

Acta Crystallographica Section E

## Structure Reports

Online

ISSN 1600-5368

# (*E,E*)-2,5-Bis(5-chloro-2-methoxyphenyl)-3,4-diazahexa-2,4-diene

 Jian-Guo Chang,<sup>a\*</sup> Jie Lu<sup>b</sup> and Ren-Gao Zhao<sup>a</sup>

<sup>a</sup>Department of Materials Science and Chemical Engineering, Taishan University, 271021 Taian, Shandong, People's Republic of China, and <sup>b</sup>Department of Architecture and Mechanical Engineering, Taishan University, 271021 Taian, Shandong, People's Republic of China  
Correspondence e-mail: tsucjg@163.com

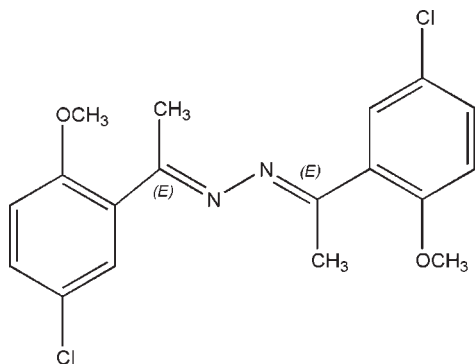
Received 25 September 2009; accepted 14 November 2009

Key indicators: single-crystal X-ray study;  $T = 295$  K; mean  $\sigma(\text{C}-\text{C}) = 0.003$  Å;  $R$  factor = 0.036;  $wR$  factor = 0.129; data-to-parameter ratio = 14.1.

The title compound,  $\text{C}_{18}\text{H}_{18}\text{Cl}_2\text{N}_2\text{O}_2$ , was synthesized by the reaction of 1-(5-chloro-2-methoxyphenyl)ethanone with hydrazine hydrate. The molecule lies on a crystallographic twofold axis passing through the mid-point of the N—N bond with one half-molecule in the asymmetric unit. The dihedral angle between the two aromatic rings is  $44.33(4)^\circ$ . In the crystal, intermolecular C—H $\cdots$ O interactions link the molecules into columns along the  $c$  axis

## Related literature

For azine compounds containing both a diimine linkage and N—N bonding, see: Kessler *et al.* (1999); Kundu *et al.* (2005). For related structures, see: Glaser *et al.* (1995); Hunig *et al.* (2000).



## Experimental

## Crystal data

$\text{C}_{18}\text{H}_{18}\text{Cl}_2\text{N}_2\text{O}_2$   
 $M_r = 365.24$   
 Orthorhombic,  $P2_12_12$   
 $a = 7.9030(19)$  Å  
 $b = 27.862(7)$  Å  
 $c = 3.9819(10)$  Å  
 $V = 876.8(4)$  Å<sup>3</sup>  
 $Z = 2$   
 Mo  $K\alpha$  radiation  
 $\mu = 0.38$  mm<sup>-1</sup>  
 $T = 295$  K  
 $0.22 \times 0.16 \times 0.12$  mm

## Data collection

Bruker APEXII CCD area detector diffractometer  
 Absorption correction: multi-scan (SADABS; Sheldrick, 2003)  
 $T_{\min} = 0.921$ ,  $T_{\max} = 0.956$   
 4469 measured reflections  
 1566 independent reflections  
 1417 reflections with  $I > 2\sigma(I)$   
 $R_{\text{int}} = 0.019$

## Refinement

$R[F^2 > 2\sigma(F^2)] = 0.036$   
 $wR(F^2) = 0.129$   
 $S = 1.01$   
 1566 reflections  
 111 parameters  
 H-atom parameters constrained  
 $\Delta\rho_{\max} = 0.12$  e Å<sup>-3</sup>  
 $\Delta\rho_{\min} = -0.22$  e Å<sup>-3</sup>  
 Absolute structure: Flack (1983), 592 Friedel pairs  
 Flack parameter: 0.08 (12)

Table 1

Hydrogen-bond geometry (Å, °).

$D-H\cdots A$	$D-H$	$H\cdots A$	$D\cdots A$	$D-H\cdots A$
$\text{C9}-\text{H9B}\cdots\text{O1}^i$	0.96	2.68	3.521 (3)	146

 Symmetry code: (i)  $x, y, z - 1$ .

Data collection: APEX2 (Bruker, 2005); cell refinement: SAINT (Bruker, 2005); data reduction: SAINT; program(s) used to solve structure: SHELXS97 (Sheldrick, 2008); program(s) used to refine structure: SHELXL97 (Sheldrick, 2008); molecular graphics: SHELXTL (Sheldrick, 2008); software used to prepare material for publication: SHELXTL.

This project was supported by the Postgraduate Foundation of Taishan University (No.Y05-2-09)

Supplementary data and figures for this paper are available from the IUCr electronic archives (Reference: FL2273).

## References

- Bruker (2005). APEX2 and SAINT. Bruker AXS Inc., Madison, Wisconsin, USA.  
 Flack, H. D. (1983). *Acta Cryst.* **A39**, 876–881.  
 Glaser, R., Chen, G. S., Anthamatten, M. & Barnes, C. L. (1995). *J. Chem. Soc. Perkin Trans. 2*, pp. 1449–1458.  
 Hunig, S., Kemmer, M. & Wenner, H. (2000). *Chem. Eur. J.* **6**, 2618–2632.  
 Kessler, E. C., Euler, W. B. & Foxman, B. M. (1999). *Chem. Mater.* **11**, 336–340.  
 Kundu, N., Chatterjee, P. B., Chaudhury, M. & Tiekink, E. R. T. (2005). *Acta Cryst.* **E61**, m1583–m1585.  
 Sheldrick, G. M. (2003). SADABS. University of Göttingen, Germany.  
 Sheldrick, G. M. (2008). *Acta Cryst.* **A64**, 112–122.

## supporting information

*Acta Cryst.* (2009). E65, o3185 [doi:10.1107/S1600536809048351]

**(*E,E*)-2,5-Bis(5-chloro-2-methoxyphenyl)-3,4-diazahexa-2,4-diene****Jian-Guo Chang, Jie Lu and Ren-Gao Zhao****S1. Comment**

Recently, a number of azine compounds containing both a diimine linkage and N—N bonding have been investigated in terms of their crystallography and coordination chemistry (Kundu *et al.*, 2005; Kesslen *et al.*, 1999). As an extension of the work on the structural characterization of azine derivatives, the title compound, (I), was synthesized and its crystal structure is reported here.

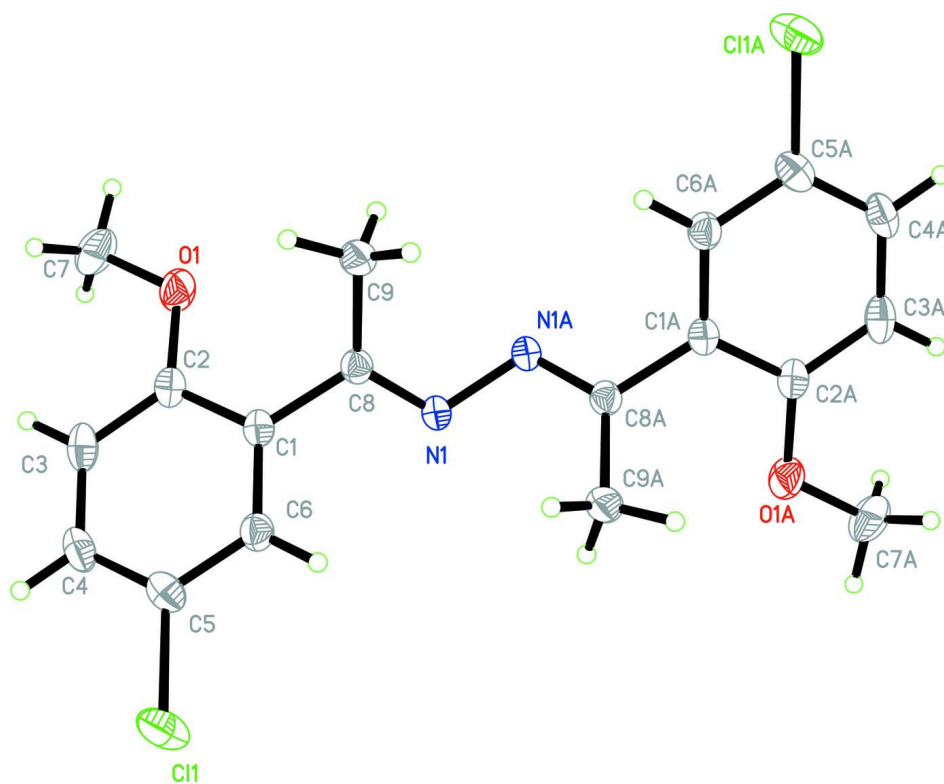
The molecule lies on a crystallographic 2-fold axis passing through the mid-point of the N—N bond to give 1/2 molecule per asymmetric unit. (Fig. 1). The dihedral angle between the two aromatic rings is 44.33 (4)°. The N atom and the phenyl ring lie on opposite side of the C8=N1 bond to give an (*E, E*) conformation with respect to the C8=N1 bond ( and its symmetry related C8a=N1a double bond (Fig. 1.). This configuration agrees with those commonly found in similar compounds (Glaser *et al.*, 1995; Hunig *et al.*, 2000). Intermolecular C—H···O interactions link the molecules into columns along the *c* axis (Table 1, Fig. 2).

**S2. Experimental**

An ethanol solution (30 ml) of hydrazine (0.02 mol) and 1-(5-chloro-2-methoxyphenyl)ethanone (0.04 mol) was refluxed and stirred for 6 h; the mixture was cooled and the resulting solid product, (I), was collected by filtration. Colourless crystals suitable for single-crystal X-ray diffraction were grown by slow evaporation of a solution in acetone.

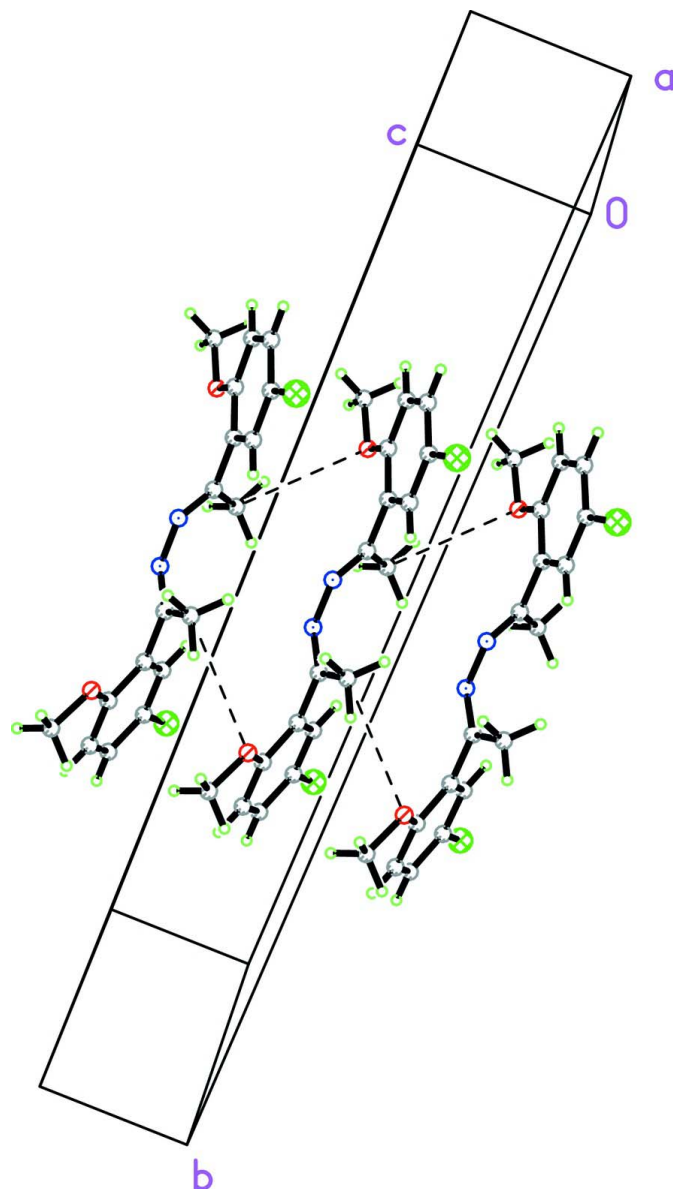
**S3. Refinement**

All H atoms were positioned geometrically and treated as riding on their parent atoms, with C—H(methyl) = 0.96 Å, C—H(aromatic) = 0.93 Å, and with  $U_{\text{iso}}(\text{H}) = 1.5U_{\text{eq}}(\text{C}_{\text{methyl}})$  and  $1.2U_{\text{eq}}(\text{C}_{\text{aromatic}})$ .



**Figure 1**

The molecular structure of compound (I), showing the atom-numbering scheme. Displacement ellipsoids are drawn at the 30% probability level.



**Figure 2**

The crystal packing of (I), Dashed lines show intermolecular C—H...O interactions.

**(*E,E*)-2,5-Bis(5-chloro-2-methoxyphenyl)-3,4-diazahexa-2,4-diene**

*Crystal data*

$C_{18}H_{18}Cl_2N_2O_2$

$M_r = 365.24$

Orthorhombic,  $P2_12_12$

Hall symbol:  $P\ 2\ 2ab$

$a = 7.9030\ (19)\ \text{\AA}$

$b = 27.862\ (7)\ \text{\AA}$

$c = 3.9819\ (10)\ \text{\AA}$

$V = 876.8\ (4)\ \text{\AA}^3$

$Z = 2$

$F(000) = 380$

$D_x = 1.383\ \text{Mg m}^{-3}$

Mo  $K\alpha$  radiation,  $\lambda = 0.71073\ \text{\AA}$

Cell parameters from 2313 reflections

$\theta = 2.7\text{--}27.8^\circ$

$\mu = 0.38\ \text{mm}^{-1}$

$T = 295\ \text{K}$

Block, colourless

$0.22 \times 0.16 \times 0.12\ \text{mm}$

*Data collection*

Bruker APEXII CCD area detector  
 diffractometer  
 Radiation source: fine-focus sealed tube  
 Graphite monochromator  
 phi and  $\omega$  scans  
 Absorption correction: multi-scan  
 (SADABS; Sheldrick, 2003)  
 $T_{\min} = 0.921$ ,  $T_{\max} = 0.956$

4469 measured reflections  
 1566 independent reflections  
 1417 reflections with  $I > 2\sigma(I)$   
 $R_{\text{int}} = 0.019$   
 $\theta_{\text{max}} = 25.0^\circ$ ,  $\theta_{\text{min}} = 1.5^\circ$   
 $h = -9 \rightarrow 8$   
 $k = -33 \rightarrow 32$   
 $l = -4 \rightarrow 4$

*Refinement*

Refinement on  $F^2$   
 Least-squares matrix: full  
 $R[F^2 > 2\sigma(F^2)] = 0.036$   
 $wR(F^2) = 0.129$   
 $S = 1.01$   
 1566 reflections  
 111 parameters  
 0 restraints  
 Primary atom site location: structure-invariant  
 direct methods  
 Secondary atom site location: difference Fourier  
 map

Hydrogen site location: inferred from  
 neighbouring sites  
 H-atom parameters constrained  
 $w = 1/[\sigma^2(F_o^2) + (0.1019P)^2 + 0.021P]$   
 where  $P = (F_o^2 + 2F_c^2)/3$   
 $(\Delta/\sigma)_{\text{max}} < 0.001$   
 $\Delta\rho_{\text{max}} = 0.12 \text{ e } \text{\AA}^{-3}$   
 $\Delta\rho_{\text{min}} = -0.22 \text{ e } \text{\AA}^{-3}$   
 Absolute structure: Flack (1983), 592 Friedel  
 pairs  
 Absolute structure parameter: 0.08 (12)

*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 of  $F^2$  against ALL reflections. The weighted R-factor  $wR$  and goodness of fit  $S$  are based on  $F^2$ , conventional R-factors  $R$  are based on  $F$ , with  $F$  set to zero for negative  $F^2$ . The threshold expression of  $F^2 > 2\sigma(F^2)$  is used only for calculating R-factors(gt) etc. and is not relevant to the choice of reflections for refinement. R-factors based on  $F^2$  are statistically about twice as large as those based on  $F$ , and R-factors based on ALL data will be even larger.

*Fractional atomic coordinates and isotropic or equivalent isotropic displacement parameters ( $\text{\AA}^2$ )*

	<i>x</i>	<i>y</i>	<i>z</i>	$U_{\text{iso}}^*/U_{\text{eq}}$
C11	0.35358 (10)	0.29469 (2)	0.1116 (2)	0.0643 (3)
O1	0.9306 (2)	0.40920 (6)	0.5876 (6)	0.0520 (5)
N1	0.5044 (3)	0.47464 (6)	0.4965 (5)	0.0367 (5)
C1	0.6541 (3)	0.40440 (7)	0.3671 (6)	0.0333 (5)
C2	0.8000 (3)	0.38071 (8)	0.4798 (6)	0.0373 (6)
C3	0.8050 (4)	0.33068 (10)	0.4871 (7)	0.0487 (7)
H3	0.9006	0.3150	0.5674	0.058*
C4	0.6670 (3)	0.30434 (8)	0.3743 (7)	0.0468 (7)
H4	0.6702	0.2710	0.3770	0.056*
C5	0.5261 (4)	0.32784 (9)	0.2588 (7)	0.0429 (6)
C6	0.5175 (3)	0.37744 (8)	0.2532 (6)	0.0381 (6)
H6	0.4208	0.3927	0.1736	0.046*
C7	1.0770 (4)	0.38654 (12)	0.7221 (9)	0.0611 (8)
H7A	1.0453	0.3671	0.9111	0.092*
H7B	1.1565	0.4106	0.7929	0.092*

H7C	1.1279	0.3667	0.5532	0.092*
C8	0.6407 (3)	0.45789 (8)	0.3653 (6)	0.0330 (5)
C9	0.7746 (3)	0.48801 (9)	0.2056 (7)	0.0413 (6)
H9A	0.7244	0.5081	0.0369	0.062*
H9B	0.8580	0.4676	0.1039	0.062*
H9C	0.8274	0.5077	0.3734	0.062*

*Atomic displacement parameters (Å<sup>2</sup>)*

	$U^{11}$	$U^{22}$	$U^{33}$	$U^{12}$	$U^{13}$	$U^{23}$
Cl1	0.0758 (5)	0.0434 (4)	0.0738 (5)	−0.0196 (3)	−0.0095 (5)	−0.0004 (4)
O1	0.0404 (9)	0.0499 (10)	0.0656 (13)	0.0104 (8)	−0.0170 (10)	−0.0051 (10)
N1	0.0331 (10)	0.0273 (9)	0.0497 (12)	0.0040 (8)	−0.0013 (9)	0.0009 (9)
C1	0.0368 (12)	0.0316 (12)	0.0315 (11)	0.0055 (9)	0.0014 (11)	0.0012 (9)
C2	0.0404 (13)	0.0386 (12)	0.0329 (12)	0.0102 (10)	−0.0014 (10)	−0.0024 (10)
C3	0.0568 (16)	0.0408 (13)	0.0485 (15)	0.0213 (12)	−0.0006 (13)	0.0047 (12)
C4	0.0618 (17)	0.0286 (11)	0.0500 (15)	0.0080 (11)	0.0084 (15)	0.0008 (11)
C5	0.0567 (16)	0.0327 (12)	0.0392 (13)	−0.0037 (12)	0.0022 (12)	0.0010 (10)
C6	0.0382 (13)	0.0343 (12)	0.0420 (13)	0.0049 (10)	−0.0002 (11)	0.0035 (10)
C7	0.0409 (14)	0.082 (2)	0.0605 (19)	0.0158 (15)	−0.0150 (13)	−0.0022 (17)
C8	0.0340 (11)	0.0311 (11)	0.0337 (11)	0.0024 (9)	−0.0040 (11)	−0.0019 (9)
C9	0.0400 (12)	0.0362 (13)	0.0475 (15)	−0.0018 (11)	0.0057 (11)	−0.0039 (11)

*Geometric parameters (Å, °)*

Cl1—C5	1.748 (3)	C4—C5	1.371 (4)
O1—C2	1.371 (3)	C4—H4	0.9300
O1—C7	1.422 (3)	C5—C6	1.384 (3)
N1—C8	1.285 (3)	C6—H6	0.9300
N1—N1 <sup>i</sup>	1.415 (3)	C7—H7A	0.9600
C1—C6	1.391 (3)	C7—H7B	0.9600
C1—C2	1.403 (3)	C7—H7C	0.9600
C1—C8	1.494 (3)	C8—C9	1.493 (3)
C2—C3	1.395 (4)	C9—H9A	0.9600
C3—C4	1.389 (4)	C9—H9B	0.9600
C3—H3	0.9300	C9—H9C	0.9600
C2—O1—C7	118.2 (2)	C5—C6—H6	120.1
C8—N1—N1 <sup>i</sup>	113.8 (2)	C1—C6—H6	120.1
C6—C1—C2	119.2 (2)	O1—C7—H7A	109.5
C6—C1—C8	118.9 (2)	O1—C7—H7B	109.5
C2—C1—C8	121.9 (2)	H7A—C7—H7B	109.5
O1—C2—C3	123.4 (2)	O1—C7—H7C	109.5
O1—C2—C1	116.53 (19)	H7A—C7—H7C	109.5
C3—C2—C1	120.0 (2)	H7B—C7—H7C	109.5
C4—C3—C2	119.9 (2)	N1—C8—C9	124.3 (2)
C4—C3—H3	120.0	N1—C8—C1	114.8 (2)
C2—C3—H3	120.0	C9—C8—C1	120.9 (2)

C5—C4—C3	119.6 (2)	C8—C9—H9A	109.5
C5—C4—H4	120.2	C8—C9—H9B	109.5
C3—C4—H4	120.2	H9A—C9—H9B	109.5
C4—C5—C6	121.5 (3)	C8—C9—H9C	109.5
C4—C5—C11	119.57 (19)	H9A—C9—H9C	109.5
C6—C5—C11	118.9 (2)	H9B—C9—H9C	109.5
C5—C6—C1	119.7 (2)		
C7—O1—C2—C3	-1.7 (4)	C4—C5—C6—C1	-0.1 (4)
C7—O1—C2—C1	176.4 (2)	C11—C5—C6—C1	-179.54 (19)
C6—C1—C2—O1	179.7 (2)	C2—C1—C6—C5	1.3 (4)
C8—C1—C2—O1	-0.2 (3)	C8—C1—C6—C5	-178.8 (2)
C6—C1—C2—C3	-2.1 (4)	N1 <sup>i</sup> —N1—C8—C9	-3.8 (3)
C8—C1—C2—C3	178.0 (2)	N1 <sup>i</sup> —N1—C8—C1	179.13 (16)
O1—C2—C3—C4	179.8 (2)	C6—C1—C8—N1	48.8 (3)
C1—C2—C3—C4	1.7 (4)	C2—C1—C8—N1	-131.3 (2)
C2—C3—C4—C5	-0.5 (4)	C6—C1—C8—C9	-128.4 (2)
C3—C4—C5—C6	-0.3 (4)	C2—C1—C8—C9	51.5 (3)
C3—C4—C5—C11	179.2 (2)		

Symmetry code: (i)  $-x+1, -y+1, z$ .

*Hydrogen-bond geometry* ( $\text{\AA}$ ,  $^\circ$ )

<i>D</i> —H $\cdots$ <i>A</i>	<i>D</i> —H	H $\cdots$ <i>A</i>	<i>D</i> $\cdots$ <i>A</i>	<i>D</i> —H $\cdots$ <i>A</i>
C9—H9B $\cdots$ O1 <sup>ii</sup>	0.96	2.68	3.521 (3)	146

Symmetry code: (ii)  $x, y, z-1$ .