Synthesis of new 3,4-di- and 1,2,3,4-tetrahydroquinazolin-4-one derivatives and X-ray diffraction study of crystal solvates of 3-methylsulfonylamino-2-(2-methylsulfonylaminophenyl)-1,2,3,4-tetrahydroquinazolin-4-one

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The reactions of hydrazide, mesyl hydrazide, succinyl hydrazide, and maleyl hydrazide of anthranilic acid with carbonyl compounds were studied, and new di- and tetrahydroquinazolin-4-one derivatives were prepared. The structure of one reaction product, *viz.*, 3-methylsulfonylamino-2-(2-methylsulfonylaminophenyl)-1,2,3,4-tetrahydroquinazolin-4-one, was established by X-ray diffraction study of two crystal solvates of this compound. The characteristic features of the crystal packings of these solvates are discussed.

Key words: anthranilic acid hydrazide, anthranilic acid mesyl hydrazide, succinic anhydride, maleic anhydride, carbonyl compounds, di- and tetrahydroquinazolin-4-one derivatives, supramolecular architecture.

First representatives of 1,2,3,4-tetrahydroquinazolin-4-ones were synthesized by condensation of *o*-aminobenzanilides with formaldehyde in 1904.[1] More recently, it has appeared that many such compounds are physiologically active. Nowadays, there are abundant data on these compounds (see, for example, Refs 2—4 and references therein).

We synthesized new substituted 1,2,3,4-tetrahydroquinazolin-4-ones with the use of anthranilic acid mesyl hydrazide **1**, which we have prepared for the first time. It was found that compound **1** smoothly reacts with aldehydes **2** on heating in PrOH to form 1,2,3,4-tetrahydroquinazolin-4-ones **3** and is transformed into 3-methylsulfonylamino-3,4-dihydroquinazolin-4-one (**4**) on refluxing with formic acid (Scheme 1).

Under the same conditions, the reaction of anthranilic acid hydrazide **5** with furfural produced quinazolinone **6**. Refluxing of hydrazide **5** with formic acid afforded 3-amino-3,4-dihydroquinazolin-4-one (**7**) (Scheme 2).

Cyclic ketones can be condensed with anthranilamides and their analogs to form spirocyclic structures.[4—6] The simplest method giving rise to the target products in high yields is based on heating of the reactants under solvent-free conditions without catalysts.[7]

We found that refluxing of hydrazide **1** with cyclopentanone or cyclohexanone in ethylene glycol afforded 3-methylsulfonylamino-2-spirocyclopentane-1,2,3,4-tetrahydroquinazolin-4-one (**8**) and 3-methylsulfonylamino-2-spirocyclohexane-1,2,3,4-tetrahydroquinazolin-4-one (**9**), respectively. Spiro derivatives **10** and **11** were prepared from hydrazide **5** and the same cyclic ketones under solvent-free conditions (Scheme 3).

Refluxing of hydrazide **1** with acetylacetone gave 2-methyl-3-methylsulfonylamino-3,4-dihydroquinazolin-4-one (**12**) presumably *via* elimination of acetone from intermediate tetrahydro derivative **A** (Scheme 4).

It is known[8] that anthranilhydrazide **5** is acylated with cyclic dicarboxylic acid anhydrides at the hydrazine nitrogen atom. Heating of this reaction product with succinic anhydride in acetic anhydride followed by hydrolysis afforded 3-(3-carboxypropanoylamino)-2-methylquinazolin-4-one.[8]

We used acylation products of anthranilhydrazide **5** with succinic and maleic anhydrides in reactions with aldehydes to synthesize 1,2,3,4-tetrahydroquinazolin-4-one derivatives **13** and **14**, respectively. The reactions were carried out without isolation of intermediate acylation products **B** and **C**. Analogous reactions can be performed with cyclic ketones, as was exemplified by com-

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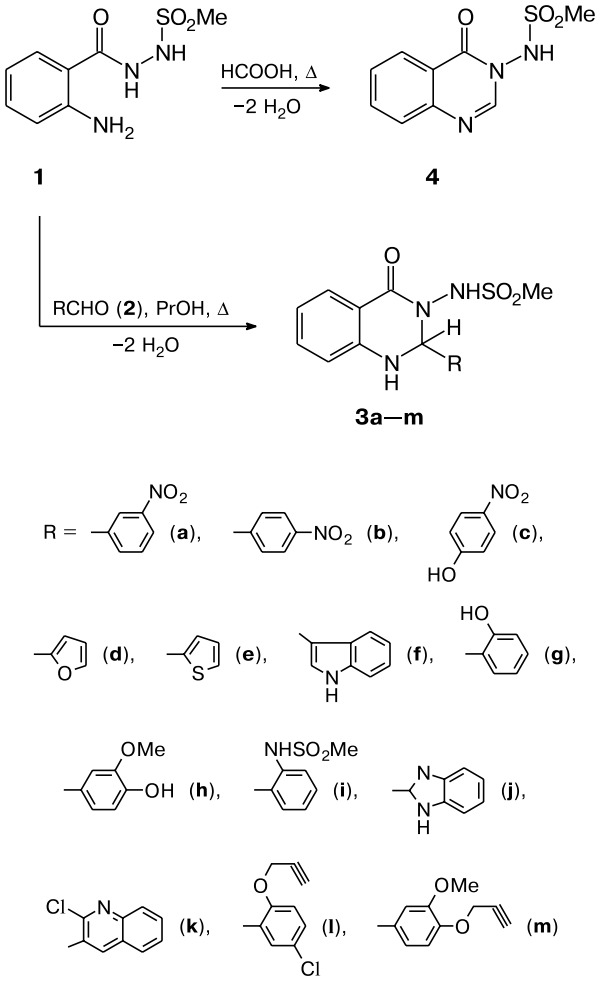
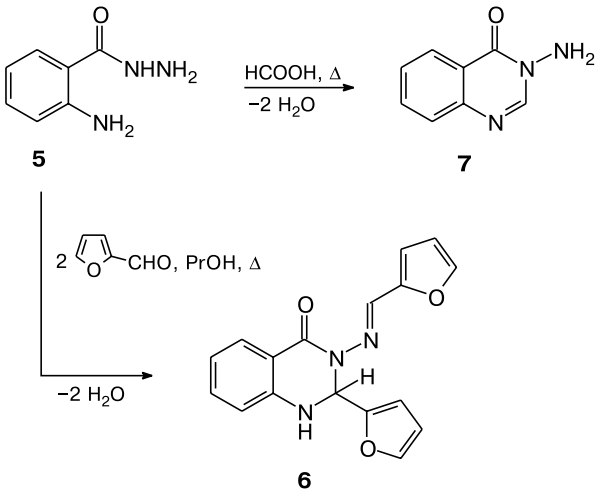
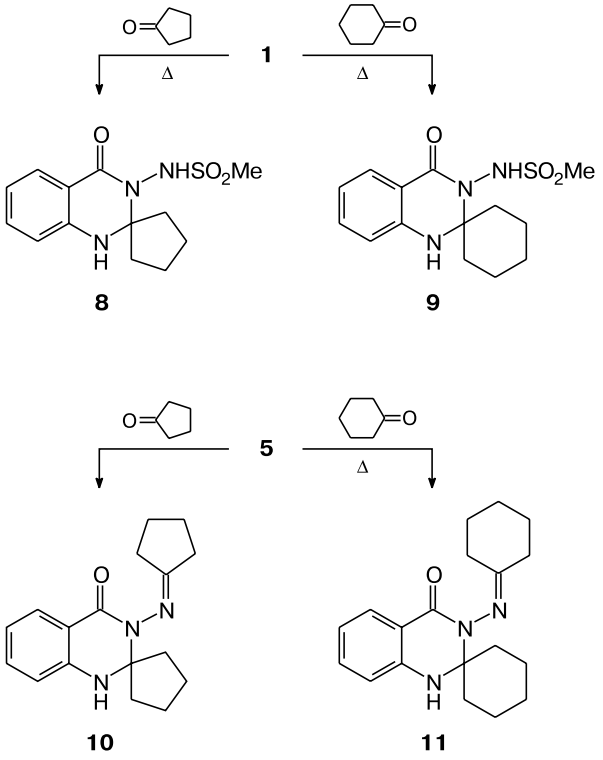
Scheme 1



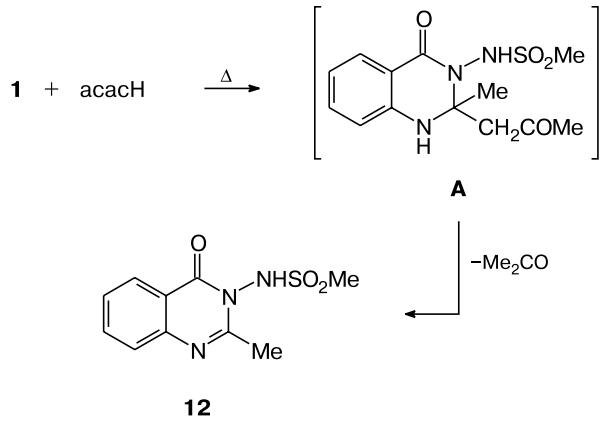
Scheme 2



Scheme 3



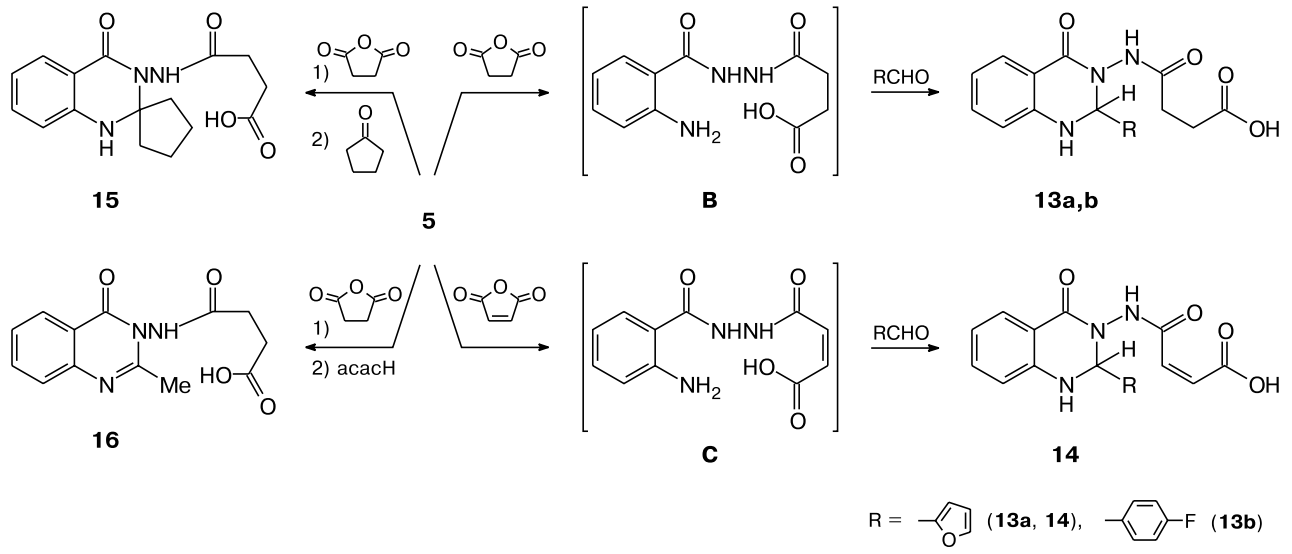
Scheme 4



pound **15**. The reaction of anthranilhydrazide **5**, succinic anhydride, and acetylacetone produced 3-(3-carboxypropanoylamino)-2-methylquinazolin-4-one (**16**) described earlier.[8]*

* The positions of the signals in the ^{1}H NMR spectrum of the compound synthesized in the study[8] are virtually identical to our data. However, the melting point of this compound is ~100 °C lower than that determined in the present study, which is apparently a technical error.

Scheme 5



The structure of compound **3i** was established by X-ray diffraction. Crystals were grown from solutions in nitromethane and acetonitrile as 1 : 1 crystal solvates **3i** · $MeNO_2$ and **3i** · MeCN. These crystal solvates were studied by X-ray diffraction (Fig. 1, Table 1). The corresponding bond lengths and bond angles in the basic molecules of the crystals of **3i** · $MeNO_2$ and **3i** · MeCN are identical within experimental error.

In molecules **3i** · $MeNO_2$ and **3i** · MeCN, the N(1) atom has a nearly planar-trigonal bond configuration. The sum of the bond angles at the N(1) atom (~355—356°) is close to the ideal value of 360° characteristic of the planar-trigonal bond configuration. The geometry of the bonds at the N(2) atom is pyramidal (the sum of the bond angles is ~342—343°). There is no conjugation between the lone electron pair of the N(1) atom and the corresponding aromatic system. The S(1)—N(1)—C(3)—C(4) torsion angle is 94.7(2) and 99.5(3)° in the basic molecules of **3i** · $MeNO_2$ and **3i** · MeCN, respectively, which is evidence for the nearly perpendicular mutual orientation of the bond systems of the nitrogen atoms and the aromatic fragment. The dihedral angle between the plane through the S(1),C(3),H(1) atoms and the plane of the benzene ring C(2)...C(7) (~59° in **3i** · $MeNO_2$ and ~57° in **3i** · MeCN) indicates that the orientation of the lone pair of this atom is unfavorable for its conjugation with the benzene ring.

The heterocycle of the tricyclic system adopts a twist-boat conformation. The C(8)—C(9)—C(14)—N(3) fragment is planar, and the C(1) and N(4) atoms deviate from this plane in the same direction, the deviations being different. In **3i** · $MeNO_2$, the deviations of the C(1) and N(4) atoms are 0.592 and 0.184 Å, respectively; in **3i** · MeCN, 0.538 and 0.133 Å.

The dihedral angle between the mean plane of the six-membered heterocycle and the plane of the benzene ring C(2)...C(7) is 87° and 83° in **3i** · $MeNO_2$ and **3i** · MeCN, respectively.

It should be noted that the conformational parameters of the basic molecules of the crystal solvates have similar values. The dihedral angles between different fragments of the molecule in the crystals of **3i** · $MeNO_2$ are very similar to those in **3i** · MeCN (Fig. 2).

Such a detailed similarity of the conformational parameters of the molecule in the crystal solvates under consideration is indicative of the similarity of their crystal environment. This is unusual taking into account the difference in the nature of the solvate molecules and the fact that **3i** · $MeNO_2$ and **3i** · MeCN crystallize in different space groups. It is well known that nitromethane is not involved in weak interactions, *i.e.*, it generally has no effect on the supramolecular architecture because it is located in the cavities of the crystal structure. Unlike nitromethane, acetonitrile can form hydrogen bonds. Taking into account the presence of three "active" protons in molecule **3i**, hydrogen bonding between the basic molecule and the acetonitrile molecule would be expected to occur in crystal solvate **3i** · MeCN. However, the present study demonstrated that in neither crystal structure are the solvate molecules involved in weak ineractions with the basic molecules; instead the solvate molecules are located in the cavities of the crystal packing.

In the crystal packing of **3i** · $MeNO_2$, the molecules are linked to each other to form infinite ribbons through hydrogen bonds involving all "active" protons (Fig. 3).

The basic molecules in the crystal structure of **3i** · MeCN form analogous ribbons.

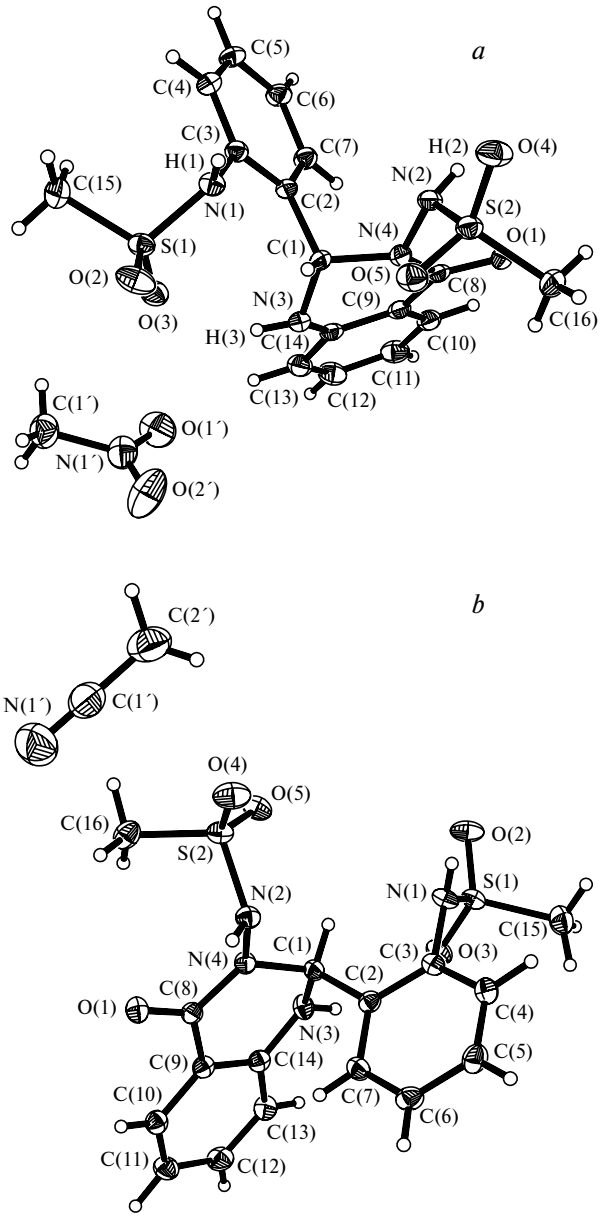


Fig. 1. Structures of the structural units of compound **3i** in crystal solvates **3i**·$MeNO_2$ (*a*) and **3i**·MeCN (*b*). The same atomic numbering scheme is used for molecule **3i** in both crystal solvates.

Table 1. Selected bond lengths (d/Å) and bond angles (ω/deg) in crystal solvates **3i**·$MeNO_2$ and **3i**·MeCN

Parameter	**3i**·$MeNO_2$	**3i**·MeCN
Bond	d/Å	
S(1)—O(2)	1.435(1)	1.439(2)
S(1)—O(3)	1.436(1)	1.440(2)
S(2)—O(4)	1.434(1)	1.437(2)
S(2)—O(5)	1.430(1)	1.433(2)
S(1)—N(1)	1.616(1)	1.619(2)
S(2)—N(2)	1.652(1)	1.653(2)
S(1)—C(15)	1.756(2)	1.762(3)
S(2)—C(16)	1.756(2)	1.762(3)
N(1)—C(3)	1.439(2)	1.439(3)
N(2)—N(4)	1.405(2)	1.403(3)
N(3)—C(14)	1.380(2)	1.383(3)
N(4)—C(1)	1.473(2)	1.476(3)
C(1)—N(3)	1.453(2)	1.462(3)
C(1)—C(2)	1.522(2)	1.525(3)
C(2)—C(3)	1.394(2)	1.400(3)
C(3)—C(4)	1.391(2)	1.403(4)
C(4)—C(5)	1.385(2)	1.386(4)
C(5)—C(6)	1.392(2)	1.393(4)
C(6)—C(7)	1.389(2)	1.387(4)
C(7)—C(2)	1.398(2)	1.399(3)
C(8)—O(1)	1.232(2)	1.234(3)
C(9)—C(8)	1.467(2)	1.478(3)
C(14)—C(9)	1.403(2)	1.404(4)
Bond angle	ω/deg	
O(2)—S(1)—O(3)	118.0(1)	118.6(1)
O(2)—S(1)—N(1)	106.4(1)	106.3(1)
O(2)—S(1)—C(15)	108.7(1)	108.2(1)
O(3)—S(1)—N(1)	107.9(1)	107.9(1)
O(3)—S(1)—C(15)	107.8(1)	107.5(1)
O(4)—S(2)—O(3)	119.5(1)	119.3(1)
O(4)—S(2)—N(2)	103.4(1)	103.3(1)
O(4)—S(2)—C(16)	109.2(1)	109.4(1)
O(5)—S(2)—N(2)	107.0(1)	107.2(1)
O(5)—S(2)—C(16)	108.9(1)	108.8(1)
N(1)—S(1)—C(15)	107.7(1)	107.9(1)
N(2)—S(2)—C(16)	108.4(1)	108.2(1)
S(1)—N(1)—C(3)	120.8(1)	121.6(2)
S(2)—N(2)—N(4)	117.3(1)	117.4(2)

In both crystals, the infinite chains are formed through centrosymmetric pairs of hydrogen bonds resulting in the closure of six-membered rings. One pair of hydrogen bonds exists in the N(1B)—H(1B)...O(2D) and N(1D)—H(1D)...O(2B) fragments in both crystals (see Fig. 3). In **3i**·$MeNO_2$, the N...O distance is 2.872(2) Å, the H...O distance is 2.13(3) Å, and the angle at the H atom is 170(1)°. In **3i**·MeCN, the corresponding parameters are 2.842(3) Å, 2.01(4) Å, and 168(2)°. Another pair of hydrogen bonds links the N(2B)—H(2B)...O(1C) and N(2C)—H(2C)...O(1B) fragments in the crystals of **3i**·$MeNO_2$ and the N(2A)—H(2A)...O(1D) and N(2D)—H(2D)...O(1A) fragments in the crystals of **3i**·MeCN. The parameters of these interactions in **3i**·$MeNO_2$ are as follows: N...O, 2.851(2) Å; H...O, 1.96(3) Å; N—H...O, 176(1)°. The corresponding parameters in **3i**·MeCN are 2.833(3) Å, 2.00(4) Å, and 172(2)°. The geometric parameters correspond to medium-strength hydrogen bonds.

The remaining "active" proton at the N(3) atom in the above-considered ribbon associate is located in the vicinity of the O(3) atom of the basic molecule. The N(3)...O(3) and H(3)...O(3) distances are 2.963(2) and 2.38(4) Å, respectively, in molecule **3i**·$MeNO_2$ and 3.001(4) and 2.38(7) Å in molecule **3i**·MeCN. The angle at the

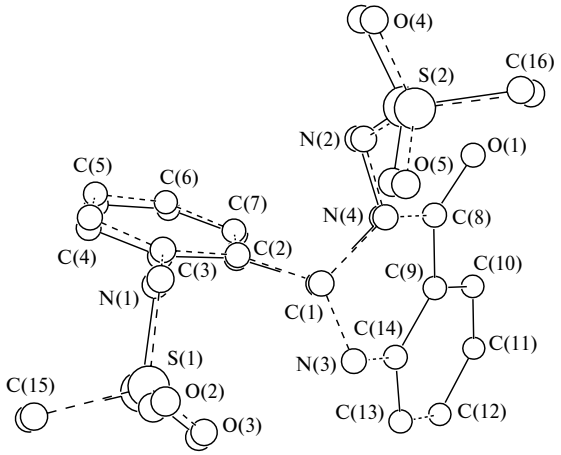


Fig. 2. Superposition of the planes of basic molecules **3i** passing through the atoms of the benzene ring C(9)—C(14) and the nearest substituents, C(8) and N(3), in crystal solvates **3i** · $MeNO_2$ and **3i** · MeCN.

hydrogen atom (134(3)° and 135(4)° in **3i** · $MeNO_2$ and **3i** · MeCN, respectively) most likely characterizes a forced contact, which efficiently shields the proton, thus hindering its involvement in hydrogen bonding with the acetonitrile solvate molecule in crystal solvate **3i** · MeCN.

Therefore, the ribbon packing of the basic molecules, which we revealed in the crystals belonging to different space groups and containing solvate molecules of different nature, is the prevailing motif in the supramolecular architecture of the compound under study.

Experimental

The IR spectra were recorded on a Specord IR-75 instrument in Nujol mulls. The ^{1}H NMR spectra of all compounds, except for **3f**, were measured on a Varian UNITY-300 spectrometer. The spectrum of **3f** was recorded on a Bruker DRX500 spectrometer. The spectroscopic characteristics of the resulting compounds are given in Tables 2 and 3. The mass spectra were obtained on a Finnigan mar. INCOS 50 GLC-mass spectrometer using a direct inlet system. Attempts to prepare anthranilic acid mesyl hydrazide according to a standard procedure[9] with the use of mesyl chloride and pyridine failed. Only a bismesylated product was isolated from the reaction mixture when the reagents were taken in a ratio of 1 : 1.

Anthranilic acid mesyl hydrazide (1). Mesyl chloride (3 mL, 0.04 mol) was added with stirring to a suspension of anthranilic acid hydrazide (6 g, 0.04 mol) in MeCN (40 mL), which was distilled over P_2O_5, and the reaction mixture was heated to boiling. The mixture rapidly became viscous and then again became liquid after 5—10 min. The reaction mixture was refluxed for 20 min and filtered in the hot state. The filtrate was cooled on ice. The crystalline precipitate that formed was filtered off, washed with cold MeCN, and dried. A colorless compound with m.p. of 174—177 °C was obtained. The yield was 2.6 g (29%). The filtrate was concentrated and an additional amount of compound **1** (1.2 g) was obtained. The total yield was 3.8 g (42%). Found (%): C, 41.65; H, 5.11; N, 18.20. $C_8H_{11}N_3O_3S$. Calculated (%): C, 41.91; H, 4.84; N, 18.33.

3-Methylsulfonylamino-2-(3-nitrophenyl)-1,2,3,4-tetrahydroquinazolin-4-one (3a). A mixture of hydrazide **1** (0.23 g, 1 mmol) and *m*-nitrobenzaldehyde (0.15 g, 1 mmol) was refluxed in PrOH (5 mL) for 1 h, cooled using rubbing with a rod, and kept on ice. The precipitate that formed was filtered off, washed with cold PrOH and petroleum ether, and dried. Yellowish crystals of compound **3a** with m.p. of 207—210 °C were obtained. The yield was 0.3 g (83%). M^+ 362. Found (%): C, 49.79; H, 4.12; N, 15.11. $C_{15}H_{14}N_4O_5S$. Calculated (%): C, 49.72; H, 3.89; N, 15.46.

3-Methylsulfonylamino-2-(4-nitrophenyl)-1,2,3,4-tetrahydroquinazolin-4-one (3b) was prepared analogously to **3a** from hydrazide **1** (0.46 g, 2 mmol) and *p*-nitrobenzaldehyde (0.33 g, 2 mmol) in Pr^{i}OH (10 mL). Compound **3b** was recrystallized

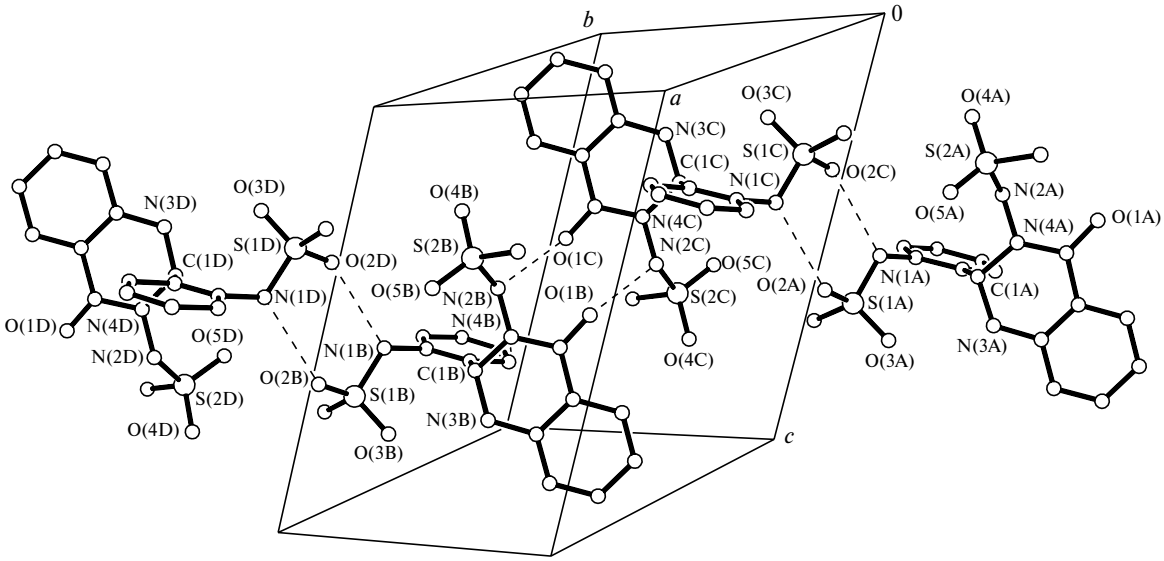


Fig. 3. Fragment of the crystal packing of **3i** · $MeNO_2$. The hydrogen atoms are omitted.

Table 2. Spectroscopic characteristics of compounds **1**, **3a—m**, and **4**

Compound	IR, ν/cm^{-1}	^{1}H NMR (DMCO-d$_6$), δ (*J*/Hz)
1	3507, 3400, 3300, 3220 (NH); 1647 (CO); 1621, 1594, 1521 (arom.); 1340, 1154 (SO_2)	2.95 (s, 3 H, Me); 6.37 (s, 2 H, NH_2); 6.47 (m, 1 H, C(4)H); 6.69 (d, 1 H, C(3)H, *J* = 8.2); 7.12 (m, 1 H, C(5)H); 7.55 (d, 1 H, C(6)H, *J* = 7.9); 9.25, 10.35 (both s, 1 H each, NH)
3a	3360, 3147 (NH); 1661 (CO); 1614, 1581, 1500, 1487 (arom.); 1527, 1341 (NO_2); 1354, 1167 (SO_2)	2.98 (s, 3 H, Me); 6.11 (d, 1 H, C(2)H, *J* = 3.2); 6.69 (m, 2 H, CH_{arom}); 7.23 (m, 1 H, CH_{arom}); 7.50—7.80 (m, 4 H, NH, CH_{arom}); 8.13 (br.dd, 1 H, C(5)H); 8.25 (s, 1 H, C(2´)H); 10.33 (s, 1 H, NH)
3b	3407, 3153 (NH); 1660 (CO); 1607, 1500 (arom.); 1521, 1341 (NO_2); 1354, 1160 (SO_2)	2.98 (s, 3 H, Me); 6.10 (d, 1 H, C(2)H, *J* = 3.2); 6.68 (m, 2 H, CH_{arom}); 7.23 (m, 1 H, CH_{arom}); 7.50—7.80 (m, 4 H, NH, CH_{arom}); 8.15 (d, 2 H, C(3´)H, C(5´)H, *J* = 8.6); 10.28 (s, 1 H, NH)
3c*	3540, 3380, 3167 (OH, NH); 1700, 1661 (CO); 1614, 1607, 1514 (arom.); 1507, 1335 (NO_2); 1340, 1167 (SO_2)	0.86 (t, 3 H, Me, *J* = 7.3); 1.42 (m, 2 H, CH_2); 2.99 (s, 3 H, Me); 3.34 (m 2 H, CH_2); 4.02 (t, 1 H, OH, *J* = 5.2); 6.22 (d, 1 H, C(2)H, *J* = 2.8); 6.82 (m, 2 H, CH_{arom}); 7.01 (d, 1 H, CH_{arom}, *J* = 9.0); 7.20 (m, 2 H, NH, CH_{arom}); 7.75 (dd, 1 H, CH_{arom}, ^3J = 7.8, ^4J = 1.25); 7.84 (s, 1 H, C(6´)H); 8.00 (dd, 1 H, C(5)H, ^3J = 9.0, ^4J = 2.8); 10.36 (s, 1 H, NH); 11.54 (s, 1 H, OH)
3d	3340, 3326, 3127 (NH); 1654 (CO); 1614, 1500, 1480 (arom.); 1334, 1327, 1160 (SO_2)	2.93 (s, 3 H, Me); 5.86 (d, 1 H, C(2)H, *J* = 3.1); 6.28 (m, 2 H, C(3´)H, C(4´)H); 6.69 (m, 2 H, C(7)H, C(8)H); 7.23 (m, 1 H, C(6)H); 7.44 (br.d, 1 H, C(5´)H); 7.55 (d, 1 H, NH, *J* = 3.1); 7.68 (d, 1 H, C(5)H, *J* = 7.8); 10.02 (s, 1 H, NH)
3e	3366, 3133, 3100 (NH); 1660, 1647 sh. (CO); 1614, 1500, 1487 (arom.); 1334, 1154 (SO_2)	2.94 (s, 3 H, Me); 6.12 (d, 1 H, C(2)H, *J* = 3.2); 6.65—6.80 (m, 2 H, CH_{arom}); 6.90 (m, 1 H, CH_{arom}); 7.07 (d, 1 H, C(3´)H, *J* = 3.3); 7.27 (m, 2 H, CH_{arom}); 7.63 (d, 1 H, NH, *J* = 3.2); 7.68 (d, 1 H, C(5)H, *J* = 7.8); 10.09 (s, 1 H, NH)
3f**	3393, 3387, 3233 (NH); 1661 (CO); 1614, 1567 (arom.); 1321, 1154 (SO_2)	2.96 (s, 3 H, Me); 6.22 (br.d, C(2)H); 6.70 (m, 2 H, CH_{arom}); 6.97 (m, 1 H, CH_{arom}); 7.17 (m, 1 H, CH_{arom}); 7.30 (br.d, CH_{arom}); 7.56 (m, 1 H, CH_{arom}); 7.69 (d, 1 H, CH_{arom}, *J* = 8.0);7.70 (br.d, 1 H, NH); 7.75 (d, 1 H, CH_{arom}, *J* = 8.00); 7.93 (d, 1 H, CH_{arom}, *J* = 8.0); 10.05 (s, 1 H, NH); 11.05 (s, 1 H, NH)
3g	3460, 3400, 3347, 3213, 3167 (OH, NH); 1661, 1647 (CO); 1621, 1614, 1601, 1501 (arom.); 1154, 1327, 1167, 1354 (SO_2)	2.95 (s, 3 H, Me); 6.19 (d, 1 H, C(2)H, *J* = 2.1); 6.65 (m, 3 H, CH_{arom}); 6.84 (d, 1 H, C(8)H, *J* = 8.0); 6.90 (d, 1 H, CH_{arom}, *J* = 7.4); 6.94 (br.d, 1 H, NH); 7.05 (m, 1 H, C(7)H); 7.15 (m, 1 H, C(6)H); 7.68 (d, 1 H, C(5)H, *J* = 7.7); 9.83 (s, 1 H, OH); 10.04 (s, 1 H, NH)
3h	3500 (OH); 3414, 3113 (NH); 1640 (CO); 1607, 1507, 1500 (arom.); 1327, 1141 (SO_2)	2.93 (s, 3 H, Me); 3.74 (s, 3 H, OMe); 5.84 (d, 1 H, CH, *J* = 2.0); 6.50—6.80 (m, 4 H, CH_{arom}); 6.88 (s, 1 H, C(2´)H); 7.18 (m, 1 H, CH_{arom}); 7.37 (br.d, 1 H, NH); 7.66 (d, 1 H, C(5)H); 8.71 (s, 1 H, OH); 9.95 (s, 1 H, NH)
3i	3380, 3293, 3140 (NH); 1714, 1641 (CO); 1614, 1467 (arom.); 1341, 1321, 1167, 1154 (SO_2)	2.97 (s, 3 H, Me); 2.98 (s, 3 H, Me); 6.38 (d, 1 H, CH, *J* = 2.6); 6.60 (d, 1 H, C(8)H, *J* = 8.0); 6.71 (m, 1 H, C(7)H); 6.82 (d br, 1 H, NH); 7.05—7.40 (m, 4 H, CH_{arom}); 7.45 (d, 1 H, C(3´)H, *J* = 7.85); 7.75 (d, 1 H, C(5)H, *J* = 7.75); 9.30, 9.98 (both s, 1 H each, NH)
3j	3380, 3220 (NH); 1674 (CO); 1621, 1607, 1527, 1487 (arom.); 1334, 1161 (SO_2)	2.99 (s, 3 H, Me); 6.12 (d, 1 H, CH, *J* = 3.4); 6.72 (m, 2 H, CH_{arom}); 7.00—7.60 (m, 6 H, NH, 5 CH_{arom}); 7.75 (dd, 1 H, C(5)H, ^3J = 7.8, ^4J = 1.25); 9.85, 12.27 (both s, 1 H each, NH)
3k	3366, 3193 (NH); 1687 (CO); 1607, 1594, 1560, 1494 (arom.); 1334, 1321, 1167, 1154 (SO_2)	3.01 (s, 3 H, Me); 6.35 (d, 1 H, C(2)H, *J* = 3.8); 6.70 (m, 2 H, CH_{arom}); 7.20, 7.57 (both m, 1 H each, CH_{arom}); 7.69 (d, 1 H, NH, *J* = 3.8); 7.75, 7.88 (both m, 2 H each, CH_{arom}); 8.00 (s, 1 H, CH_{arom}); 10.25 (s, 1 H, NH)
3l	3387, 3300 (NH); 3113 (≡CH); 3127 (C≡C); 1647 sh, 1634 (CO); 1614, 1500, 1487, 1474 (arom.); 1354, 1327, 1170, 1160 (SO_2)	2.96 (s, 3 H, Me); 3.30 (br.t, 1 H, ≡CH); 4.84, 4.86 (both d, 2 H, CH_2, *J* = 2.2); 6.15 (d, 1 H, C(2)H, *J* = 2.9); 6.66 (m, 2 H, CH_{arom}); 6.97 (br.d, 1 H, NH); 7.10—7.25 (m, 4 H, CH_{arom}); 7.71 (d, 1 H, CH_{arom}, *J* = 7.9); 10.30 (s, 1 H, NH)

(*to be continued*)

Table 2 (*continued*)

Compound	IR, ν/cm^{-1}	^{1}H NMR (DMCO-d$_6$), δ (*J*/Hz)
3m	3333, 3306, 3067 (NH); 3287 (≡CH); 2127 (C≡C); 1660 (CO); 1621, 1614, 1534, 1514, 1487, 1480, 1474 (arom.); 1334, 1154, 1147 (SO_2)	2.94 (s, 3 H, Me); 3.16 (t, 1 H, ≡CH, J = 2.4); 3.73 (s, 3 H, OMe); 4.66 (d, 2 H, CH_2, J = 2.4); 5.88 (d, 1 H, C(2)H, J = 2.1); 6.65 (m, 2 H, CH_{arom}); 6.80 (dd, 1 H, CH_{arom}, ^3J = 8.3, ^4J = 1.8); 6.89 (d, 1 H, CH_{arom}, J = 8.1); 6.95 (br.d, 1 H, NH); 7.20 (m, 1 H, CH_{arom}); 7.45 (c, 1 H, C(2′)H); 7.66 (dd, 1 H, C(5)H, ^3J = 7.8, ^4J = 1.2); 10.07 (s, 1 H, NH)
4	3033 (NH); 1700 (CO); 1614, 1567, 1480 (arom.); 1347, 1165 (SO_2)	3.14 (s, 3 H, Me); 7.56 (m, 1 H, C(7)H); 7.70 (d, 1 H, C(8)H, J = 7.9); 7.84 (m, 1 H, C(6)H); 8.15 (s, 1 H, C(2)H); 8.21 (dd, 1 H, C(5)H, ^3J = 8.0, ^4J = 1.25); 11.33 (s, 1 H, NH)

* The 1 : 1 solvate with PrOH.
** The spectrum was recorded on a Bruker DRX 500 spectrometer.

Table 3. Spectroscopic characteristics of compounds **6—16**

Compound	IR, ν/cm^{-1}	^{1}H NMR (DMCO-d$_6$), δ (*J*/Hz)
6	3340 (NH); 1634 (CO); 1607 (CH=N); 1500, 1487 (arom.)	6.18 (d, 1 H, C(3′)H, J = 3.3); 6.24 (d, 1 H, CH, J = 3.4); 6.26, 6.53 (both m, 1 H each, C(4′)H); 6.72 (m, 1 H, C(7)H); 6.78 (d, 1 H, C(8)H, J = 8.0); 6.84 (d, 1 H, C(3′)H, J = 3.3); 7.24 (m, 1 H, C(6)H); 7.44 (d, 1 H, C(5′)H, J = 1.7); 7.63 (d, 1 H, NH, J = 3.4); 7.68 (d, 1 H, C(5′)H, J = 1.7); 7.71 (dd, C(5)H, ^3J = 7.9, ^4J = 1.4)
7	3287, 3153 (NH); 1687 (CO); 1634, 1607, 1594, 1567 (arom.)	5.73 (s, 2 H, NH_2); 7.50 (m, 1 H, C(7)H); 7.63 (d, 1 H, C(8)H, J = 7.9); 7.74 (m, 1 H, C(6)H); 8.17 (dd, 1 H, C(5)H, ^3J = 8.0, ^4J = 1.0); 8.23 (s, 1 H, C(2)H)
8	3380, 3153 (NH); 1660 (CO); 1620, 1520, 1487 (arom.); 1340, 1160 (SO_2)	1.40—2.00 (m, 7 H, CH_2); 2.66 (m, 1 H, CH_2); 2.91 (s, 3 H, Me); 6.65 (m, 1 H, C(7)H); 6.73 (d, 1 H, C(8)H, J = 8.1); 7.06 (s, 1 H, NH); 7.23 (m, 1 H, C(6)H); 7.66 (dd, 1 H, C(5)H, ^3J = 7.8, ^4J = 1.3); 9.44 (s, 1 H, NH)
9	3387, 3193, 3167 (NH); 1660 (CO); 1614, 1514, 1487 (arom.); 1330, 1160 (SO_2)	1.00—2.20 (m, 10 H, CH_2); 2.90 (s, 3 H, Me); 6.65 (m, 1 H, C(7)H); 6.79 (s, 1 H, NH); 6.90 (d, 1 H, C(8)H, J = 8.0); 7.24 (m, 1 H, C(6)H); 7.65 (dd, 1 H, C(5)H, ^3J = 7.9, ^4J = 1.3); 9.38 (s, 1 H, NH)
10	3267 (NH); 1640, 1634 (CO, C=N); 1614, 1580, 1514, 1487 (arom.)	1.50—2.60 (m, 16 H, CH_2); 6.61 (m, 1 H, C(7)H); 6.67 (d, 1 H, C(8)H, J = 8.0); 6.75 (s, 1 H, NH); 7.13 (m, 1 H, C(6)H); 7.59 (d, 1 H, C(5)H, J = 7.0)
11	3320 (NH); 1640 sh, 1620 (CO, C=N); 1607, 1580, 1514, 1487 (arom.)	1.00—2.50 (m, 20 H, CH_2); 6.51 (s, 1 H, NH); 6.61 (m, 1 H, C(7)H); 6.87 (d, 1 H, C(8)H, J = 8.0); 7.16 (m, 1 H, C(6)H); 7.57 (dd, 1 H, C(5)H, ^3J = 7.8, ^4J = 1.4)
12	3167 (NH); 1660 (CO); 1607, 1600, 1567, 1475 (arom.); 1354, 1167 (SO_2)	2.61, 3.18 (both s, 3 H each, Me); 7.47 (m, 1 H, C(7)H); 7.60 (d, 1 H, C(8)H, J = 8.0); 7.77 (m, 1 H, C(6)H); 8.12 (dd, 1 H, C(5)H, ^3J = 7.9, ^4J = 1.1); 10.95 (s, 1 H, NH)
13a	3313, 3260 (OH, NH); 1707, 1687, 1634 (CO); 1607 (arom.)	2.40 (m, 4 H, CH_2); 5.90 (d, 1 H, C(2)H, J = 2.0); 6.35 (m, 2 H, C(3′)H, C(4′)H); 6.72 (m, 2 H, CH_{arom}); 7.23 (m, 1 H, CH_{arom}); 7.29 (d, 1 H, C(5′)H, J = 1.7); 7.51 (br.d, 1 H, NH); 7.65 (d, C(5)H, J = 7.0); 10.08 (s, 1 H, NH); 11.92 (s, 1 H, OH)
13b	3319, 3260, 3033 (OH, NH); 1734, 1714, 1667 (CO); 1614, 1540, 1500, 1487 (arom.)	2.30 (m, 4 H, CH_2); 6.00 (br.d, 1 H, C(2)H); 6.70 (m, 2 H, CH_{arom}); 7.10 (m, 3 H, CH_{arom}); 7.23 (m, 1 H, CH_{arom}); 7.49 (m, 2 H, NH, CH_{arom}); 7.66 (br.dd, C(5)H); 9.77 (s, 1 H, NH); 11.90 (s, 1 H, OH)
14	3320, 3193, 3020 (OH, NH); 1700, 1687, 1640 (CO); 1614, 1527, 1507, 1487 (arom.)	5.98 (d, 1 H, C(2)H, J = 2.6); 6.20—6.40 (m, 4 H, C(3′)H, C(4′)H, CH_{olef}); 6.72 (m, 2 H, CH_{arom}); 7.25 (m, 1 H, CH_{arom}); 7.44 (d, 1 H, NH, J = 2.6); 7.50 (m, 1 H, C(5′)H); 7.68 (dd, 1 H, C(5)H, ^3J = 7.8, ^4J = 1.5); 10.87 (s, 1 H, NH); 13.33 (s, 1 H, OH).
15	3326, 3253 (OH, NH); 1707, 1674 (CO); 1634, 1620, 1607, 1580 (arom.)	1.40—2.30 (m, 8 H, CH_2); 2.45 (m, 4 H, CH_2); 6.88 (m, 2 H, C(7)H, C(8)H); 6.85 (s, 1 H, NH); 7.19 (m, 1 H, C(6)H); 7.60 (dd, 1 H, C(5)H, ^3J = 7.8, ^4J = 1.2); 9.72 (s, 1 H, NH); 11.91 (s, 1 H, OH)
16	3333 (NH); 1714, 1660 (CO); 1607, 1567, 1520, 1480 (arom.)	2.40 (s, 3 H, Me); 2.58 (m, 4 H, CH_2); 7.44 (m, 1 H, C(7)H); 7.57 (d, 1 H, C(8)H, J = 8.1); 7.72 (m, 1 H, C(6)H); 8.11 (d, 1 H, C(5)H, J = 7.9); 10.97 (s, 1 H, NH); 12.03 (s, 1 H, OH)

from EtOH (75 mL). Lemon-yellow crystals with m.p. of 241—249 °C were obtained. The yield was 0.55 g (75%). Found (%): C, 49.83; H, 4.16; N, 15.32. $C_{15}H_{14}N_4O_5S$. Calculated (%): C, 49.72; H, 3.89; N, 15.46.

2-(2-Hydroxy-5-nitrophenyl)-3-methylsulfonylamino-1,2,3,4-tetrahydroquinazolin-4-one (3c) was prepared analogously to **3a**. The reaction time was 15 min. Yellowish crystals containing a PrOH solvate molecule were obtained, the solvate molecule being gradually eliminated on heating. M.p. 225—240 °C. The yield was 0.33 g (75%). M^+ 378.

2-(2-Furyl)-3-methylsulfonylamino-1,2,3,4-tetrahydroquinazolin-4-one (3d) was prepared analogously to **3a** with the use of furfural (0.2 mL, a 100% excess). Colorless crystals with m.p. of 215—220 °C were obtained. The yield was 0.25 g (81%). Found (%): C, 51.13; H, 4.37; N, 13.31. $C_{13}H_{13}N_3O_4S$. Calculated (%): C, 50.81; H, 4.26; N, 13.67.

3-Methylsulfonylamino-2-(2-thienyl)-1,2,3,4-tetrahydroquinazolin-4-one (3e) was prepared analogously to **3a** from hydrazide **1** (0.46 g, 2 mmol) and 2-thiophenecarbaldehyde (0.3 mL, 3.2 mmol) in Pr^iOH (10 mL). Compound **3e** was recrystallized from EtOH (20 mL). Colorless crystals with m.p. of 230—234 °C were obtained. The yield was 0.45 g (69%). Found (%): C, 47.92; H, 4.37; N, 12.63. $C_{13}H_{13}N_3O_3S_2$. Calculated (%): C, 48.28; H, 4.05; N, 12.99.

2-(3-Indolyl)-3-methylsulfonylamino-1,2,3,4-tetrahydroquinazolin-4-one (3f) was prepared analogously to **3a**. The reaction time was 35 min. Colorless crystals with m.p. of 225—228 °C were obtained. The yield was 0.2 g (56%). The filtrate was partially concentrated and an additional amount (0.07 g) of compound **3f** was obtained. The total yield was 0.27 g (76%). Found (%): C, 57.35; H, 4.72; N, 15.54. $C_{17}H_{16}N_4O_3S$. Calculated (%): C, 57.29; H, 4.53; N, 15.72.

2-(2-Hydroxyphenyl)-3-methylsulfonylamino-1,2,3,4-tetrahydroquinazolin-4-one (3g) was prepared analogously to **3a** from hydrazide **1** (1.3 g) and salicylaldehyde (0.7 mL, 0.1 mL excess) in PrOH (5 mL). After cooling on ice, Pr^iOH (3 mL) was added. Colorless crystals with m.p. of 237—240 °C were obtained. The yield was 1.5 g (79%). A compound with m.p. of 232—239 °C was precipitated from the filtrate with petroleum ether in a yield of 0.19 g. The IR spectrum of the latter compound was identical with that of **3g**. Found (%): C, 53.82; H, 4.79; N, 12.83. $C_{15}H_{15}N_3O_4S$. Calculated (%): C, 54.05; H, 4.53; N, 12.60.

2-(4-Hydroxy-3-methoxyphenyl)-3-methylsulfonylamino-1,2,3,4-tetrahydroquinazolin-4-one (3h) was prepared analogously to **3a** by the reaction with vanillin. The reaction mixture was cooled to room temperature, water (20 mL) was added, and the mixture was cooled on ice. The precipitate was filtered off, washed with water, and dried. A colorless compound with m.p. of 232—238 °C was obtained. The yield was 0.28 g (77%). M^+ 363. Found (%): C, 52.50; H, 5.11; N, 11.34. $C_{16}H_{17}N_3O_5S$. Calculated (%): C, 52.88; H, 4.72; N, 11.56.

3-Methylsulfonylamino-2-(2-methylsulfonylaminophenyl)-1,2,3,4-tetrahydroquinazolin-4-one (3i) was prepared analogously to **3a** by the reaction with *o*-mesylaminobenzaldehyde.[9] A colorless compound with m.p. of 157 °C was obtained. The yield was 0.26 g (63%). The structure was established by X-ray diffraction.

2-(2-Benzimidazolyl)-3-methylsulfonylamino-1,2,3,4-tetrahydroquinazolin-4-one (3j) was prepared analogously to **3a** in PrOH (10 mL). The reaction time was 2 h. The reaction mixture was filtered off from a small insoluble precipitate and cooled. Then H_2O (30 mL) was added and the mixture was kept on ice for 1 h. The precipitate was filtered off and dried *in vacuo*. A colorless compound with m.p. of 195—200 °C was obtained. The yield was 0.2 g (56%). Found (%): C, 53.34; H, 4.32; N, 19.30. $C_{16}H_{15}N_5O_3S$. Calculated (%): C, 53.77; H, 4.23; N, 19.60.

2-[2-(Chloroquinolin-3-yl)]-3-methylsulfonylamino-1,2,3,4-tetrahydroquinazolin-4-one (3k). A mixture of hydrazide **1** (0.46 g, 2 mmol) and 2-chloro-3-formylquinoline[10] (0.38 g, 2 mmol) was refluxed in PrOH (5 mL) for 15 min. The new crystalline precipitate that formed after dissolution of the starting compounds was filtered off from the hot solution, washed with PrOH and petroleum ether, and dried. A colorless compound, which decomposes upon heating above 250 °C, was obtained. The yield was 0.5 g (62%). Found (%): C, 53.55; H, 3.91; N, 14.11. $C_{18}H_{15}N_4ClO_3S$. Calculated (%): C, 53.67; H, 3.75; N, 13.91.

2-(5-Chloro-2-propargyloxyphenyl)-3-methylsulfonylamino-1,2,3,4-tetrahydroquinazolin-4-one (3l). Propargyl ether of 5-chlorosalicylaldehyde[11] (0.39 g, 2 mmol) was added to a boiling solution of hydrazide **1** (0.46 g, 2 mmol) in PrOH (10 mL). The mixture was refluxed for 30 min. The precipitate that formed was filtered off in the hot state and washed with Pr^iOH and petroleum ether. Colorless crystals with m.p. of 213—220 °C were obtained. The yield was 0.57 g (70%). Found (%): C, 52.94; H, 4.27; N, 10.41. $C_{18}H_{16}N_3ClO_4S$. Calculated (%): C, 53.27; H, 3.97; N, 10.35.

2-(3-Methoxy-4-propargyloxyphenyl)-3-methylsulfonylamino-1,2,3,4-tetrahydroquinazolin-4-one (3m). A mixture of hydrazide **1** (0.46 g, 2 mmol) and propargyl ether of vanillin[11] (0.38 g, 2 mmol) was refluxed in PrOH (10 mL) for 30 min and then cooled to room temperature by gradually adding water with stirring and rubbing with a rod. Then the mixture was kept on ice for 3 h. The precipitate that formed was filtered off and recrystallized from toluene with cooling to room temperature and rubbing with a rod. A colorless compound with m.p. of 141—142 °C was obtained. The yield was 0.43 g (54%). Found (%): C, 56.63; H, 5.11; N, 10.24. $C_{19}H_{19}N_3O_5S$. Calculated (%): C, 56.85; H, 4.77; N, 10.47.

3-Methylsulfonylamino-3,4-dihydroquinazolin-4-one (4). Hydrazide **1** (0.46 g, 2 mmol) was refluxed in HCOOH (2 mL) for 1 h and then cooled. An oil that was precipitated with water crystallized out by rubbing with a rod. The precipitate was filtered off and recrystallized from MeOH. A colorless compound with m.p. of 222—228 °C was obtained. The yield was 0.42 g (87%). M^+ 239. Found (%): C, 45.23; H, 4.13; N, 17.32. $C_9H_9N_3O_3S$. Calculated (%): C, 45.18; H, 3.79; N, 17.56.

2-(2-Furyl)-3-(2-furfurylideneamino)-1,2,3,4-tetrahydroquinazolin-4-one (6). A mixture of hydrazide **5** (0.75 g, 5 mmol) and furfural (1 mL, 12 mmol) was refluxed in PrOH (5 mL) for 1 h and cooled on ice using rubbing with a rod. The precipitate that formed was filtered off, washed with cold Pr^iOH and petroleum ether, and dried. Cream-color crystals with m.p. of 127—130 °C (from Pr^iOH) were obtained. The yield was 1.27 g (83%). Found (%): C, 66.72; H, 4.34; N, 13.41. $C_{17}H_{13}N_3O_3$. Calculated (%): C, 66.44; H, 4.26; N, 13.67.

3-Amino-3,4-dihydroquinazolin-4-one (7). Hydrazide **5** (1 g, 6.6 mmol) was refluxed in HCOOH (2 mL) for 1 h and cooled. Then H_2O (10 mL) and concentrated NH_4OH (6 mL) were added. The precipitate that formed was filtered off and recrystallied from MeOH. A colorless compound with m.p. of

212—214 °C was obtained. The yield was 0.3 g (28%). Found (%): C, 63.21; H, 5.41; N, 31.72. $C_7H_7N_3O$. Calculated (%): C, 63.14; H, 5.30; N, 31.56.

3-Methylsulfonylamino-2-spirocyclopentane-1,2,3,4-tetrahydroquinazolin-4-one (8). A mixture of hydrazide **1** (0.46 g, 2 mmol) and cyclopentanone (1 mL) in ethylene glycol (3 mL) was refluxed for 1.5 h and cooled. Then Pr^iOH (5 mL) was added and the mixture was kept on ice for 1 h. The precipitate that formed was filtered off and washed with cold Pr^iOH and petroleum ether. A colorless compound with m.p. of 283—286 °C was obtained. The yield was 0.45 g (76%). Found (%): C, 52.69; H, 5.98; N, 14.00. $C_{13}H_{17}N_3O_3S$. Calculated (%): C, 52.87; H, 5.80; N, 14.23.

3-Methylsulfonylamino-2-spirocyclohexane-1,2,3,4-tetrahydroquinazolin-4-one (9). A mixture of hydrazide **1** (0.23 g, 1 mmol) and cyclohexanone (1 mL) in glycol (3 mL) was refluxed for 2 h and then cooled. The precipitate that formed was filtered off and washed with EtOH. A colorless compound with m.p. of 282—284 °C was obtained. The yield was 0.21 g (68%). Found (%): C, 54.11; H, 6.51; N, 13.21. $C_{14}H_{19}N_3O_3S$. Calculated (%): C, 54.35; H, 6.19; N, 13.58.

2-Spirocyclopentane-3-cyclopentylideneamino-1,2,3,4-tetrahydroquinazolin-4-one (10). Hydrazide **5** (0.5 g, 3.3 mmol) was refluxed in cyclopentanone (3 mL) for 1 h, the reaction mixture was cooled, and petroleum ether (10 mL) was added to the precipitate that formed. Then the precipitate was filtered off and washed with petroleum ether. A colorless compound with m.p. of 155—166 °C (from isooctane) was obtained. The yield was 0.9 g (96%). Found (%): C, 72.60; H, 7.68; N, 14.52. $C_{17}H_{21}N_3O$. Calculated (%): C, 72.31; H, 7.50; N, 14.88.

2-Spirocyclohexane-3-cyclohexylideneamino-1,2,3,4-tetrahydroquinazolin-4-one (11) was prepared analogously to compound **10** in cyclohexanone (3 mL). A colorless compound with m.p. of 228—230 °C was obtained. The yield was 0.85 g (82.5%). Found (%): C, 73.16; H, 8.24; N, 13.51. $C_{19}H_{25}N_3O$. Calculated (%): C, 73.28; H, 8.09; N, 13.49.

2-Methyl-3-methylsulfonylamino-3,4-dihydroquinazolin-4-one (12). Hydrazide **1** (1 g, 4.3 mmol) was refluxed in acetylacetone (4 mL) for 30 min, Pr^iOH (4 mL) was added, and the mixture was cooled on ice using rubbing with a rod. The precipitate that formed was filtered off and washed with Pr^iOH and petroleum ether. A colorless compound with m.p. of 195—197 °C (from Pr^iOH) was obtained. The yield was 0.7 g (63%). M^+ 253. Found (%): C, 47.20; H, 4.53; N, 16.41. $C_{10}H_{11}N_3O_3S$. Calculated (%): C, 47.42; H, 4.38; N, 16.59.

Synthesis of compounds 13 and 14 (general procedure). Hydrazide **5** (0.75 g, 5 mmol) and succinic (in the case of **13a,b**) or maleic (in the case of **14**) anhydride (0.5 g, 5 mmol) were triturated in a mortar and placed in a round-bottom flask. Then MeCN (5 mL), which was distilled over P_2O_5 before use, was added, and the reaction mixture was refluxed for 5 min. Furfural (in the case of **13a** and **14**) or *p*-fluorobenzaldehyde (in the case of **13b**) (0.6 mL) was added. The reaction mixture was refluxed for 30 min (in the case of **13a,b**) or 15 min (in the case of **14**) and cooled on ice. The precipitate that formed was filtered off, washed with cold MeCN and petroleum ether, and dried.

***N*-[2-(2-Furyl)-4-oxo-1,2,3,4-tetrahydroquinazolin-3-yl]succinamic acid (13a).** A colorless compound with m.p. of 154—156 °C was obtained. The yield was 1 g (61%). M^+ 329. Found (%): C, 58.11; H, 4.78; N, 12.50. $C_{16}H_{15}N_3O_5$. Calculated (%): C, 58.36; H, 4.59; N, 12.76.

***N*-[2-(4-Fluorophenyl)-4-oxo-1,2,3,4-tetrahydroquinazolin-3-yl]succinamic acid (13b).** A colorless compound with m.p. of 155—160 °C (from MeCN) was obtained. The yield was 1 g (56%). Found (%): C, 60.31; H, 4.62; N, 11.54. $C_{18}H_{16}N_3O_4F$. Calculated, (%): C, 60.50; H, 4.51; N, 11.76.

***N*-[2-(2-Furyl)-4-oxo-1,2,3,4-tetrahydroquinazolin-3-yl]maleinamic acid (14).** A colorless compound with m.p. of 177—178 °C (from propylene carbonate) was obtained. The yield was 0.8 g (50%). M^+ 327. Found (%): C, 58.41; H, 4.35; N, 12.60. $C_{16}H_{13}N_3O_5$. Calculated (%): C, 58.72; H, 4.00; N, 12.84.

Table 4. Crystallographic parameters and details of X-ray diffraction study of crystal solvates **3i** · $MeNO_2$ and **3i** · MeCN

Parameter	**3i** · $MeNO_2$	**3i** · MeCN
Molecular formula	$C_{17}H_{21}N_5O_7S_2$	$C_{18}H_{21}N_5O_5S_2$
Molecular weight /kg kmol^{-1}	471.51	451.52
Crystal system	Monoclinic	Triclinic
Space group	$P2_1/c$	$P\bar{1}$
a/Å	9.3609(5)	9.4160(4)
b/Å	22.979(1)	9.8709(3)
c/Å	9.8990(6)	12.3807(4)
α/deg	—	72.796(2)
β/deg	96.083(2)	73.176(2)
γ/deg	—	84.917(2)
V/Å^3	2117.3(2)	1052.18(6)
Z	4	2
ρ_{calc}/g cm^{-1}	1.479	1.425
F(000)	984	472
μ(Mo-Kα)/mm^{-1}	0.302	0.294
Crystal dimensions/mm	0.28×0.12×0.10	0.18×0.08×0.02
T/K	120.0(2)	120.0(2)
Radiation (λ/Å)	Mo-K_α (0.71073)	Mo-Kα (0.71073)
Scanning mode	ω	ω
Scan range, θ/deg	1.77—28.00	1.79—29.82
Ranges of indices of measured reflections	$-12 \le h \le 12$ $-29 \le k \le 30$ $-8 \le l \le 13$	$-12 \le h \le 10$ $-13 \le k \le 11$ $-17 \le l \le 16$
Number of measured reflections	15029	7818
Number of independent reflections	5096	5279
(R_{int})	(0.0277)	(0.0221)
Number of reflections with $I > 2\sigma(I)$	4309	4163
Variables in refinement	365	344
R factors based on reflections with $I > 2\sigma(I)$		
R_1	0.0401	0.0533
wR_2	0.1008	0.1365
based on all reflections		
R_1	0.0491	0.0740
wR_2	0.1048	0.1471
Goodness-of-fit on F^2	1.055	1.005
Residual electron density (min/max)/e Å^3	−0.426/0.437	−0.518/0.370

***N*-(4-Oxo-2-spirocyclopentane-1,2,3,4-tetrahydroquinazolin-3-yl)succinamic acid (15)** was prepared analogously to **13** from hydrazide **5** (1.5 g), succinic anhydride (1 g), and cyclopentanone (2 mL) in MeCN (5 mL). A colorless compound with m.p. of 191—193 °C (from MeOH) was obtained. The yield was 2.4 g (75%). Found (%): C, 60.44; H, 6.35; N, 13.42. $C_{16}H_{19}N_3O_4$. Calculated (%): C, 60.56; H, 6.04; N, 13.24.

***N*-(2-Methyl-4-oxo-3,4-dihydroquinazolin-3-yl)succinamic acid (16)** was prepared analogously to **15** with the use of acetylacetone (2 mL). After refluxing for 1 h, the solution was cooled and kept on ice for 3 h. The precipitate that formed was filtered off, washed with cold MeCN, diethyl ether, and petroleum ether, and dried. Colorless crystals with m.p. of 203—205 °C were obtained. The yield was 0.65 g (24%). M^+ 275. Found (%): C, 56.38; H, 5.13; N, 15.10. $C_{13}H_{13}N_3O_4$. Calculated (%): C, 56.73; H, 4.76; N, 15.27.

X-ray diffraction study of crystal solvates 3i · $MeNO_2$ and 3i · MeCN. Single crystals of crystal solvates **3i** · $MeNO_2$ and **3i** · MeCN were coated with a perfluorinated oil and mounted on a Bruker SMART-CCD diffractometer under a stream of cold nitrogen. The experimental X-ray data sets were collected from single crystals (Mo-Kα radiation) using the ω-scanning technique. The crystallographic parameters and characteristics of X-ray diffraction study are given in Table 4.

The experimental reflections were processed using the Bruker SAINT software.[12]

Both structures were solved by direct methods and refined by the full-matrix least-squares method against F^2 with anisotropic displacement parameters for all nonhydrogen atoms. The positions of the hydrogen atoms, including the hydrogen atoms of the methyl groups of the nitromethane solvate molecule in **3i** · $MeNO_2$ and the acetonitrile solvate molecule in **3i** · MeCN, were revealed in difference Fourier maps and refined isotropically.

All calculations were carried out with the use of the SHELXTL-Plus program package.[13] The atomic coordinates and other experimental data were deposited with the Cambridge Crystallographic Data Center.*

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