—O1—C2—C3	−171.3 (3)	O3—C4—C5—C6	−178.7 (2)
C6—C1—C2—O1	178.0 (2)	C3—C4—C5—C6	0.3 (4)
C6—C1—C2—C3	−0.1 (4)	C2—C1—C6—C5	1.3 (4)
C12—O2—C3—C4	−81.8 (3)	C2—C1—C6—C7	−175.5 (2)
C12—O2—C3—C2	103.3 (3)	C4—C5—C6—C1	−1.4 (4)
O1—C2—C3—O2	−4.3 (3)	C4—C5—C6—C7	175.4 (2)
C1—C2—C3—O2	174.0 (2)	N1—C7—C6—C1	−178.0 (2)
O1—C2—C3—C4	−179.2 (2)	N1—C7—C6—C5	5.1 (4)
C1—C2—C3—C4	−1.0 (4)	C7—N1—N2—C8	176.4 (2)
C13—O3—C4—C5	1.6 (4)	N1—N2—C8—O4	3.2 (4)
C13—O3—C4—C3	−177.5 (2)	N1—N2—C8—C9	−176.8 (2)
O2—C3—C4—O3	5.1 (3)	O4—C8—C9—C10	−11.6 (3)
C2—C3—C4—O3	180.0 (2)	N2—C8—C9—C10	168.4 (2)

Hydrogen-bond geometry (Å, º)

D—H···*A*	*D*—H	H···*A*	*D*···*A*	*D*—H···*A*
C9—H9*B*···O2^{i}	0.97	2.50	3.448 (3)	165
C13—H13*A*···O4^{ii}	0.96	2.51	3.350 (3)	146
N2—H2···O4^{iii}	0.86	1.98	2.802 (3)	160

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