Definitive Structural Assignment of Condensation Products from
Anthranilamide and 3-Amino-2-carbamoylthiophene with Ketones.
Formation of Tetrahydroquinazolinones and Their Thiophene Isosteres

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Products formed from dehydrative cyclization of cyclopentanone, cyclohexanone, and cycloheptanone with anthranilamide (2) under neutral or acidic conditions are established as the spiro compounds 2,2-poly-methylene-1,2,3,4-tetrahydroquinazolin-4-ones 6 by means of (a) the presence of a ^{13}C nmr signal at 66-79 ppm for atom C-2 and (b) X-ray crystallography on the tetramethylene compound 6a. Analogously, products from reactions of these cycloalkanones with 3-amino-2-carbamoylthiophene are now shown by ^{13}C nmr spectra to have structures isosteric to 6, i.e. 5,5-polymethylene-4,5,6,7-tetrahydrothieno[2,3-e]pyrimidin-7-ones 5, rather than the chelate ring structures previously proposed. Additionally, conflicting literature reports on product 3 from reaction of acetone with 2 are partially clarified. Correlation of ^{13}C chemical shifts in systems 3, 5, and 6 is presented.

In a previous publication [2] Klemm and coworkers reported the reaction of 3-amino-2-carbamoylthiophene (1) with cycloalkanones in absolute ethanol at 25-78° and pH 3.4 with the loss of a mole of water per mole of 1 reacted. On the basis of infrared, mass, and ^1H nmr spectra they proposed chelate structures 4a-4c for the crystalline products formed. In the present paper we present new experimental data that show definitively that structures 4a-4c are incorrect and should be replaced by the tetrahydrothieno-[2,3-e]pyrimidinone structures 5a-5c, respectively.

Klemm et al. [2] noted that their synthetic method "resembles literature procedures for the preparation of tetrahydroquinazolinones" 3, 6a, and 6b from anthranil-amide (2a) under more strenuous acidic reaction conditions [3,4]. However, non-chelated colorless, crystalline iminoamides 7 were reported by Russian workers [5] from refluxing N-alkylanthranilamides 2b with cyclopentanone in benzene alone. Also Moore and Kornreich [6] isolated a neutral compound $C_{13}H_{16}N_2O$ from treatment of a mixture of anthranilonitrile (8) and cyclohexanone with Triton B. They assigned iminoamide structure 9 to their product since it could be hydrolyzed to 2a and cyclohexanone and also synthesized (under unspecified conditions) from these two components. Only the Russians characterized their
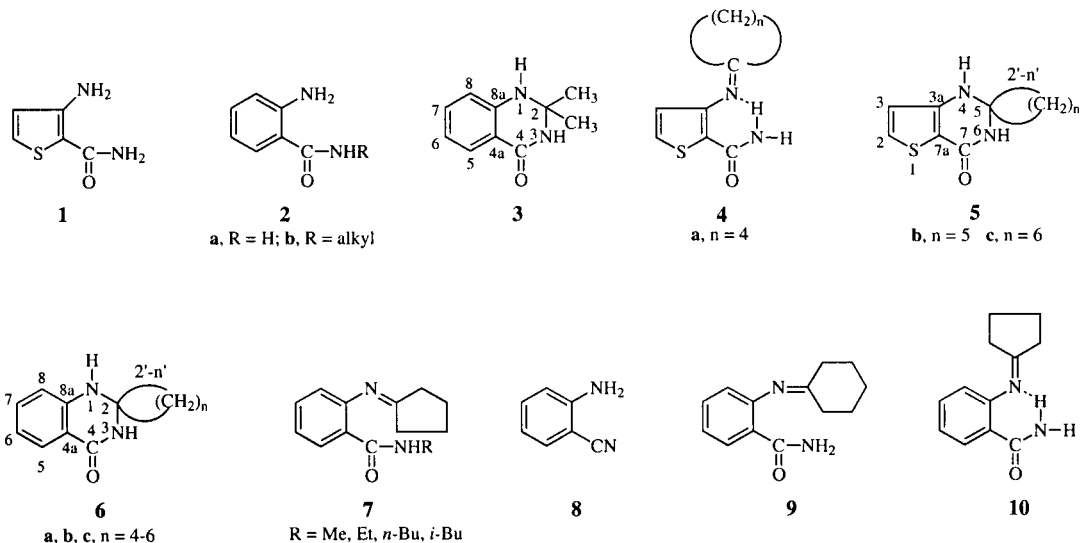

Figure 1.

Table 1

Comparison of Observed ^{13}C NMR Chemical Shifts (δ) for Isosteric Compounds of Series **5** and **6**
Plus Compound **3** in Hexadeuteriodimethyl Sulfoxide

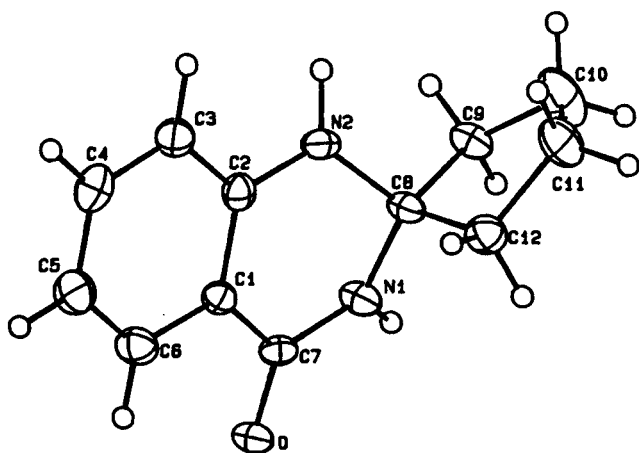| Parent ring series	Carbon position	Chemical Shift Observed			for 3 [a]	Calcd. δ [b]	Average Δδ [c]
		for series **a**	for series **b**	for series **c**			
6	4	163.5	163.2	163.0	163.1		
5	7	161.7	161.2	161.0			-1.9
6	8a	147.6	146.8	146.8	147.1	146.5	
5	3a [d]	152.1	151.3	151.2			+4.4
6	7	133.1	133.2	133.1	133.2	132.2	
5	2 [d]	132.3	132.2	132.2			-1.0
6	5	127.3	127.1	127.1	127.2	128.1	
5	3 [d]	118.0	118.2	118.1			+3.7 [e]
6	6 [f]	116.6	116.5	116.3	116.5	116.3	
6	8 [f]	114.6	114.6	114.3	114.2	113.7	
6	4a [f]	114.4	114.4	114.2	113.8	118.1	
5	7a [d]	105.8	105.1	104.8			-9.0
6	2	77.1	67.8	71.9	66.8		
5	5	79.0	70.1	74.2			+2.1 [g]
6	2',n'	39.3	37.2	41.0	29.0 [h]		
5	2',n'	38.6	36.2	39.9			-1.0 [g]
6	4',(5') [i]		24.7	29.3			
5	4',(5') [i]		24.7	29.1			-0.1
6	3',(n-1)'	22.0	20.9	20.9			
5	3',(n-1)'	21.9	21.1	20.9			0.0

[a] Structural assignments on this compound were aided by means of an Attached Proton Test [13]. [b] Based on assumed additivity effects of the two benzene ring substituents -NHR and -C(=O)NHR (where R is an alkyl group) [11]. See discussion. [c] Average chemical shift for compounds **5a-5c** minus that for compounds **6a-6c** and **3**. [d] An Attached Proton Test on **5b** serves to distinguish the 3a,7a-pair from the 2,3-pair. [e] Compare data given for position 8 in **6a-6c** and **3**. [f] Data for positions 6, 8, and 4a may be interchanged, except for compound **3** where the chemical shift of 4a was established by an Attached Proton Test. [g] This value excludes the chemical shift shown for **3**. [h] Value for methyl carbon atoms at C-2. [i] Position 5' applies only to series **c**.

products by spectral means, specifically by ultraviolet and infrared absorptions. Signals at 3394 (±2) and 3285 (±8) cm^{-1} in solvent carbon tetrachloride were assigned to NH bands, but amide bands were not reported. Based on these literature results, it appeared that our presumed chelate compounds were really intermediates which should be convertible into isosteres of tetrahydroquinazolinones by refluxing with concentrated hydrochloric acid. Attempts to effect such transformation, however, yielded only less pure recovered starting materials. Thus, proposed structures **4a-4c** were brought into question for our compounds [2].

Meanwhile, it came to our attention that Lessel [7] had reacted **2a** with cyclopentanone simply by heating the two components (sans acid) at 160° for 3 hours to give a 99% yield of **6a**. Likewise, heating **2a** with cyclohexanone for 15 minutes at 100° gave a 97% yield of **6b**. His infrared, mass, and ^{1}H nmr spectra, analogous to ours in the thiophene series, could be interpreted either in terms of chelate structures (*cf.* **4** [2]) or of tetrahydroquinazolinones. However, for **6a** he also reported a ^{13}C nmr spectrum which showed a signal at 77.0 ppm, assigned to quaternary carbon C-2, and inconsistent with any expected chemical shift to be observed for the benzene isostere of structure **4a**, *i.e.* **10**. To corroborate the **6a** and **6b** structures and to

correlate reaction conditions used in both the thiophene and benzene series we repeated the syntheses of **6a** (82% yield) and **6b** (60%) by our methodology at pH 3.4 in absolute ethanol. Also we synthesized the new homolog **6c** (37%) in the same manner [8]. Data on our samples of **6a** and **6b** compared closely with those recorded by Lessel [9]. Comparative ^{13}C nmr spectral data and proposed structural assignments for compounds **3** (*vide infra*), **5a-5c**, and **6a-6c** are presented in Table 1. In two cases, **3** and **5b**, Attached Proton Test spectra helped to delineate assignments further. From this Table one notes that each of these compounds shows a ^{13}C signal at 67-79 ppm for a quaternary aliphatic carbon atom, at C-2 in **3** and **6a-6c** or at C-5 in **5a-5c**. This chemical shift appears to be reasonable for an -NHCR$_2$NH- system, where R is an alkyl group [10].

Positional assignments of the ^{13}C nmr signals in compounds **6a-6c** and **3** (Table 1) were made with the aid of chemical shifts reported by Ewing [11] for substituents -NHR (*ipso*, + 19.0; *ortho*, -15.0; *meta*, + 0.6; *para*, -12.0) and -C(=O)NHR (*ipso*, + 5.6; *ortho*, -1.0; *meta*, -0.2; *para*, + 3.0) (where R is alkyl) on the benzene ring and the assumption of additivity of effects [12]. Thus, observed chemical shifts were readily assigned to carbon atoms 5, 7,

and 8a. Chemical shifts for carbons 6, 8, and 4a were too close together (113.8-116.6) to differentiate amongst them by this method, though the signal at 113.8 was identified as quaternary carbon C-4a in **3** by use of an Attached Proton Test [13]. Other signals in the benzene series were readily recognized by their chemical shifts [14]. The similarities in chemical shifts and an Attached Proton Test on **5b** were used to make structural assignments in the thiophene series **5** [15].

Figure 2. ORTEP drawing of compound **6a**.

Table 2

Crystallographic Data and Structural Refinement for **6a**

Empirical formula	$C_{12}H_{14}N_2O$
Formula weight	202.25
Crystal appearance	colorless lath
Crystal dimensions	0.04 x 0.15 x 0.43 mm
Crystal system	orthorhombic
Space group	Pbca
Unit cell dimensions	a = 10.320 (2) Å
	b = 11.993 (3) Å
	c = 16.749 (3) Å
	$\alpha = \beta = \gamma = 90°$
Unit cell volume	2073 (2) Å^3
Z	8
Density (calculated)	1.296 g/cm^3
Linear absorption coefficient (μ)	0.79 cm^{-1}
F(000)	864
Diffractometer	Enraf-Nonius CAD-4
Radiation wavelength (λ)	MoKα, 0.71073 Å
Monochromator	graphite
Temperature	21°C
Maximum 2θ	50°
Index ranges	h, 0→12; k, 0→14; l, 0→19
Scan mode	$\omega/2\theta$
Scan speed (on ω)	0.96° min^{-1}
Scan width	(0.80 + 0.35tan θ)°
Standard reflections	3 for every 3600 s exposure
Independent reflections scanned	1822 (excluding systematic absences)
R$_{int}$	no equivalents measured
Reflections in refinement (N)	1141 [I ≥ 1σ (I)]
Absorption correction	none
Secondary extinction parameter (g)	1.9 (2) x 10^{-6}
Number of parameters (V)	137
Weighting factor (w)	$1/\sigma^2(F)$
R(F), wR(F)	0.097, 0.054
Function minimized	$\Sigma w(|F_0| - |F_c|)^2$
S	1.57
Maximum Δ/σ, last cycle	0.006
Maximum, minimum in final diffraction map	0.42, -0.38e Å^{-3}

Algebraic relationships:
$R(F) = \Sigma ||F_0| - |F_c||/\Sigma |F_0|$　　$S = [\Sigma w(|F_0| - |F_c|)^2 /(N - V)]^{1/2}$
$wR(F) = [\Sigma w(|F_0| - |F_c|)^2/\Sigma w|F_0|^2]^{1/2}$
I_0 (corr) $= I_0(1 + 2gI_c)$

Table 3

Fractional Atomic Coordinates and Equivalent Isotropic Thermal Parameters (Å^2) for **6a**. Standard Deviations in Parentheses Refer to the Least Significant Digits

$B_{eq} = (8\pi^2/3)\Sigma_i\Sigma_j U_{ij}a^*{}_i a^*{}_j a_i \bullet a_j$

Atom [a]	x	y	z	B$_{eq}$
O	0.5823 (3)	0.4247 (3)	0.5829 (2)	3.3 (2)
N (1)	0.6472 (3)	0.5799 (3)	0.5183 (2)	3.1 (2)
N (2)	0.8075 (4)	0.6970 (3)	0.5697 (2)	3.3 (2)
C (1)	0.7726 (4)	0.5179 (4)	0.6292 (3)	2.5 (2)
C (2)	0.8393 (4)	0.6202 (4)	0.6277 (3)	2.8 (2)
C (3)	0.9333 (5)	0.6406 (4)	0.6854 (3)	3.6 (3)
C (4)	0.9616 (5)	0.5616 (5)	0.7424 (3)	4.1 (3)
C (5)	0.8973 (6)	0.4609 (4)	0.7434 (3)	4.3 (3)
C (6)	0.8022 (5)	0.4402 (4)	0.6879 (3)	3.6 (3)
C (7)	0.6619 (4)	0.4026 (4)	0.5750 (3)	2.6 (2)
C (8)	0.7469 (4)	0.6602 (4)	0.4967 (3)	2.7 (2)
C (9)	0.6892 (4)	0.7628 (4)	0.4531 (3)	3.4 (3)
C (10)	0.7878 (6)	0.7970 (5)	0.3924 (3)	6.2 (4)
C (11)	0.8962 (5)	0.7149 (5)	0.3912 (3)	4.8 (3)
C (12)	0.8449 (5)	0.6142 (4)	0.4367 (3)	3.7 (3)

[a] Atom numbers refer to Figure 2.

Finally, direct determination of the structure of **6a** was obtained by X-ray crystallography, as shown in Figure 2 and detailed in Tables 2-5. From Tables 4 and 5 one notes that the cyclopentane ring is distorted from a planar, equilateral pentagon. Thus, bond lengths in this ring vary from 1.49 to 1.55 Å and internal ring angles vary from 103.4° (*i.e.* less than 108°, expected for a regular polygon) to

Table 4

Bond Lengths for **6a**. Standard Deviations in Parentheses Refer to the Least Significant Digits

Atom [a]	Atom	Distance (Å)	Atom	Atom	Distance (Å)
O	C7	1.252 (5)	C3	C4	1.376 (6)
N1	C7	1.335 (5)	C4	C5	1.378 (7)
N1	C8	1.455 (5)	C5	C6	1.375 (6)
N2	C2	1.379 (5)	C8	C9	1.549 (6)
N2	C8	1.443 (5)	C8	C12	1.529 (6)
C1	C2	1.407 (6)	C9	C10	1.496 (6)
C1	C6	1.388 (6)	C10	C11	1.491 (7)
C1	C7	1.471 (6)	C11	C12	1.523 (6)
C2	C3	1.391 (6)			

[a] Atom numbers refer to Figure 2.

Table 5

Bond Angles in **6a**. Standard Deviations in Parentheses Refer to the Decimal Digits

Atom [a]	Atom	Atom	Angle (°)	Atom	Atom	Atom	Angle (°)
C7	N1	C8	123.7 (4)	O	C7	C1	122.5 (4)
C2	N2	C8	119.7 (4)	N1	C7	C1	116.2 (4)
C2	C1	C6	119.3 (4)	N1	C8	N2	107.4 (4)
C2	C1	C7	118.5 (4)	N1	C8	C9	111.8 (4)
C6	C1	C7	121.6 (5)	N1	C8	C12	113.1 (4)
N2	C2	C1	118.6 (4)	N2	C8	C9	108.8 (4)
N2	C2	C3	122.5 (5)	N2	C8	C12	112.3 (4)
C1	C2	C3	118.9 (5)	C9	C8	C12	103.4 (4)
C2	C3	C4	120.6 (5)	C8	C9	C10	106.1 (4)
C3	C4	C5	120.7 (5)	C9	C10	C11	109.8 (4)
C4	C5	C6	119.6 (5)	C10	C11	C12	104.8 (4)
C1	C6	C5	121.0 (5)	C8	C12	C11	105.8 (4)
O	C7	N1	121.3 (4)				

[a] Atom numbers refer to Figure 2.

109.8° (*i.e.* close to the tetrahedral angle). Also the sum of the internal angles in the ring is 529.9 ± 2°, *i.e.* less than the 540° required for planarity. Likewise, the pyrimidinone ring is nonplanar, while the benzene ring (within the accuracy of measurement) is planar (sums of internal angles = 704.1±2.4° and 720.1±2.9°, respectively).

As indicated earlier [2], both Carrington [3] and Böhme and Böing [4] reported the synthesis of dimethyltetrahydroquinazolinone **3** from reaction of **2a** with acetone. Although each group obtained a product which gave the expected elemental analysis for C, H, and N, the former worker reported a melting point of 262° and the latter workers, of 182°. To investigate this discrepancy we carried out the reaction exactly as described by Carrington, including a single crystallization [16] of the crude product (mp 169-170.5°) from methanol to give colorless needles [17], mp 260-261°. However, tlc, a mass spectrum, and chromatographic separation of the 261° crystals showed the presence of three fluorescent products: **3** (colorless micaceous plates from water [18], mp 184-185.5°, 35% yield); an unknown powder (mp > 230° dec, major component, readily soluble in ethanol and precipitated by addition of ether, largely soluble in water and clearly a mixture of various substances), and a volatile, minor component (apparent mol wt 392). Spectral properties of our 185° product are consistent with structure **3**. One should note a preference for the synthesis of **3** by the method of Böhme and Böing over that of Carrington. One does wonder why Carrington obtained the correct analysis for his sample.

EXPERIMENTAL [19]

2,2-Tetramethylene-1,2,3,4-tetrahydroquinazolin-4-one (**6a**).

A mixture of 2.22 g (16.3 mmoles) of anthranilamide (Aldrich), 2.10 g (25 mmoles) of cyclopentanone, 100 ml of absolute ethanol, and 0.8 ml of concentrated hydrochloric acid (pH of mixture *ca.* 3.4) was stirred at room temperature for four hours and then cooled to 0°. Crude **6a** (2.70 g, 82%, mp 260-264°) was collected by filtration, washed with a small amount of cold 95% ethanol, and recrystallized as colorless prisms (blue fluorescent in solution) from methanol, mp 259-260° (lit 254° [4,7]); ir: 3283, 3167 (NH stretching), 3009, 1643 (amide I), 1609, 1520 (amide II), 1274 (amide III), 755 cm^{-1} [20]; ^{1}H nmr: δ 8.11 (s, 1H, NH-3), 7.57 (d, $J_{5,6}$ = 7.8 Hz, 1H, H-5), 7.21 (dt, J = 7.8, 1.5 Hz, 1H, H-7), 6.75 (s, 1H, NH-1), 6.69 (d, $J_{7,8}$ = 7.8 Hz, 1H, H-8), 6.63 (t, J = 7.8 Hz, 1H, H-6), 1.78 and 1.67 (2 split s, 8H, cyclopentane H atoms); ^{13}C nmr: see Table 1; ms: m/z 202 (M$^+$, 17), 174 (16), 173 (M$^+$ - C$_2$H$_5$, 100), 160 (M$^+$ - C$_3$H$_6$, 8), 119 (13); X-ray analysis: see Tables 2-5 and Figure 2.

Anal. Calcd. for C$_{12}$H$_{14}$N$_2$O: C, 71.26; H, 6.98; N, 13.85. Found: C, 71.34; H, 7.02; N, 13.83.

2,2-Pentamethylene-1,2,3,4-tetrahydroquinazolin-4-one (**6b**).

In the preceding manner 2.22 g of anthranilamide and 2.51 g (25.6 mmoles) of cyclohexanone produced 2.10 g (60%) of crude **6b**, mp 227-230°, obtained as colorless prisms (blue fluorescent in solution) on recrystallization from 95% ethanol, mp 225-226° (lit 225° [4,7]); ir: 3363, 3173 (NH stretching), 2922, 1647 (amide I), 1606, 1498 (amide II), 753 cm^{-1} [20]; ^{1}H nmr: δ 7.93 (s, 1H, NH-3), 7.55 (dd, $J_{5,6}$ = 7.5 Hz, $J_{5,7}$ = 1.0 Hz, 1H, H-5), 7.21 (dt, J = 7.5, 1.5 Hz, 1H, H-7), 6.80 (d, $J_{7,8}$ = 8.1 Hz, 1H, H-8), 6.61 (s, 1H, NH-1) superimposed on 6.61 (t, J = 7.5 Hz, 1H, H-6), 1.2-1.8 (m, 10H, cyclohexane H atoms); ^{13}C nmr: see Table 1; ms: m/z 216 (M$^+$, 20), 187 (10), 173 (M$^+$ - C$_3$H$_7$, 100), 160 (M$^+$ - C$_4$H$_8$, 34), 120 (13).

2,2-Hexamethylene-1,2,3,4-tetrahydroquinazolin-4-one (**6c**).

In a modification of the preceding method, the reaction mixture from 2.22 g of anthranilamide, 2.73 g (24.4 mmoles) of cycloheptanone and the acidified ethanol was stirred for one hour, refluxed for 50 minutes, and then refrigerated to give 1.40 g (37%) of crude **6c**, mp 202-204°, obtained as colorless prisms (blue fluorescent in solution) on recrystallization from 95% ethanol, mp 204-205°; ir: 3336, 3269, 3186, and 3051 (NH-1 and NH-3 stretching), 2922, 2847, 1647, (amide I), 1607, 1512 (amide II), 1275 (amide III), 753 cm^{-1} [20]; ^{1}H nmr: δ 8.04 (s, 1H, NH-3), 7.54 (dd, $J_{5,6}$ = 7.8 Hz, $J_{5,7}$ = 1.5 Hz, 1H, H-5), 7.20 (dt, J = 7.8, 1.5 Hz, 1H, H-7), 6.73 (s, 1H, NH-1), 6.70 (d, $J_{7,8}$ = 7.8 Hz, 1H, H-8), 6.60 (t, J =7.8 Hz, 1H, H-6), 1.86 (broad s, 4H, 2 H-2' and 2 H-7'), 1.50 (s, 8H, 2 each H-3' to H-6'); ^{13}C nmr: see Table 1; ms: m/z 230 (M$^+$, 18), 187 (27), 173 (M$^+$ - C$_4$H$_9$, 100), 160 (M$^+$ - C$_5$H$_{10}$, 19).

Anal. Calcd. for C$_{14}$H$_{18}$N$_2$O: C, 73.01; H, 7.88; N, 12.16. Found: C, 72.98; H, 7.59; N, 12.14.

Carrington Procedure [3].

Following the published procedure a mixture of 2.7 g of **2**, 40 ml of 3.5% hydrochloric acid, and 40 ml of acetone was refluxed for 15 hours. The mixture was evaporated using a rotary evaporator until crystals started to form, cooled, scratched, and filtered to yield 2.08 g of off-white powder, mp 169-170.5°. One crystallization from methanol gave colorless needles, mp 260-261° (lit 262°); R$_f$ (chloroform/silica gel) 0 only; R$_f$ (ethyl acetate/silica gel) two strongly fluorescent spots (blue) 0.05 and 0.29; ms: m/z 392 (volatile impurity), 176 (apparently **3**). Evaporation of the combined residues gave 1.34 g of crude product, R$_f$ (ethyl acetate/silica gel) 0.05 (medium), 0.29 (strong), 0.44 (faint).

Column chromatography (silica gel/chloroform, ethyl acetate, methanol in succession) of the total product gave 1.22 g of **3** (35%) and then 1.54 g of an unknown material. This unknown material was readily soluble in 95% ethanol, from which it precipitated as a non-crystalline powder on addition of ether, mp, sinters at 230° and vaporizes slowly up to 259° dec; non-sublimable at 200° (0.07 mm); found to be a mixture of byproducts, largely soluble in water.

2,2-Dimethyl-1,2,3,4-tetrahydroquinazolin-4-one (**3**).

The preceding crude **3** crystallized as colorless, fluorescent, micaceous plates from water, mp 184-185.5° (lit [4], 182° with partial sublimation); ir: 3330, 3254, 3178, and 3045 (NH-1 and NH-3 stretching), 2970, 1644 (amide I), 1609, 1513 and/or 1535 (amide II), 1388, 752 cm^{-1} [20]; ^{1}H nmr: δ 7.94 (s, 1H, NH-3), 7.57 (split d, $J_{5,6}$ = 7.2 Hz, $J_{5,7}$ = 1.2 Hz, 1H, H-5), 7.21 (dt, J = 7.5, 1.5 Hz, 1H, H-7), 6.64 (s, 1H, NH-1) which overlaps 6.62 (pseudo-t, 2H, H-6 and H-8), 1.37 (s, 6H, 2 CH$_3$ groups); ^{13}C nmr: see Table 1, Attached Proton Test spectrum: positive peaks at δ 163.1 (C-4), 147.1 (C-8a), and 113.8 (C-4a); negative peaks at δ 133.2 (C-7), 127.2 (C-5), 116.5 and 114.2 (C-6 and C-8), and 29.0 (CH$_3$); C-2 signal not observed [21]; ms: m/z 176 (M$^+$, 8), 162 (10), 161 (M$^+$ - CH$_3$, 100), 120 (**161** - CH$_3$CN, 23), 92 (**120** - CO, 10).

5,5-Tetramethylene-4,5,6,7-tetrahydrothieno[2,3-*e*]pyrimidin-7-one (**5a**).

This compound was incorrectly assigned structure **4a** previously [2]. New structural assignments are ir: 3264, 3161, 3155, and 3072 cm^{-1} (NH stretching); ^{1}H nmr: δ 7.54 (s, 1H, NH-6), 7.05 (s, 1H, NH-4); ^{13}C mnr: see Table 1.

5,5-Pentamethylene-4,5,6,7-tetrahydrothieno[2,3-*e*]-pyrimidin-7-one (**5b**).

This compound was incorrectly assigned structure **4b** previously [2]. New structural assignments are ir: 3356, 3229, 3163, and 3027 cm^{-1} (NH stretching); ^{1}H nmr: δ 7.36 (s, 1H, NH-6), 6.96 (broad s, 1H, NH-4); ^{13}C nmr: see Table 1; Attached Proton Test spectrum: positive peaks at δ 161.2 (C-7), 151.3 (C-3a), 70.1 (C-5), 36.2, 24.7, and 21.1 (3 CH$_2$ groups); negative peaks at δ 132.2 (C-2) and 118.2 (C-3); C-7a signal not observed [22].

5,5-Hexamethylene-4,5,6,7-tetrahydrothieno[2,3-*e*]pyrimidin-7-one (**5c**).

This compound was incorrectly assigned structure **4c** previously [2]. New structural assignments are ir: 3304, 3232, 3176, and 3024 cm^{-1} (NH stretching); ^{1}H nmr: δ 7.48 (s, 1H, NH-6), 7.05 (s, 1H, NH-4); ^{13}C nmr: see Table 1.

Acknowledgment.

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[18]   Consistent with reported characteristics [4].
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[22]   Same as [21], except for 2784 acquisitions.