$CONH_2$), 3.72 (t, 1, J = 5 Hz, proton 4), and 3.3 (d, 2, J = 5 Hz, methylene).

Anal. Calcd for $C_{11}H_{10}N_2O_3$: C, 60.54; H, 4.62; N, 12.84; mol wt, 218.21. Found: C, 60.71; H, 4.39; N, 12.54; M^+, 218.

The compound did *not* induce sneezing.

2-Benzazepine-1,3-dione (14). A. *cis-o*-Carboxycinnamonitrile (13) was prepared, following literature procedure, by treating 21 g of 2-nitroso-1-naphthol in 1 l. of ligroin with 26 g of PCl_5, stirring for several hours while HCl was evolved copiously, allowing the mixture to stand for 3 days, and taking the crude solid into *ca.* 200 ml of 10% NaOH solution; acidification of the filtered solution with 15% HCl and recrystallization of the cyano acid from methanol gave crystals of **13**, mp 177–179° (lit.[9] mp 172°, 179°).

B. PPA Closure.—A suspension of 2 g of **13** in 42 g of PPA was heated in a steam cone and stirred for 0.5 hr. The purple-brown solution was cooled and hydrolyzed with cold water; the crude crystals were collected, washed with water, triturated with dilute $NaHCO_3$ solution, and again collected, washed with water, and dried. Recrystallization from ether, filtering the solution free of dark sediment, gave 0.5 g of crystals: mp 142–143°; ir 3.14–3.28 (bonded NH), and 6.03–6.06 μ, with shoulders at 5.90–5.95 μ; uv 224 nm (ϵ 31,980), 281 (8600), and 321 (3830), with inflections at 288 (8360) and 306 (5740); nmr ($CDCl_3$) δ 9.4 (m, 1, exchanges with D_2O, imide NH), 8.5 (m, 1, proton 9), 7.6 (m, 3, ArH), 7.16 (d, 1, J = 12.5 Hz, proton 5), and 6.4 [q, 1, J = 12.5 Hz, coupling to proton 5, J' = 2.5 Hz (long range coupling to imide NH, disappearing on D_2O exchange of NH), proton 4].

Anal. Calcd for $C_{10}H_7NO_2$: C, 69.36; H, 4.07; N, 8.09. Found: C, 69.49; H, 4.14; N, 8.03.

4,5-Dihydro-2-benzazepine-1,3-dione (15).—Hydrogenation of **14** as for **11** at 65° for 1.5–2 hr, and evaporation of the filtered, colorless solution, gave crystals, from ethyl acetate–ether: mp 118.5–120.5°; ir 3.11–3.25, 5.90, and 6.00 μ; uv 239 nm (ϵ 12,870) and 282 (1760); nmr ($CDCl_3$) δ 8.76 (m, 1, exchanges with D_2O, imide NH), 8.1 (m, 1, proton 9), 7.34 (m, 3, ArH), and 2.96 (resembling q, 4, J's not first order, methylenes).

Anal. Calcd for $C_{10}H_9NO_2$: C, 68.56; H, 5.18; N, 8.00. Found: C, 68.22; H, 5.25; N, 7.93.

Registry No.—**2**, 36004-42-9; **3**, 36004-43-0; **4**, 36004-44-1; **5**, 36004-45-2; **6**, 36015-22-2; **6** (tricarboxylic acid derivative), 36004-46-3; **7**, 36004-47-4; **8**, 36004-48-5; **9**, 36004-49-6; **11**, 36004-50-9; **12**, 36004-51-0; **14**, 36004-52-1; **15**, 36004-53-2.

Synthesis of Some 7-Aryl-6-azapteridines from 1,2,4-Triazine Intermediates

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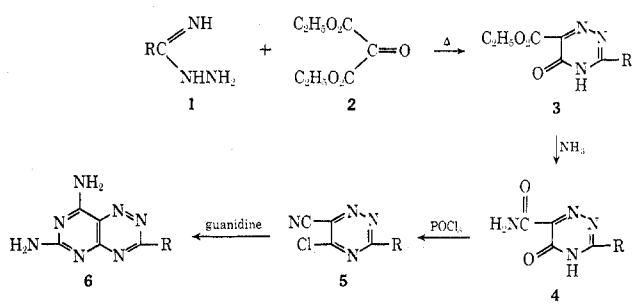
Interest in the pyrimido[4,5-*e*]-*as*-triazine (6-azapteridine) and pyrimido[5,4-*e*]-*as*-triazine (7-azapteridine) ring systems has intensified recently[2–7] as a consequence of the discovery that certain derivatives of the former ring system exhibit antiviral activity,[8] and that a number of broad-spectrum antibiotics (toxoflavin, fervenulin, and 2-methylfervenulone) are derivatives of the 7-azapteridine ring system.[9] We have explored the possibility of utilizing a common intermediate for the synthesis of both isomeric ring systems, and our results are summarized in this brief report.

The condensation of amidrazones with α,β-dicarbonyl compounds to give *as*-triazines is well known,[10–12] but none of the many *as*-triazines thus far prepared by this route possesses substituent groups suitable for subsequent cyclization to an azapteridine. It occurred to us that the reaction of amidrazones (**1**) with diethyl oxomalonate (**2**) should afford *as*-triazines[13] (**3**) which could readily be converted into 6-azapteridines (**6**) by the sequence of reactions depicted in Scheme I.

SCHEME I



Indeed, this concept proved to be successful when R = aryl, but it failed at the dehydration–chlorination step (**4** → **5**) with R = alkyl (CH_3, C_2H_5). Many different reaction conditions were explored ($POCl_3$, $POCl_3$–pyridine, $POCl_3$–DMF, $SOCl_2$–pyridine, $SOCl_2$–DMF), but in all cases only resinous, uncharacterizable products were obtained.

There is little ambiguity as to the structure of the condensation products formed in the above reaction (see Scheme I), since the keto grouping of diethyl oxomalonate is considerably more reactive than the ester groups, and N^1 of the amidrazone[14] is the most nucleophilic nitrogen. However, the structural assignments were confirmed independently by the unequivocal synthesis of **3** (R = C_6H_5, CH_3) by the reaction of the hydrazone of diethyl oxomalonate (**8**)[15] with the imidate esters **7** (R = C_6H_5, CH_3) (see Scheme II). The products of this latter condensation were identical in every respect with the corresponding compounds prepared by the alternate procedure described in Scheme I.

In principle, protection of N^1 of the amidrazone fol-

(1) NIH Predoctoral Fellow, 1969–1972.

(2) C. Temple, Jr., C. L. Kussner, and J. A. Montgomery, *J. Org. Chem.*, **36**, 3502 (1971), and references cited therein.

(3) C. Temple, Jr., C. L. Kussner, and J. A. Montgomery, *J. Heterocycl. Chem.*, **8**, 1099 (1971).

(4) F. Yoneda, K. Shinomura, and S. Nishigaki, *Tetrahedron Lett.*, 851 (1971).

(5) F. Yoneda, M. Kanahori, and S. Nishigaki, *J. Heterocycl. Chem.*, **8**, 523 (1971).

(6) F. Yoneda, M. Kanahori, K. Ogiwara, and S. Nishigaki, *ibid.*, **7**, 1443 (1970).

(7) D. J. Brown and T. Sugimoto, *J. Chem. Soc. C*, 2616 (1971).

(8) C. Küchler, W. Küchler, and L. Heinisch, *Arzneim.-Forsch.*, **16**, 1122 (1966).

(9) E. C. Taylor and S. F. Martin, *J. Org. Chem.*, **35**, 3792 (1970), and references cited therein.

(10) R. L. Jones and J. R. Kershaw, *Rev. Pure Appl. Chem.*, **21**, 23 (1971).

(11) H. Neunhoeffer, H. Hennig, H.-W. Fruhauf, and M. Mutterer, *Tetrahedron Lett.*, 3147 (1969).

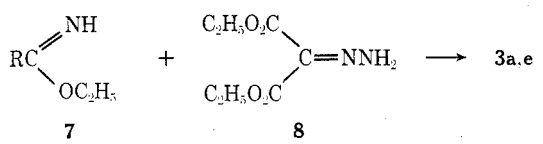
(12) M. Brugger, H. Wamhoff, and F. Korte, *Justus Liebigs Ann. Chem.*, **755**, 101 (1972).

(13) This route to *as*-triazines is not new; diethyl oxomalonate and thiosemicarbazide, for example, are known to give **3**, R = SH [R. B. Barlow and A. D. Welch, *J. Amer. Chem. Soc.*, **78**, 1258 (1956)].

(14) See H. Rapoport and R. M. Bonner, *J. Amer. Chem. Soc.*, **72**, 2783 (1950), for the nomenclature of amidrazones.

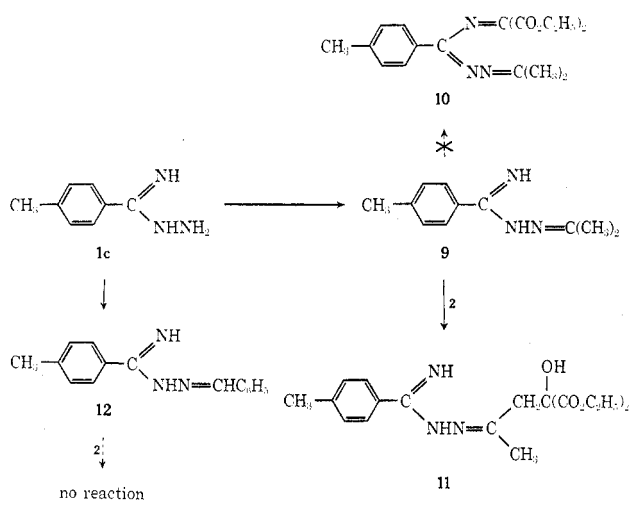
(15) E. Ciganek, *J. Org. Chem.*, **30**, 4366 (1965).

SCHEME II



lowed by condensation with diethyl oxomalonate should result in initial imine formation; removal of the protecting group and subsequent intramolecular cyclization would then lead to the isomeric 7-azapteridine ring system, thus realizing our objective of preparing both ring systems from a common precursor. Unfortunately, our efforts thus far to effect this reverse condensation mode have been unsuccessful. Compound **1c** was successfully converted into its isopropylidene derivative **9** by reaction with acetone, but subsequent reaction with diethyl oxomalonate (**2**) proceeded anomalously to give **11** rather than **10**, the expected product. In an effort to avoid this unexpected reaction, **1c** was converted into its benzylidene derivative **12**, but this failed to react with diethyl oxomalonate (see Scheme III).

SCHEME III



Experimental Section

General.—Melting points were determined on a Thomas-Hoover apparatus and are uncorrected. Imidate ester hydrochlorides were prepared from the corresponding nitriles using standard procedures.[16] All of the aryl-substituted amidrazones were prepared by the reaction of anhydrous hydrazine (1 equiv) with the appropriate imino ester free base (as described below for *p*-chlorobenzamidrazone) except for the case of **1d** (R = 2-pyridyl) which was prepared by the method of Case.[17] Aliphatic amidrazones were prepared by the method of Wright, Halliday, and Davis.[18]

***p*-Chlorobenzamidrazone (1b).**—Ethyl *p*-chlorobenzimidate hydrochloride (10.0 g, 0.0454 mol) was shaken with 100 ml of 5% aqueous sodium hydroxide solution and 150 ml of ether. The aqueous layer was extracted with ether (1 × 75 ml); the combined ether layers were washed with water until they were no longer basic and then dried over 5A molecular sieves. The ether was evaporated under reduced pressure giving 7.2 g (0.039 mol) of crude ethyl *p*-chlorobenzimidate. A solution of 97% anhydrous hydrazine (1.30 g, 0.039 mol) in a mixture of 100 ml of anhydrous ether and 15 ml of anhydrous ethanol was added and the solution allowed to stand at 0° for 2 days. The excess solvent was evaporated under reduced pressure and the product recrystallized from isopropyl ether to give 4.28 g (56%) of white flakes, mp 87–89°. *Anal.* Calcd for $C_7H_8N_3Cl$: C, 49.57; H, 4.75; N, 24.79; Cl, 20.90. Found: C, 49.24; H, 4.72; N, 24.96; Cl, 21.21.

6-Carbethoxy-3-phenyl-5(4*H*)-*as*-triazinone (3a). Method A.[19] —To a solution of diethyl oxomalonate (**2**) (3.64 g, 20.9 mmol) dissolved in 10 ml of isopropyl alcohol was slowly added a solution of benzamidrazone (3.25 g, 20.9 mmol) in 20 ml of isopropyl alcohol. The solution was stirred at room temperature for 6 hr and then refluxed 2 hr to complete the cyclization. Evaporation of the excess solvent under reduced pressure, trituration of the residue with a small volume of cold ethyl acetate (*ca.* 20 ml), and recrystallization from ethyl acetate afforded 3.28 g (64%) of a colorless granular solid, mp 202–203°. *Anal.* Calcd for $C_{12}H_{11}N_3O_3$: C, 58.77; H, 4.52; N, 17.14. Found: C, 58.62; H, 4.59; N, 17.09.

Method B.—A solution of the hydrazone of diethyl oxomalonate (1.26 g, 6.72 mmol) and ethyl benzimidate (1.00 g, 6.72 mmol) in 25 ml of isopropyl alcohol was refluxed 16 hr. The excess solvent was evaporated under reduced pressure and about 10 ml of ethyl acetate added to the residue. Cooling and filtering gave 0.61 g of crude product which was recrystallized from a small volume of ethyl acetate, yielding 0.43 g (26%). No attempt was made to optimize the yield. The product was identical with that obtained by method A.

The following compounds were prepared analogously by method A.

6-Carbethoxy-3-(*p*-chlorophenyl)-5(4*H*)-*as*-triazinone (3b): 62%, mp 244° dec (translucent plates from isopropyl alcohol). *Anal.* Calcd for $C_{12}H_{10}N_3ClO_3$: C, 51.53; H, 3.36; N, 15.03; Cl, 12.68. Found: C, 51.40; H, 3.57; N, 14.91; Cl, 12.86.

6-Carbethoxy-3-(*p*-tolyl)-5(4*H*)-*as*-triazinone (3c): 53%, mp 222–223° dec (colorless needles from isopropyl alcohol). *Anal.* Calcd for $C_{13}H_{13}N_3O_3$: C, 60.22; H, 5.05; N, 16.21. Found: C, 60.01; H, 5.04; N, 16.16.

6-Carbethoxy-3-(2-pyridyl)-5(4*H*)-*as*-triazinone (3d): 62%, mp 142–143° (very fine colorless needles from ethyl acetate). *Anal.* Calcd for $C_{11}H_{10}N_4O_3$: C, 53.66; H, 4.09; N, 22.76. Found: C, 53.55; H, 4.21; N, 22.80.

6-Carbethoxy-3-methyl-5(4*H*)-*as*-triazinone (3e) (method A): 55%, mp 160–161° (colorless granular solid from ethyl acetate). *Anal.* Calcd for $C_7H_9N_3O_3$: C, 45.90; H, 4.95; N, 22.94. Found: C, 45.82; H, 5.14; N, 23.13.

Method B gave 68% yield.

6-Carbethoxy-3-ethyl-5(4*H*)-*as*-triazinone (3f): 61%, mp 202–204° (colorless granular solid from benzene–ether). *Anal.* Calcd for $C_8H_{11}N_3O_3$: C, 48.72; H, 5.62; N, 21.31. Found: C, 48.46; H, 5.89; N, 21.48.

6-Carbamoyl-3-phenyl-5(4*H*)-*as*-triazinone (4a).—Compound **3a** (1.75 g) was suspended in 50 ml of anhydrous methanol and the mixture saturated at 0° with dry ammonia gas. The resulting mixture was stirred for 18 hr at room temperature and concentrated to about half of its original volume under reduced pressure; the product was collected by filtration and then recrystallized from isopropyl alcohol to give 1.45 g (83%) of fine colorless needles, mp 301° dec. *Anal.* Calcd for $C_{10}H_8N_4O_2$: C, 55.55; H, 3.73; N, 25.92. Found: C, 55.58; H, 3.77; N, 25.88.

The following compounds were prepared analogously.

6-Carbamoyl-3-(*p*-chlorophenyl)-5(4*H*)-*as*-triazinone (4b): 85%, mp 322–323° dec (colorless flakes from ethanol). *Anal.* Calcd for $C_{10}H_7N_4ClO_2$: C, 47.92; H, 2.81; N, 22.35. Found: C, 48.05; H, 2.77; N, 22.40.

6-Carbamoyl-3-(*p*-tolyl)-5(4*H*)-*as*-triazinone (4c): 84%, mp 319–320° dec (white microcrystalline solid from isopropyl alcohol). *Anal.* Calcd for $C_{11}H_{10}N_4O_2$: C, 57.38; H, 4.38; N, 24.34. Found: C, 57.43; H, 4.52; N, 24.48.

6-Carbamoyl-3-(2-pyridyl)-5(4*H*)-*as*-triazinone (4d): 89%, mp 294–295° dec (colorless granular solid from ethanol). *Anal.*

(16) (a) S. M. McElvain and J. W. Nelson, *J. Amer. Chem. Soc.*, **64**, 1825 (1942); (b) "Organic Syntheses," Collect. Vol. I, Wiley, New York, N. Y., 1943, p 6.

(17) F. H. Case, *J. Org. Chem.*, **30**, 931 (1965).

(18) G. C. Wright, R. P. Halliday, and C. S. Davis, *J. Pharm. Sci.*, **59**, 105 (1970).

(19) NOTE ADDED IN PROOF.—The preparation of this compound (mp 197–198° from ethanol) by the same route, and its conversion into several 6-azapteridines by reaction with PCl_5 to give 6-carbethoxy-5-chloro-3-phenyl-*as*-triazine, followed by condensation with acetamidine, benzam idine, and 1,3-dimethylurea, has recently been described [M. Brugger, H. Wamhoff, and F. Korte, *Justus Liebigs Ann. Chem.*, **758**, 173 (1972)].

Calcd for $C_9H_7N_5O_2$: C, 49.77; H, 3.25; N, 32.25. Found: C, 49.94; H, 3.20; N, 32.38.

6-Carbamoyl-3-methyl-5(4*H*)-*as*-triazinone (**4e**): 68%, mp 270° dec (cream-colored powdery solid from isopropyl alcohol). *Anal.* Calcd for $C_5H_6N_4O_2$: C, 38.96; H, 3.92; N, 36.35. Found: C, 38.80; H, 3.92; N, 36.53.

6-Carbamoyl-3-ethyl-5(4*H*)-*as*-triazinone (**4f**): 57%, mp 266° dec (short pale yellow needles from isopropyl alcohol). *Anal.* Calcd for $C_6H_8N_4O_2$: C, 42.85; H, 4.80; N, 33.32. Found: C, 42.66; H, 4.80; N, 33.25.

5-Chloro-6-cyano-3-phenyl-*as*-triazine (**5a**).—To a cooled mixture of phosphorus oxychloride (5 ml) and anhydrous pyridine (0.92 g, 11.6 mmol) was added portionwise with stirring **4a** (0.50 g, 2.32 mmol). The mixture was gently refluxed for 15 min, the excess phosphorus oxychloride evaporated under reduced pressure, and the viscous residue poured over ice. The aqueous solution was neutralized with sodium bicarbonate and then extracted with chloroform (4 × 25 ml). The combined extracts were dried over anhydrous magnesium sulfate and concentrated to dryness under reduced pressure. Recrystallization of the crude product from benzene–hexane afforded 0.34 g (68%) of yellow plates, mp 184–185°. *Anal.* Calcd for $C_{10}H_5N_4Cl$: C, 55.44; H, 2.32; N, 25.86. Found: C, 55.49; H, 2.20; N, 25.82.

The following compounds were prepared analogously.

5-Chloro-6-cyano-3-(*p*-chlorophenyl)-*as*-triazine (**5b**): 69%, mp 152–153° dec (very fine pale yellow needles from benzene–hexane). *Anal.* Calcd for $C_{10}H_4N_4Cl_2$: C, 47.83; H, 1.61; N, 22.31. Found: C, 47.64; H, 1.63; N, 21.99.

5-Chloro-6-cyano-3-(*p*-tolyl)-*as*-triazine (**5c**): 70%, mp 201–202° (yellow plates from acetonitrile). *Anal.* Calcd for $C_{11}H_7N_4Cl$: C, 57.27; H, 3.05; N, 24.29. Found: C, 57.25; H, 3.01; N, 24.28.

5-Chloro-6-cyano-3-(2-pyridyl)-*as*-triazine (**5d**): 73%, mp 151–153° dec (blunt yellow prisms from benzene–hexane). *Anal.* Calcd for $C_9H_4N_5Cl$: C, 49.67; H, 1.85; N, 32.19. Found: C, 49.47; H, 1.64; N, 32.15.

2,4-Diamino-7-phenylpyrimido[4,5-*e*]-*as*-triazine (**6a**).—Compound **5a** (0.25 g, 1.15 mmol) was suspended in a methanolic solution of guanidine (prepared from guanidine hydrochloride (0.22 g, 2.30 mmol) and metallic sodium (0.053 g, 2.30 g-atoms) in 10 ml of anhydrous methanol) and the mixture refluxed for 16 hr. The mixture was cooled and filtered; the solid was washed with water and recrystallized from dimethylformamide to give 0.15 g (55%) of a yellow microcrystalline solid, mp >300°. *Anal.* Calcd for $C_{11}H_9N_7$: C, 55.22; H, 3.79; N, 40.99. Found: C, 55.22; H, 3.93; N, 41.24.

The following compounds were prepared analogously.

2,4-Diamino-7-(*p*-chlorophenyl)pyrimido[4,5-*e*]-*as*-triazine (**6b**): 56%, mp >300° (yellow microcrystalline solid from dimethylformamide). *Anal.* Calcd for $C_{11}H_8N_7Cl$: C, 48.27; H, 2.95; N, 35.83. Found: C, 48.16; H, 2.91; N, 35.77.

2,4-Diamino-7-(*p*-tolyl)pyrimido[4,5-*e*]-*as*-triazine (**6c**): 64%, mp >300° (yellow microcrystalline solid from dimethylformamide). *Anal.* Calcd for $C_{12}H_{11}N_7$: C, 56.91; H, 4.38; N, 38.72. Found: C, 56.98; H, 4.52; N, 38.74.

2,4-Diamino-7-(2-pyridyl)pyrimido[4,5-*e*]-*as*-triazine (**6d**): 44%, mp >300° (yellow microcrystalline solid from dimethylformamide). *Anal.* Calcd for $C_{10}H_8N_8$: C, 49.99; H, 3.36; N, 46.65. Found: C, 49.88; H, 3.40; N, 46.75.

N^1-Isopropylidine-*p*-toluamidrazone (**9**).—*p*-Toluamidrazone (6.00 g) was dissolved in 100 ml of acetone and the solution stirred at room temperature for 30 min. Evaporation of the excess solvent under reduced pressure and recrystallization from hexane afforded 6.41 g (84%) of colorless needles, mp 72–73°. *Anal.* Calcd for $C_{11}H_{15}N_3$: C, 69.81; H, 7.99; N, 22.20. Found: C, 69.74; H, 8.06; M, 22.34.

Reaction of N^1-Isopropylidene-*p*-toluamidrazone with Diethyl Oxomalonate. Formation of 11.—To a solution of **9** (0.50 g, 2.65 mmol) in 10 ml of anhydrous benzene was added diethyl oxomalonate (**2**) (0.46 g, 2.65 mmol), and the solution was refluxed (Dean-Stark trap) for 3 hr. The excess solvent was evaporated under reduced pressure, the residual viscous oil dissolved in a minimum volume of hexane, and the solution cooled and filtered. Recrystallization from hexane gave 0.64 g (67%) of blunt colorless prisms, mp 78–79°. *Anal.* Calcd for $C_{18}H_{25}N_3O_5$: C, 59.49; H, 6.93; N, 11.56. Found: C, 59.33; H, 6.91; N, 11.37. Nmr ($CDCl_3$): δ 1.27 (t, 6), 2.15 (s, 3), 2.40 (s, 3), 3.18 (s, 2), 4.29 (q, 4), 7.23 (d, 2, J = 8 Hz), 7.74 (d, 2, J = 8 Hz).

Registry No.—**1b**, 36286-75-6; **3a**, 36286-76-7; **3b**, 36286-77-8; **3c**, 36286-78-9; **3d**, 36286-79-0; **3e**, 36286-80-3; **3f**, 36286-81-4; **4a**, 36294-41-4; **4b**, 36286-82-5; **4c**, 36286-83-6; **4d**, 36286-84-7; **4e**, 36286-85-8; **4f**, 36286-86-9; **5a**, 36286-87-0; **5b**, 36286-88-1; **5c**, 36286-89-2; **5d**, 36286-90-5; **6a**, 36286-91-6; **6b**, 36286-92-7; **6c**, 36286-93-8; **6d**, 36286-94-9; **9**, 36286-95-0; **11**, 36286-96-1.

Chloral–Hydrazone Adducts

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The acid-catalyzed oxidative dimerization of formaldehyde dimethylhydrazone (**1**), to glyoxal bis-dimethylhydrazone (**3**) by way of the head-to-head dimer, 2,2-dimethylhydrazinoacetaldehyde dimethylhydrazone (**2**) (eq 1), was described recently.[1] We

$$\underset{\mathbf{1}}{2Me_2NN{=}CH_2} \xrightarrow{H^-} \underset{\mathbf{2}}{[Me_2NN{=}CHCH_2NHNMe_2]} \xrightarrow{-2H} \underset{\mathbf{3}}{Me_2NN{=}CHCH{=}NNMe_2} \quad (1)$$

now wish to report the spontaneous addition of hydrazones, including **1**, to chloral (trichloroacetaldehyde, **4**), as in eq 2, a process that is analogous to the first step in eq 1.

$$R_1R_2NN{=}CHR_3 + \underset{\mathbf{4}}{Cl_3CCHO} \longrightarrow \underset{\mathbf{5}}{R_1R_2NN{=}CR_3CHOHCCl_3} \quad (2)$$

Chloral–hydrazone adducts that have been prepared and characterized are described in Table I. Those obtained from formaldehyde hydrazones are formulated as hydrazones of 2-hydroxy-3,3,3-trichloro-1-propanone (**5**, R_3 = H) on the bases of elemental analyses and the nmr spectral data, which include tests for exchangeable hydrogen with D_2O in acetone-d_6. Adducts were obtained also from benzaldehyde dimethylhydrazone and from formaldehyde methylphenylhydrazone, but they were too unstable to permit complete characterization.

Most of the adducts were light yellow, crystalline solids; one, from formaldehyde diethylhydrazone, was an oil. They all decomposed to black tars if kept at room temperature, but most could be kept indefinitely under refrigeration at −10°. Half-lives at room temperature ranged from about 1 hr to about 4 days. The decomposition products gave a positive test for chloride ion with aqueous silver nitrate.

The first chloral–hydrazone adduct resulted from an attempt to prepare chloral dimethylhydrazone by an exchange reaction between chloral and **1** under anhydrous conditions, since the reaction of chloral with 1,1-dimethylhydrazine gave only tarry decomposition products.[2] The formation of an adduct with structure **5** was unexpected, but the result can be rationalized in terms of structural features peculiar to

(1) F. E. Condon and D. Farcasiu, *J. Amer. Chem. Soc.*, **92**, 6625 (1970).
(2) *Cf.* R. L. Hinman and D. Fulton, *ibid.*, **80**, 1895 (1958).