Acta Crystallographica Section C
Crystal Structure Communications
ISSN 0108-2701

4-(9,10-Dihydroacridin-9-ylidene)-thiosemicarbazide and its five-membered thiazole and six-membered thiazine derivatives

Ivan Potočňák,[a]* Ján Imrich,[a] Ivan Danihel,[a] Jozef Kožíšek[b] and Karel Douglas Klika[c]

[a]Institute of Chemistry, Faculty of Science, P.J. Šafárik University, Moyzesova 11, SK-041 54 Košice, Slovakia, [b]Institute of Physical Chemistry and Chemical Physics, Slovak University of Technology, Radlinského 9, SK-812 37 Bratislava, Slovakia, and [c]Department of Chemistry, University of Turku, Vatselankatu 2, FIN-20014 Turku, Finland
Correspondence e-mail: ivan.potocnak@upjs.sk

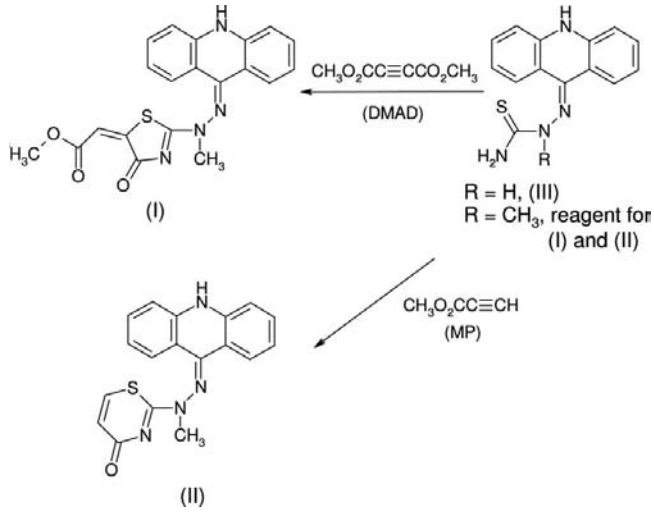
Received 15 December 2009
Accepted 11 January 2010
Online 22 January 2010

Two methyl derivatives, five-membered methyl 2-{2-[2-(9,10-dihydroacridin-9-ylidene)-1-methylhydrazinyl]-4-oxo-4,5-dihydro-1,3-thiazol-5-ylidene}acetate, $C_{20}H_{16}N_4O_3S$, (I), and six-membered 2-[2-(9,10-dihydroacridin-9-ylidene)-1-methylhydrazinyl]-4*H*-1,3-thiazin-4-one, $C_{18}H_{14}N_4OS$, (II), were prepared by the reaction of the *N*-methyl derivative of 4-(9,10-dihydroacridin-9-ylidene)thiosemicarbazide, $C_{14}H_{12}N_4S$, (III), with dimethyl acetylenedicarboxylate and methyl propiolate, respectively. The crystal structures of (I), (II) and (III) are molecular and can be considered in two parts: (i) the nearly planar acridine moiety and (ii) the singular heterocyclic ring portion [thiazolidine for (I) and thiazine for (II)] including the linking amine and imine N atoms and the methyl C atom, or the full side chain in the case of (III). The structures of (I) and (II) are stabilized by N—H···O hydrogen bonds and different π–π interactions between acridine moieties and thiazolidine and thiazine rings, respectively.

Comment

Acridinylthiosemicarbazides are especially interesting for their inherent proclivity towards spiro-open-chain tautomerism (Tomaščiková, Danihel *et al.*, 2008; Tomaščiková, Imrich *et al.*, 2008; Klika *et al.*, 2006*a*,*b*) which can lead to an array of structures and, on occasion, confusion regarding the identity of products arising from a reaction. We had found previously that thiosemicarbazides or thioureas differ in their behaviour towards bielectrophilic reagents in substitution reactions and hence the consequent cyclization to form a heterocyclic ring can yield unexpected or difficult to rationalize structures (Klika *et al.*, 2006*a*,*b*; Klika, Imrich *et al.*, 2006; Balentová *et al.*, 2006).

Herein we continue our study of the products of reactions of thiosemicarbazides with dimethyl acetylenedicarboxylate (DMAD) (Tomaščiková *et al.*, 2007; Tomaščiková, Danihel *et al.*, 2008; Tomaščiková, Imrich *et al.*, 2008) and augment it by contrasting it with methyl propiolate (MP). This is because it seemed plausible to us that control of five- *versus* six-membered ring formation (*i.e.*1,3-thiazolidin-4-one *versus* 1,3-thiazin-4-one products, respectively) using ethyne acid esters could be accomplished by adduct reagent selection.



For unequivocal gross structural confirmation, in particular the size of the newly formed rings, as well as to elucidate the fine structural elements, crystals of three derivatives, *viz*. five-membered methyl 2-{2-[2-(9,10-dihydroacridin-9-ylidene)-1-methylhydrazinyl]-4-oxo-4,5-dihydro-1,3-thiazol-5-ylidene}-acetate, (I), six-membered 2-[2-(9,10-dihydroacridin-9-ylidene)-1-methylhydrazino]-4*H*-1,3-thiazin-4-one, (II), and unsubstituted 4-(9,10-dihydroacridin-9-ylidene)thiosemicarba-

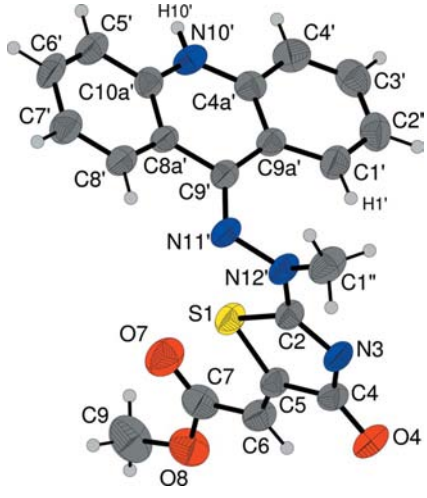


Figure 1
The molecular structure of (I). Displacement ellipsoids are plotted at the 50% probability level and H atoms are shown as small spheres of arbitrary radii.

 doi:10.1107/S0108270110001277

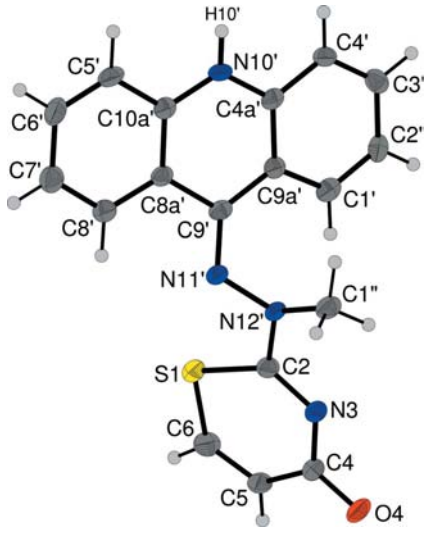


Figure 2
The molecular structure of (II). Displacement ellipsoids are plotted at the 50% probability level and H atoms are shown as small spheres of arbitrary radii.

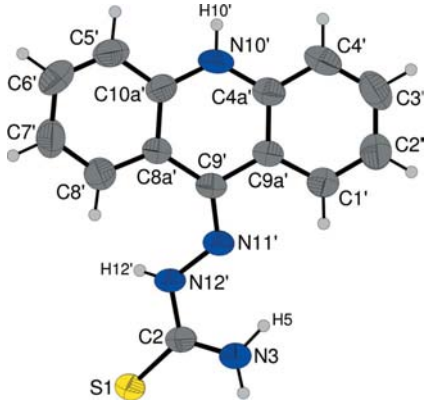


Figure 3
The molecular structure of (III). Displacement ellipsoids are plotted at the 50% probability level and H atoms are shown as small spheres of arbitrary radii. For clarity, only one disordered S atom is depicted.

zide, (III), were obtained (Imrich *et al.*, 2010) and subjected to X-ray single-crystal analysis. Of more than 70 crystallographic descriptions of structures containing an acridine moiety and several hundred descriptions of structures containing thiazolidine rings (Allen, 2002), only four reports exist which contain both acridine and thiazolidine entities linked in some manner (Klika *et al.*, 2001; Tomaščiková, Danihel *et al.*, 2008; Imrich *et al.*, 2005; Černák *et al.*, 1995). Furthermore, there are no reports of structures having been examined that contain both acridine and thiazine entities. The structures obtained for (I), (II) and (III) are shown in Figs. 1, 2 and 3, respectively. Selected bond lengths, angles and torsion angles are presented in Tables 1, 3 and 5 for (I), (II) and (III), respectively. The crystal structures of all three compounds are molecular and can be conveniently considered in two parts: the acridine moiety and the singular heterocyclic ring portion [thiazolidine for (I) and thiazine for (II)] including the linking amine N12′ and imine N11′ atoms and methyl atom C1″ [the second part is the full side chain in the case of (III)].

The geometric parameters for the acridine moiety in (I), (II) and (III) correspond to aromatic portions (outer rings), whilst the central C9′—C9a′ and C8a′—C9′ bonds pertain very much to typical C—C single bonds and overall the parameters are similar to previous reports (Klika *et al.*, 2001; Tomaščiková, Danihel *et al.*, 2008; Imrich *et al.*, 2005; Černák *et al.*, 1995). The placement of a labile H atom on N10′ confirms the NMR assignments (Imrich *et al.*, 2010). Although the acridine moieties are not aromatic over the entire segment, they are, nevertheless, almost planar in all structures; the largest deviations of C9′ from the mean planes of the 14 non-H atoms of the acridine moieties are 0.184 (3) and 0.180 (3) Å for (I) and (II), respectively. These deviations from planarity, though, are larger than the deviation of 0.088 (1) Å observed in a previous study (Tomaščiková, Danihel *et al.*, 2008) for an acridine moiety that was aromatic over the entire segment. On the other hand, not only is the acridine moiety itself in (III) very planar, with the largest deviation [0.101 (3) Å] observed for C7′ from the mean plane of the 14 non-H atoms of the acridine moiety, but even all non-H atoms, excluding the disordered S1 atom, lie nearly in the same plane with the largest deviation [0.197 (3) Å] observed for N3 from the mean plane of the 18 non-H atoms.

The geometric parameters for the 1,3-thiazolidin-4-one ring in (I) resemble those found in similar compounds containing a 1,3-thiazolidin-4-one ring (Tomaščiková, Danihel *et al.*, 2008; Orrell & Wallis, 1984; Heravi *et al.*, 2006; Cameron & Hair, 1971; Deepthi *et al.*, 2001). Also, the geometric parameters of the 1,3-thiazin-4-one ring in (II) are similar to those reported in the literature (Hilton *et al.*, 1994). Both of the 1,3-thiazolidin-4-one and 1,3-thiazin-4-one rings of (I) and (II), respectively, are also planar. Moreover, not only are these singular heterocyclic rings themselves planar, but all of the heavy atoms from N11′ on one side of the rings, including methyl atom C1″, all the way to O8 on the other side of the 1,3-thiazolidin-4-one ring for (I) lie almost in the one plane in each case. The largest deviation, 0.166 (2) Å, from the mean plane of these 13 atoms in (I) was observed for O8. For (II), the largest deviation, 0.189 (3) Å, was observed for N11′. Most interestingly, the angle between the acridine ring plane and the plane of the singular heterocyclic rings was 79.61 (3)° in (I), whilst in (II) the corresponding angle was only 56.48 (4)°. These compare to dihedral angles of *ca* 59° and *ca* 58° for (I) and (II), respectively, from modelling studies (Böhm *et al.*, 2009). Thus, only in the solid state is a wide dihedral angle observed. As a result of the larger dihedral angle in the crystal structure of (I), a weak intramolecular C1′—H1′···N12′ hydrogen bond (see Table 2) is present in (I) but not in (II) and the N12′—C2 bond in (I) is much shorter than in (II) as a consequence [1.309 (3) *versus* 1.341 (4) Å, respectively]. This is consistent with the known conformational mobility and geometric flexibility of N which can easily adopt flattened geometries (Tähtinen *et al.*, 2003; Rosling, Hotokka *et al.*, 1999; Rosling *et al.*, 1999*a*,*b*; Pawłowicz *et al.*, 2006, 2007; Olsen *et al.*, 2007; Klika, Mäki *et al.*, 2006) to permit such interactions,

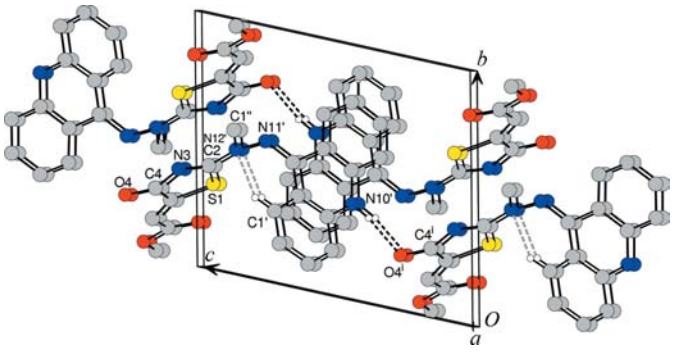


Figure 4
Packing diagram showing the parallel stacking of molecules in (I). Intermolecular hydrogen bonding, N10′—H10′···O4^{i} (solid black dashed line), links the molecules into chains and the π-stacking of both the acridine moieties (acridine-to-acridine) and the thiazolidine rings (thiazolidine-to-thiazolidine) is evident. Intramolecular hydrogen bonding, C1′—H1′···N12′ (hollow dashed line), is also present. For clarity, only H atoms involved in hydrogen bonds are depicted. [Symmetry code: (i) x, y, $z-1$.]

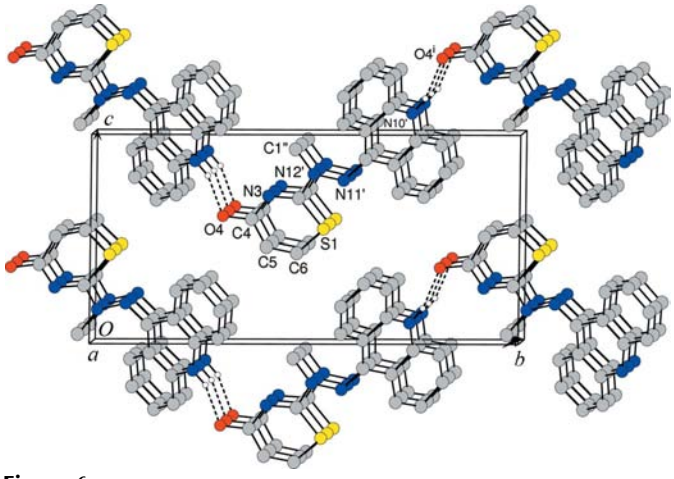


Figure 6
Packing diagram showing the parallel stacking of molecules in (II). Intermolecular hydrogen bonding, N10′—H10′···O4^{i}, links the molecules into chains. For clarity, only H atoms involved in hydrogen bonds are depicted. [Symmetry code: (i) $-x+1$, $y+\frac{1}{2}$, $-z+2$.]

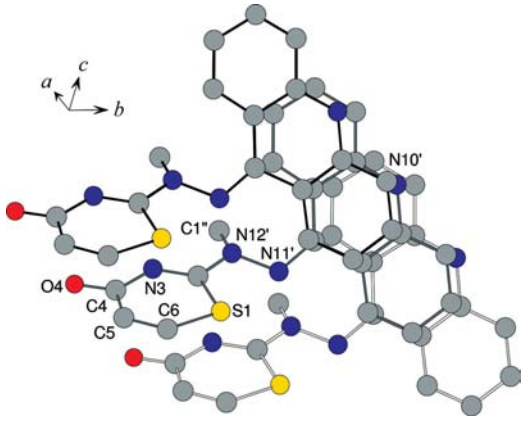


Figure 5
View of the π-stacking in (II), showing an overlap of two of the three six-membered rings of neighbouring acridine moieties. For clarity, H atoms have been omitted and black, grey and hollow lines are used to represent bonds in the upper, middle and bottom molecules, respectively.

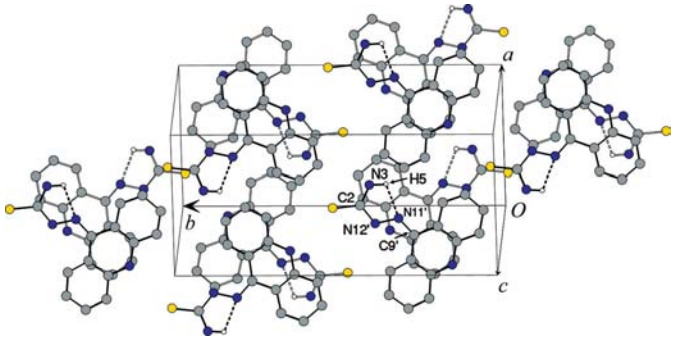


Figure 7
Packing diagram showing the parallel stacking of acridine units in (III). Intramolecular N3—H5···N11′ hydrogen bonding and the π-stacking of the central and outer rings is evident. For clarity, only H atoms involved in hydrogen bonds are depicted and hollow lines are used to represent bonds in the bottom molecules.

although in this instance the geometric permutation is driven by other factors since N12′ is also flattened in (II) and the only requirement is for an increase in the dihedral angle between the segmental planes to enable the hydrogen-bonding interaction to occur.

The geometric parameters between non-H atoms in the side chain of (III) correspond to the single and double bonds represented between the associated atoms. The H atoms of the two amine groups were all located from a difference electron map with all N—H bond distances adopting typical values, as do the angles concerned with N3. On the other hand, the C2—N12′—H12′ angle of 112 (2)° strongly deviates from the expected value of 120°. As a consequence, the H12′ atom deviates from the mean plane of all seven atoms of the side chain (excluding the disordered S1 atom) by 0.25 (3) Å. Probably because of the terminal bonding of the S1 atom in the side chain, the thermal motion of this atom is much larger than the motion of the S1 atoms in the 1,3-thiazolidin-4-one and 1,3-thiazin-4-one rings of (I) and (II), respectively. Disordering of the S1 atom in (III) over two positions with site-occupation factors of 0.730 (15) and 0.270 (15) resulted in a substantially better R factor and goodness-of-fit values in comparison to a model containing a single S1 atom, hence the preference for a model with a statistically disordered S1 atom. The Z configuration about the C5═C6 double bond and the *s–cis* configuration of the C5═C6—C7═O7 segment in (I) were both re-confirmed (Tomaščiková, Danihel *et al.*, 2008).

Regarding the crystal packing of the molecules, for both (I) and (II) (see Figs. 4, 5 and 6) intermolecular N10′—H10′···O4^{i} hydrogen bonds [symmetry code: (i) x, y, $z-1$; Tables 2 and 4] link pairs of molecules, giving rise to the formation of chains of molecules. By contrast, in (III) only intramolecular hydrogen bonding is present, *viz.* N3—H5···N11′ (Table 6), and thus the molecules are, for all intents and purposes, isolated from one another. For both compounds (I) and (II), the acridine rings

exhibit π–π stacking to other acridine units in neighbouring molecules. Probably because of the different dihedral angles for the two compounds between the planes of the acridine and singular heterocyclic rings, the stacking of the acridine moieties, however, is different for the two compounds. In (I), the neighbouring acridine moieties, with a centroid–centroid distance of 4.03 (1) Å, are staggered similarly as in graphite and with acridine moieties from next-but-one planes wholly aligned. In (II), the acridine moieties from neighbouring layers are shifted with respect to one another [centroid–centroid distance = 3.60 (1) Å] and thus only two of the three six-membered rings of neighbouring acridine moieties are appropriately aligned (see Fig. 5). The distances between the mean planes of neighbouring acridine moieties are 3.64 (1) and 3.42 (1) Å for (I) and (II), respectively, and the offset angles are 25.6 and 18.9° for (I) and (II), respectively. Thus, it can be realized that these face-to-face π-stacking interactions dictate to a certain degree the crystal packing of the molecules. Such parallel π-stacking with ring separations of 3.3–3.8 Å is an important noncovalent organizational force in supramolecular aggregates (Janiak *et al.*, 2000), although in this instance the contribution of the intermolecular hydrogen-bonding interactions to the crystal packing of the molecules dominates. Nevertheless, the alignment of the planes between neighbouring acridine moieties appears to be influenced by the π–π interactions. The respective planes of neighbouring thiazolidine rings in (I) and thiazine rings in (II) are also parallel and the distances between the mean planes of these rings are 3.58 (1) and 3.97 (1) Å, respectively. However, the thiazine rings in (II) are staggered [centroid–centroid distance = 4.70 (1) Å] and therefore considerable π–π interactions can only be expected between neighbouring thiazolidine rings in (I) and as borne out by its more comparable interplanar distance. Since the natural interplanar angle for both (I) and (II) seems to be acute, based on the calculated isolated states for both (I) and (II) (Böhm *et al.*, 2009), the allowance for greater π–π interaction between the thiazolidine rings in (I) in the solid state leads to an increase in the interplanar angle. The resultant obtuse angle then promulgates change to the acridine–acridine interactions for (I), hence the observed differences between (I) and (II).

The acridine rings in (III) exhibit different π–π stacking than in (I) and (II). Acridine units in neighbouring molecules of (III) are nearly planar; however, they are not in a parallel but in an almost perpendicular orientation [83.2 (1)°; see Fig. 7]. Thus, only the central ring from one acridine unit is overlapped with one of the outer rings of an acridine unit from a neighbouring molecule with the dihedral angle between these two rings being 6.0 (1)°. The distance between the centroids of these two rings is 3.62 (1)Å, while the offset angle is 5.2°.

Experimental

Compounds (I), (II) and (III) were prepared according to the method of Imrich *et al.* (2010) and recrystallized from hot saturated methanol solutions.

Compound (I)

Crystal data

$C_{20}H_{16}N_4O_3S$
M_r = 392.43
Triclinic, $P\overline{1}$
a = 9.059 (2) Å
b = 10.238 (3) Å
c = 10.983 (2) Å
α = 74.18 (2)°
β = 76.324 (18)°
γ = 71.74 (2)°
V = 917.9 (4) Å^3
Z = 2
Mo $K\alpha$ radiation
μ = 0.21 mm^{-1}
T = 298 K
0.58 × 0.07 × 0.05 mm

Data collection

Oxford Diffraction Gemini R CCD diffractometer
Absorption correction: analytical (Clark & Reid, 1995)
T_{min} = 0.987, T_{max} = 0.992
9089 measured reflections
3237 independent reflections
1591 reflections with $I > 2\sigma(I)$
R_{int} = 0.057

Refinement

$R[F^2 > 2\sigma(F^2)]$ = 0.040
$wR(F^2)$ = 0.079
S = 0.89
3237 reflections
255 parameters
H-atom parameters constrained
$\Delta\rho_{max}$ = 0.18 e Å^{-3}
$\Delta\rho_{min}$ = −0.18 e Å^{-3}

Compound (II)

Crystal data

$C_{18}H_{14}N_4OS$
M_r = 334.39
Monoclinic, $P2_1$
a = 4.7035 (2) Å
b = 18.5073 (4) Å
c = 9.1172 (2) Å
β = 102.689 (3)°
V = 774.26 (4) Å^3
Z = 2
Mo $K\alpha$ radiation
μ = 0.22 mm^{-1}
T = 100 K
0.12 × 0.08 × 0.04 mm

Data collection

Oxford Diffraction Gemini R CCD diffractometer
Absorption correction: analytical (Clark & Reid, 1995)
T_{min} = 0.968, T_{max} = 0.993
3900 measured reflections
2659 independent reflections
2207 reflections with $I > 2\sigma(I)$
R_{int} = 0.024

Refinement

$R[F^2 > 2\sigma(F^2)]$ = 0.042
$wR(F^2)$ = 0.095
S = 1.04
2659 reflections
218 parameters
1 restraint
H-atom parameters constrained
$\Delta\rho_{max}$ = 0.41 e Å^{-3}
$\Delta\rho_{min}$ = −0.25 e Å^{-3}
Absolute structure: Flack (1983), 1240 Friedel pairs
Flack parameter: −0.03 (9)

Compound (III)

Crystal data

$C_{14}H_{12}N_4S$
M_r = 268.34
Monoclinic, $P2_1/c$
a = 8.6535 (5) Å
b = 18.6374 (9) Å
c = 7.8350 (6) Å
β = 99.372 (6)°
V = 1246.75 (13) Å^3
Z = 4
Mo $K\alpha$ radiation
μ = 0.25 mm^{-1}
T = 293 K
0.36 × 0.12 × 0.08 mm

Data collection

Oxford Diffraction Xcalibur2 diffractometer with a Sapphire2 CCD detector
Absorption correction: analytical (Clark & Reid, 1995)
T_{min} = 0.942, T_{max} = 0.982
13277 measured reflections
2208 independent reflections
1077 reflections with $I > 2s(I)$
R_{int} = 0.046

Table 1
Selected geometric parameters (Å, °) for (I).

C5—C6	1.330 (3)	C8*A*′—C9′	1.473 (3)
C6—C7	1.457 (4)	C9*A*′—C9′	1.464 (3)
C7—O7	1.203 (3)	C9′—N11′	1.318 (3)
C7—O8	1.346 (3)	N11′—N12′	1.418 (3)
O8—C9	1.450 (3)		
C9′—N11′—N12′	117.8 (2)	C2—N12′—N11′	120.6 (2)
C5—C6—C7—O7	7.9 (5)	C9′—N11′—N12′—C2	−99.6 (3)
C9*A*′—C9′—N11′—N12′	8.7 (4)		

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

D—H···*A*	*D*—H	H···*A*	*D*···*A*	*D*—H···*A*
N10′—H10′···O4^{i}	0.86	2.13	2.974 (3)	165
C1′—H1′···N12′	0.93	2.29	2.899 (4)	122

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

Table 3
Selected geometric parameters (Å, °) for (II).

C8*A*′—C9′	1.468 (4)	C9′—N11′	1.312 (4)
C9*A*′—C9′	1.468 (5)	N11′—N12′	1.417 (3)
C9′—N11′—N12′	118.1 (2)	C2—N12′—N11′	116.0 (2)
C9*A*′—C9′—N11′—N12′	10.6 (5)		

Table 4
Hydrogen-bond geometry (Å, °) for (II).

D—H···*A*	*D*—H	H···*A*	*D*···*A*	*D*—H···*A*
N10′—H10′···O4^{i}	0.86	2.02	2.837 (3)	159

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

Table 5
Selected geometric parameters (Å, °) for (III).

N3—C2	1.286 (3)	N11′—C9′	1.304 (2)
C2—N12′	1.349 (3)	C9′—C8A'	1.454 (3)
N12′—N11′	1.383 (2)	C9′—C9A'	1.464 (3)
C2—N12′—N11′	117.4 (2)	C9′—N11′—N12′	120.75 (18)
N3—C2—N12′—N11′	8.4 (3)	N12′—N11′—C9′—C9A'	−176.6 (2)

Table 6
Hydrogen-bond geometry (Å, °) for (III).

D—H···*A*	*D*—H	H···*A*	*D*···*A*	*D*—H···*A*
N3—H5···N11′	0.96 (3)	2.11 (3)	2.554 (3)	106.3 (19)

Refinement

$R[F^2 > 2\sigma(F^2)] = 0.040$	H atoms treated by a mixture of independent and constrained refinement
$wR(F^2) = 0.077$	
$S = 0.97$	
2208 reflections	$\Delta\rho_{max} = 0.13$ e Å^{-3}
198 parameters	$\Delta\rho_{min} = -0.14$ e Å^{-3}

All H atoms of (I) and (II), as well as the C-bound H atoms of (III), were placed in calculated positions and refined riding on their parent C/N atoms, with C—H = 0.93 (aromatic) or 0.96 Å (methyl) and N—H = 0.86 Å, and with U_{iso}(H) = 1.5U_{eq}(C) for the methyl groups and 1.2U_{eq}(C,N) otherwise. The amine H atoms of (III) were found in a difference electron-density map and were refined with free isotropic displacement parameters.

For all compounds, data collection: *CrysAlis CCD* (Oxford Diffraction, 2006); cell refinement: *CrysAlis RED* (Oxford Diffraction, 2006); data reduction: *CrysAlis RED*; program(s) used to solve structure: *SHELXS97* (Sheldrick, 2008); program(s) used to refine structure: *SHELXL97* (Sheldrick, 2008); molecular graphics: *DIAMOND* (Brandenburg, 2000); software used to prepare material for publication: *SHELXL97* and *PARST* (Nardelli, 1995).

Financial support for this work by the Slovak Grant Agency VEGA (grant No. 1/0476/08) and Turun Yliopistosäätiö is gratefully acknowledged.

Supplementary data for this paper are available from the IUCr electronic archives (Reference: DN3135). Services for accessing these data are described at the back of the journal.

References

Allen, F. H. (2002). *Acta Cryst.* B**58**, 380–388.

Balentová, E., Imrich, J., Bernát, J., Suchá, L., Vilková, M., Prónayová, N., Kristian, P., Pihlaja, K. & Klika, K. D. (2006). *J. Heterocycl. Chem.* **43**, 645–656.

Böhm, S., Tomaščiková, J., Imrich, J., Danihel, I., Kristian, P., Koch, A., Kleinpeter, E. & Klika, K. D. (2009). *J. Mol. Struct. (THEOCHEM)*, **916**, 105–118.

Brandenburg, K. (2000). *DIAMOND*. Release 2.1e. Crystal Impact GbR, Bonn, Germany.

Cameron, A. F. & Hair, N. J. (1971). *J. Chem. Soc. B*, pp. 1733–1736.

Černák, J., Kristian, P., Bernát, J. & Lipkowski, J. (1995). *Acta Cryst.* C**51**, 2397–2399.

Clark, R. C. & Reid, J. S. (1995). *Acta Cryst.* A**51**, 887–897.

Deepthi, S., Rajalakshmi, K., Gunasekaran, K., Velmurugan, D. & Nagarajan, K. (2001). *Mol. Cryst. Liq. Cryst.* **369**, 221–242.

Flack, H. D. (1983). *Acta Cryst.* A**39**, 876–881.

Heravi, M. M., Nami, N., Neumuller, B., Oskooie, H. A. & Hekmatshoar, R. (2006). *J. Chem. Res.* pp. 722–724.

Hilton, M., Fisk, P., Odell, B., Rzepa, H. S., Williams, D. J. & Yi, M. Y. (1994). *J. Chem. Soc. Perkin Trans. 2*, pp. 185–188.

Imrich, J., Tomaščiková, J., Danihel, I., Kristian, P., Böhm, S. & Klika, K. D. (2010). *Heterocycles*, **80**, 489–503.

Imrich, J., Vilková, M. & Černák, J. (2005). *Acta Cryst.* C**61**, o231–o233.

Janiak, C., Temizdemir, S., Dechert, S., Deck, W., Girgsdies, F., Heinze, J., Kolm, M. J., Scharmann, T. G. & Zipffel, O. M. (2000). *Eur. J. Inorg. Chem.* pp. 1229–1241.

Klika, K. D., Balentová, E., Bernát, J., Imrich, J., Vavrušová, M., Kleinpeter, E., Pihlaja, K. & Koch, A. (2006*a*). *J. Heterocycl. Chem.* **43**, 633–643.

Klika, K. D., Balentová, E., Bernát, J., Imrich, J., Vavrušová, M., Pihlaja, K., Koch, A., Kleinpeter, E., Kelling, A. & Schilde, U. (2006*b*). *ARKIVOC*, **xvi**, 93–108.

Klika, K. D., Bernát, J., Imrich, J., Chomča, I., Sillanpää, R. & Pihlaja, K. (2001). *J. Org. Chem.* **66**, 4416–4418.

Klika, K. D., Imrich, J., Vilková, M., Bernát, J. & Pihlaja, K. (2006). *J. Heterocycl. Chem.* **43**, 739–743.

Klika, K. D., Mäki, J., Sjöholm, R. & Kronberg, L. (2006). *ARKIVOC*, **xii**, 65–74.

Nardelli, M. (1995). *J. Appl. Cryst.* **28**, 659.
Olsen, R., Backman, J., Molander, P., Klika, K. D. & Kronberg, L. (2007). *Eur. J. Org. Chem.* pp. 4011–4018.
Orrell, A. P. K. & Wallis, J. D. (1984). *J. Chem. Soc. Perkin Trans. 2*, pp. 227–230.
Oxford Diffraction (2006). *CrysAlis CCD* and *CrysAlis RED*. Oxford Diffraction Ltd, Abingdon, Oxfordshire, England.
Pawłowicz, A. J., Klika, K. D. & Kronberg, L. (2007). *Eur. J. Org. Chem.* pp. 1429–1437.
Pawłowicz, A. J., Munter, T., Klika, K. D. & Kronberg, L. (2006). *Bioorg. Chem.* **34**, 39–48.
Rosling, A., Hotokka, M., Klika, K. D., Fülöp, F., Sillanpää, R. & Mattinen, J. (1999). *Acta Chem. Scand.* **53**, 213–221.
Rosling, A., Klika, K. D., Fülöp, F., Sillanpää, R. & Mattinen, J. (1999*a*). *Heterocycles*, **51**, 2575–2588.
Rosling, A., Klika, K. D., Fülöp, F., Sillanpää, R. & Mattinen, J. (1999*b*). *Acta Chem. Scand.* **53**, 103–113.
Sheldrick, G. M. (2008). *Acta Cryst.* A**64**, 112–122.
Tähtinen, P., Bagno, A., Klika, K. D. & Pihlaja, K. (2003). *J. Am. Chem. Soc.* **125**, 4609–4618.
Tomaščiková, J., Danihel, I., Böhm, S., Imrich, J., Kristian, P., Potočňák, I., Čejka, J. & Klika, K. D. (2008). *J. Mol. Struct.* **875**, 419–426.
Tomaščiková, J., Imrich, J., Danihel, I., Böhm, S. & Kristian, P. (2007). *Collect. Czech. Chem. Commun.* **72**, 347–362.
Tomaščiková, J., Imrich, J., Danihel, I., Böhm, S., Kristian, P., Pisarčíková, J., Sabol, M. & Klika, K. D. (2008). *Molecules*, **13**, 501–518.