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Key indicators

Single-crystal X-ray study
T = 293 K
Mean $\sigma(\text{C}-\text{C}) = 0.004 \text{ \AA}$
R factor = 0.052
wR factor = 0.151
Data-to-parameter ratio = 14.1

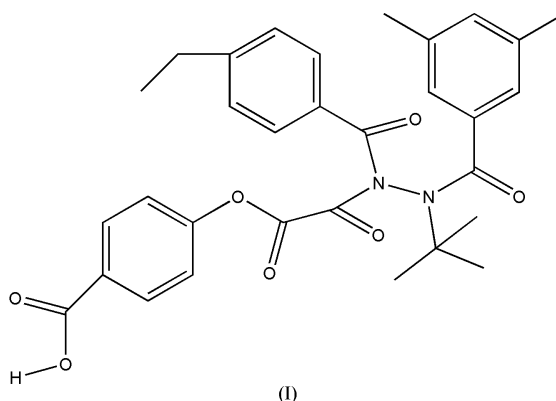
For details of how these key indicators were automatically derived from the article, see <http://journals.iucr.org/e>.

N'-*tert*-Butyl-*N'*-(3,5-dimethylbenzoyl)-*N*-(4-carboxyphenoxy)oxalyl-*N*-(4-ethylbenzoyl)hydrazine

The title compound, $\text{C}_{31}\text{H}_{32}\text{N}_2\text{O}_7$, was synthesized by hydrogenation of *N'*-*tert*-butyl-*N'*-(3,5-dimethylbenzoyl)-*N*-(4-benzyloxycarbonylphenoxy)oxalyl-*N*-(4-ethylbenzoyl)hydrazine using palladium on carbon as catalyst. In the crystal structure, the molecules are linked *via* intermolecular O—H···O hydrogen bonds, forming chains. In addition, intermolecular C—H···O hydrogen bonds and C—H··· π interactions are observed.

Comment

Synthetic substituted *N'*-*tert*-butyl-*N,N'*-diacylhydrazines have been found to be effective as non-steroidal ecdysone agonists inducing, especially in Lepidoptera, precocious molting, leading to death (Wing, 1988; Wing *et al.*, 1988; Dhadialla *et al.*, 1998). *N'*-*tert*-Butyl-*N'*-4-ethylbenzoyl-*N*-3,5-dimethylbenzoylhydrazide was the first to be commercialized as a lepidopteran-specific insecticide (Dhadialla & Jansson, 1999). In the search for new insect growth regulators with improved profiles, we have synthesized a series of derivatives of *N'*-*tert*-butyl-*N'*-4-ethylbenzoyl-*N*-3,5-dimethylbenzoylhydrazide (Mao *et al.*, 2004). We report here the crystal structure of *N'*-*tert*-butyl-*N'*-(3,5-dimethylbenzoyl)-*N*-(4-carboxyphenoxy)oxalyl-*N*-(4-ethylbenzoyl)hydrazine, (I).



The sums of the bond angles around N1 and N2 (364.4 and 359.8°, respectively) indicate sp^2 hybridization. The C15–C20 benzene ring forms dihedral angles of 44.72 (11) and 61.18 (10)° with the C2–C7 and C25–C30 benzene rings, respectively. The carboxyl group attached to the C25–C30 benzene ring is coplanar with it. The molecular structure is stabilized by weak C—H···O hydrogen bonds and C—H··· π interactions involving the C2–C7 (centroid Cg1) and C15–C20 rings (centroid Cg2) (Table 1 and Fig. 1). The carboxyl and carbonyl O atoms are involved in O—H···O intermolecular hydrogen bonds which link the molecules into a chain along

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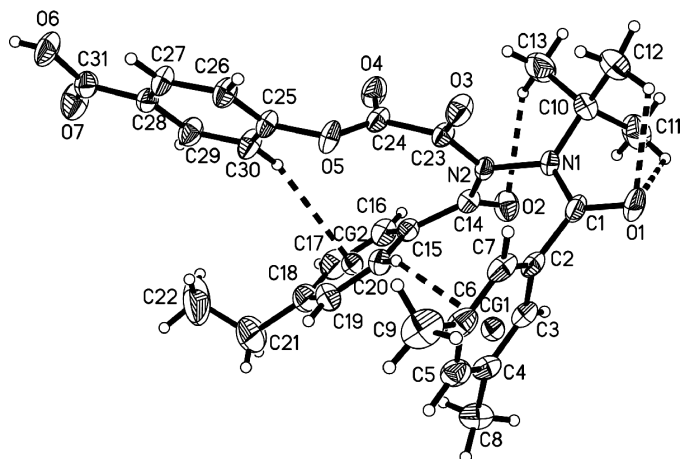


Figure 1
The structure of (I), showing 40% probability displacement ellipsoids and the atom-numbering scheme. C—H...O and C—H... π interactions are shown as dashed lines.

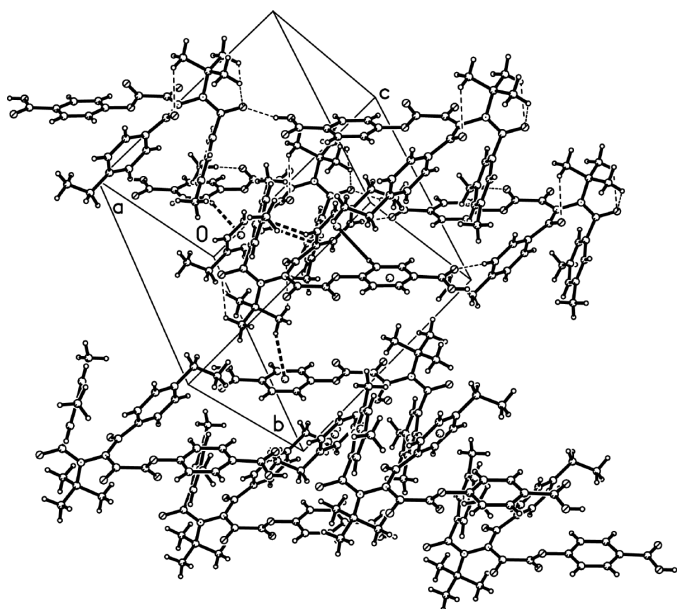


Figure 2
The crystal packing of (I). Intermolecular hydrogen bonds are shown as dashed lines.

[011]. The crystal packing is further stabilized by C—H... π interactions involving the C25–C30 benzene ring (centroid Cg3) and C—H...O hydrogen bonds (Table 1 and Fig. 2).

Experimental

According to the reported procedure of Mao *et al.* (2004), the title compound was synthesized by hydrogenation of *N*-*tert*-butyl-*N'*-(3,5-dimethylbenzoyl)-*N*-(4-benzyloxycarbonylphenoxy)oxalyl-*N*-(4-ethylbenzoyl)hydrazine using 5% palladium on carbon as catalyst in ethyl acetate at room temperature. The reaction mixture was filtered to remove solids, and the organic filtrate was concentrated under reduced pressure to yield the title compound, (I), which was recrystallized from acetonitrile to give single crystals suitable for X-ray diffraction.

Crystal data

$C_{31}H_{32}N_2O_7$
 $M_r = 544.59$
Triclinic, $P\bar{1}$
 $a = 9.971(4) \text{ \AA}$
 $b = 13.239(5) \text{ \AA}$
 $c = 13.649(5) \text{ \AA}$
 $\alpha = 111.141(6)^\circ$
 $\beta = 103.553(6)^\circ$
 $\gamma = 107.231(6)^\circ$
 $V = 1482.7(10) \text{ \AA}^3$

$Z = 2$
 $D_x = 1.220 \text{ Mg m}^{-3}$
Mo $K\alpha$ radiation
Cell parameters from 738 reflections
 $\theta = 2.3\text{--}23.5^\circ$
 $\mu = 0.09 \text{ mm}^{-1}$
 $T = 293(2) \text{ K}$
Prism, colourless
 $0.28 \times 0.22 \times 0.20 \text{ mm}$

Data collection

Bruker SMART CCD area-detector
diffractometer
 φ and ω scans
Absorption correction: none
7682 measured reflections
5192 independent reflections

3119 reflections with $I > 2\sigma(I)$
 $R_{\text{int}} = 0.024$
 $\theta_{\text{max}} = 25.0^\circ$
 $h = -11 \rightarrow 11$
 $k = -15 \rightarrow 10$
 $l = -16 \rightarrow 16$

Refinement

Refinement on F^2
 $R[F^2 > 2\sigma(F^2)] = 0.052$
 $wR(F^2) = 0.151$
 $S = 1.03$
5192 reflections
368 parameters
H-atom parameters constrained

$w = 1/[\sigma^2(F_o^2) + (0.0687P)^2 + 0.2554P]$
where $P = (F_o^2 + 2F_c^2)/3$
 $(\Delta/\sigma)_{\text{max}} < 0.001$
 $\Delta\rho_{\text{max}} = 0.21 \text{ e \AA}^{-3}$
 $\Delta\rho_{\text{min}} = -0.20 \text{ e \AA}^{-3}$

Table 1

Hydrogen-bonding geometry (\AA , $^\circ$).

D—H...A	D—H	H...A	D...A	D—H...A
O6—H6...O1 ⁱ	0.82	1.86	2.655 (2)	163
C9—H9A...O4 ⁱⁱ	0.96	2.50	3.393 (4)	154
C11—H11A...O1	0.96	2.46	3.052 (5)	120
C12—H12C...O1	0.96	2.36	2.937 (5)	118
C13—H13A...O2	0.96	2.55	3.261 (5)	131
C17—H17...O7 ⁱⁱⁱ	0.93	2.49	3.422 (5)	178
C13—H13B...Cg3 ^{iv}	0.96	2.80	3.670 (4)	151
C20—H20...Cg1	0.93	2.64	3.365 (3)	136
C30—H30...Cg2	0.93	2.82	3.618 (3)	145

Symmetry codes: (i) $x, 1+y, 1+z$; (ii) $1+x, y, z$; (iii) $1-x, 2-y, 2-z$; (iv) $1-x, 2-y, 1-z$. Cg1, Cg2 and Cg3 are the centroids of the C2–C7, C15–C20 and C25–C30 benzene rings, respectively.

H atoms were placed in calculated positions, with C—H distances in the range 0.93–0.97 \AA and O—H = 0.82 \AA , and included in the final cycles of refinement using a riding model, with $U_{\text{iso}}(\text{H})$ values of 1.2 or 1.5(methyl) times $U_{\text{eq}}(\text{parent atom})$.

Data collection: *SMART* (Bruker, 1998); cell refinement: *SAINTE* (Bruker, 1999); data reduction: *SAINTE*; program(s) used to solve structure: *SHELXTL* (Bruker, 1999); program(s) used to refine structure: *SHELXTL*; molecular graphics: *SHELXTL*; software used to prepare material for publication: *SHELXTL*.

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References

Bruker (1998). *SMART*. Bruker AXS Inc., Madison, Wisconsin, USA.

- Bruker (1999). *SAINTE* and *SHELXTL*. Bruker AXS Inc., Madison, Wisconsin, USA.
- Dhadialla, T. S., Carlson, G. R. & Le, D. P. (1998). *Annu. Rev. Entomol.* **43**, 545–569.
- Dhadialla, T. S. & Jansson, R. K. (1999). *Pest. Sci.* **55**, 357–359.
- Mao, C. H., Wang, Q. M., Huang, R. Q., Bi, F. C., Chen, L., Liu, Y. X. & Shang, J. (2004). *J. Agric. Food Chem.* **52**, 6737–6741.
- Wing, K. D. (1988). *Science*, **241**, 467–269.
- Wing, K. D., Slawecky, R. A. & Carlson, G. R. (1988). *Science*, **241**, 470–472.