Bottromycin A_1 benzylamide was prepared similarly using benzylamine crystallized from EtOAc; mp 160–168°, $[\alpha]^{25}D$ −56° (*c* 1, 95% EtOH). *Anal.* Calcd for $C_{48}H_{67}N_9O_6S \cdot H_2O$: C, 62.9; H, 7.5; N, 13.7; S, 3.5. Found: C, 63.0, H, 7.6; N, 13.4; S, 3.5.

Bottromycin A_1 Ethyl Ester.—A 10-mg/ml solution of bottromycin A_1 free base in anhydrous EtOH containing 15% Et_3N was heated at 50° in a sealed tube for 20 hr. The resultant mixture of bottromycin A_1 and the ethyl ester of bottromycin A_1 was separated by partition chromatography. *Anal.* Calcd for $C_{43}H_{64}N_8O_7S \cdot H_2O$; C, 60.4; H, 7.5; N, 13.1; S, 3.7. Found: C, 60.1; H, 7.4; N, 12.7; S, 3.4.

Bottromycin *t*-Butylamide.—Bottromycin carboxylate[16] (480 mg) was dissolved in 5 ml of anhydrous DMF and was treated successively at 0° with 219 mg of bis(2,4-dinitrophenyl) carbonate and 0.14 ml of Et_3N. The solution was stirred at 0° for 75 min and then at room temperature for 10 min. To this mixture was added 2 ml of *t*-$BuNH_2$. A precipitate formed in a few minutes. The mixture was heated at 50–60° for 30 min, diluted with 20 ml of $CHCl_3$, and filtered to remove the yellow by-product. The filtrate was concentrated to near dryness under reduced pressure. The residue was dissolved in $CHCl_3$ and then washed (saturated NaCl containing 5% NH_3) until the aqueous layer was colorless. The organic solution was dried (Na_2SO_4) and concentrated to dryness. A yellow glass (390 mg) was obtained. Tlc (10% MeOH in $CHCl_3$, silica gel) gave two zones when sprayed with bromophenol blue. The desired amide was located at R_f 0.70, and a slower moving substance R_f 0.15 was observed. These components were separated by preparative tlc yielding 197 mg of the *t*-butylamide as an off-white glass.

This general procedure was used to prepare the derivatives listed in Table III. In some cases the thin layer chromatograms were developed with MeOH–$CHCl_3$ of different composition to improve resolution of the components. The zones were sometimes developed by staining with iodine vapor or located by spraying the dried plate with H_2O.

(16) J. M. Waisvisz and M. G. van der Horven, *J. Am. Chem. Soc.*, **80**, 383 (1958).

5,5-Diarylpenta-2,4-dienoic Acid Amides as Potential Antimalarial Agents[1]

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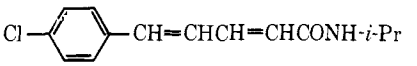
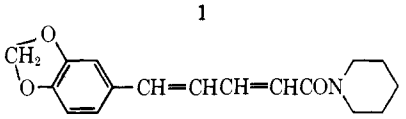
A series of 5,5-diarylpenta-2,4-dienoic acids and their amides have been synthesized and evaluated as antimalarial agents. The acids were prepared from the corresponding diaryl ketones either directly by a Reformatsky procedure or through acetylenic alcohol and acrolein intermediates. The preparation of a series of 3,3-bis(4-chlorophenyl)acrylic acid amides is also reported. One compound, N,N-diethyl-5,5-bis(4-chlorophenyl)-penta-2,4-dienoic acid amide, provided significant antiplasmodial activity.

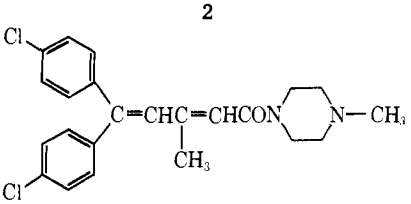
Among the more novel compounds revealed by the World War II malaria program to have interesting antiplasmodial action was N-isopropyl-5-(*p*-chlorophenyl)penta-2,4-dienoic acid amide (**1**).[2] This compound was four times more active than quinine in the chick *Plasmodium gallinaceum* assay employed in that work and had a therapeutic index of 12.5. Two other amides, obtained by coupling the same acid with guanidine and cyanoguanidine, were inactive.[3] Because of the development of resistance to chloroquine in many parts of the world by *Plasmodium falciparum*, there is an increasing need for new antimalarial drugs of novel structural type.[4,5] This need suggested that examination of additional chemical structures related to **1** for antiplasmodial properties would be of value.



1



2



3

In addition to antimalarial activity, other biological properties have been associated with pentadienoic acid derivatives. The hydrazide of 5-phenylpenta-2,4-dienoic acid has *in vitro* antituberculous activity,[6] and a series of the free acids shows inhibitory action against bacteria, yeast, and fungi.[7] Sorbic acid (2,4-hexadienoic acid) is widely used for its antifungal properties[8] and piperine (**2**, the pungent element of pepper) has insecticidal properties.[9] The use of derivatives of 5-(5-nitro-2-furyl)penta-2,4-dienoic acid as antibacterial agents has been patented.[10]

Of more pertinence to parasitic disease chemotherapy, it has been reported that a series of 5,5-diarylpenta-2,4-dienoic acid derivatives of piperazine (*e.g.*, **3**) possess marked activity against *Dicrocoelium dendriticum*, a liver fluke of considerable veterinary importance.[11,12] The closely related fluke, *Fasciola hepatica*, which infests both animals and man, is affected by these

(1) This work was supported by the U. S. Army Medical Research and Development Command under Contract No. DA-49-193-MO-2750. This contribution is no. 331 from the Army Research Program on Malaria.

(2) G. R. Coatney, W. C. Cooper, N. B. Eddy, and J. Greenberg, "Survey of Antimalarial Agents," Public Health Service Publication No. 193, Washington, D. C., 1953, p 139.

(3) Reference 2, p 137.

(4) *World Health Organ. Tech. Rept. Ser.*, **No. 296**, 1 (1965).

(5) P. J. Bartelloni, F. W. Sheely, and W. D. Tigertt, *J. Amer. Med. Ass.*, **199**, 141 (1967), and references cited therein.

(6) S. Kakimoto, I. Sekikawa, and K. Yamamoto, *J. Pharm. Soc. Japan*, **75**, 353 (1955); *Chem. Abstr.*, **50**, 1663e (1956).

(7) K. Takeichi, *Hakko Kogaku Zasshi*, **38**, 99, 224, 313, 431, 539, 602 (1960); *Chem. Abstr.*, **55**, 2791b (1961); **57**, 2669 (1962).

(8) Chemicals Used in Food Processing, Publication 1274, National Academy of Science, National Research Council, Washington, D. C., 1965, p 5.

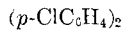
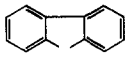
(9) E. K. Haevill, A. Hartzell, and J. M. Arthur, *Contrib. Boyce Thompson Inst.*, **13**, 87 (1943).

(10) H. Saikachi and S. Ogawa, Japanese Patent 17,981 (1962); *Chem. Abstr.*, **59**, P11426c (1963).

(11) M. Schorr, H. Loewe, E. Jürgens, H. Weber, and G. Lämmler, *Arzneim.-Forsch.*, **14**, 1151 (1964).

(12) G. Lämmler, *Z. Tropenmed. Parasitol.*, **15**, 164 (1964).

Table I
ω,ω-Diarylpentadienoic Acids and Derived Amides
$Ar_2C{=}CHCH{=}CHCOX$

No.	Ar₂	X	Yield, %	Mp, °C	Analyses[a]	Antimalarial act.[b] 40	160	640	Toxicity deaths[f]
8	$(p\text{-ClC}_6H_4)_2$		51	248–250 dec	C, H, N	0.5	1.9	...	5/5, 2/5
						0.2	0.7	...	5/5, 3/5
9			50	153–155	C, H, N	0.7	0.7	...	5/5 3/5
10		NEt_2	60	126–127	C, H, N	1.1	1.9	4.7	1/5
						0.3	1.1	6.2	1/5
						0.3	0.7	1.3[c]	
11		NMe_2	59	160–162	C, H, N	0.5	0.5	0.9	2/5
12		$NHCH(CH_3)_2$	59	194–199	C, H, N	0.3	0.3	0.3	
13		NH_2	47	190–194	C, H, N	0.1	0.3	0.3	
14		OH	53	212–216	C, H	0.1	0.3	0.3	
15	$(C_6H_5)_2$		51	Oil	C, H, N	0.0	0.2	0.8	2/5
16			49	83–91	C, H, N	0.6	0.8	0.8	3/5
17		NEt_2	41	Oil	C, H, N	0.0	0.2	0.2	
18		NMe_2	68	118–125	C, H, N	0.2	0.2	0.4	
19		$NCH(CH_3)_2$	60	179–180	C, H, N	0.1	0.1	0.5	
20		NH_2	54	161–163	C, H, N	0.1	0.1	0.1	
21		OH[d]	37	192–193		0.2	...	...	5/5, 5/5
22			38	132–134	C, H, N	0.0	0.2	0.2	
23			26	143–147	C, H, N	0.0	0.2	...	5/5
24		NEt_2	41	85–97	C, H, N	0.0	0.0	0.0	
25		NMe_2	42	142–145	C, H, N	0.6	0.6	1.0	
26		$NCH(CH_3)_2$	66	188–190	C, H, N	0.1	0.1	0.3	
27		NH_2	67	226–226.5	C, H, N	0.1	0.1	0.1	
28		OH	30	273–277	C, H	...	...	...	
29	$(p\text{-CH}_3OC_6H_4)_2$	NEt_2	36	Oil	C, H, N	0.1	0.3	0.3[c]	
30		$NHCH(CH_3)_2$	75	118–120	C, H, N	0.3	0.3	0.3	
31		NH_2	37	172–178	C, H, N	...	...	...	
32		OH	[e]	186–189	C, H	...	...	...	
33	$(p\text{-F-C}_6H_4)_2$	NEt_2	40	125–128	C, H, N	0.2	0.6	0.8	1/5
34		NH_2	37	217–222	C, H, N	0.0	0.6	0.8	1/5
35		OH	10	180–185	C, H	...	...	...	

[a] Where analyses are indicated only by symbols of the elements, analytical results obtained for those elements were within ±0.4% of the theoretical values. Absorption bands of spectra (uv, ir, pmr) were as expected. [b] See ref 17. [c] Dosages of 20, 80, and 320 mg/kg, respectively. [d] Reference 20. [e] See Experimental Section. [f] First fraction indicates deaths for the 640-mg/kg dose and the second fraction for the 160-mg/kg dose.

compounds to a lesser extent.[12] Malaria and liver fluke chemotherapy have previously been correlated through the drug 1,4-bis(trichloromethyl)benzene. This compound is curative for *Plasmodium berghei* infected mice[13] and has had successful veterinary[14] and human clinical[15,16] use against several liver flukes.

Because we were aware of interest in monoarylpentadienoic acid amides in another laboratory, we chose to concentrate our development of this lead on 5,5-diarylpenta-2,4-dienoic acid amides, thus expanding on the possible malaria–liver fluke chemotherapy correlation. A series of five diaryl acids were prepared and converted to various amides. A short series of amides from a lower vinylog, 3,3-bis(4-chlorophenyl)-acrylic acid, was also prepared.

During the course of this study the lead compound (**1**) was found to be inactive against *P. berghei* in the mouse screen.[17] Subsequently Werbel, Headen, and Elslager reported a series of nineteen closely related 5-phenylpenta-2,4-dienoic acid amides bearing a variety of substituents on the phenyl and amide functions.[18] Although no numerical results were provided, none of these compounds appeared to provide interesting antimalarial activity against *P. berghei*.[17,18]

Chemistry.—The dienoic acids have been prepared by several procedures. Initial attempts to prepare the diphenyl derivative (**21**, Table I) by the direct base-catalyzed condensation of benzophenone with methyl crotonate in dimethyl sulfoxide solution did not yield the desired acid. This procedure has proven effective in the synthesis of diaryldienoic acids when β-methylcrotonic ester was employed as the active methyl substrate.[11]

(13) Information given in separate presentations by D. P. Jacobus and E. F. Elslager at the Symposium on Drug-Refractory Malaria, 153rd National Meeting of the American Chemical Society, Miami Beach, Fla., April 10, 1967.

(14) J. C. Boray, F. A. Happich, and J. C. Andrews, *Vet. Rec.*, **80**, 218 (1967), and references cited therein.

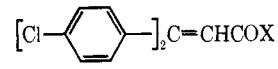
(15) W. Sheng-miao, *et al.*, *Chinese Med. J.*, **84**, 748 (1965).

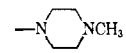
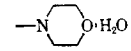
(16) C. Huei-lan, K. Hsiai-ying, F. Wei-chi, and H. Chih-piao, *ibid.*, **84**, 756 (1965).

(17) All *P. berghei* bioassays reported in this paper were performed by Dr. Leo Rane of the University of Miami by a published procedure [T. S. Osdene, P. B. Russell, and L. Rane, *J. Med. Chem.*, **10**, 431 (1967)]. Testing results were supplied through the courtesy of Dr. David P. Jacobus of the Walter Reed Army Institute of Research.

(18) L. M. Werbel, N. Headen, and E. F. Elslager, *ibid.*, **10**, 366 (1967).

Table II
3,3-Bis(4-chlorophenyl)acrylic Acid and Derived Amides



No.	X	Yield, %	Mp, °C	Analyses[a]	Antimalarial act.[b] 40	160	640	Toxicity deaths[d]
36		60	141–145	C, H, N	...	...	...	
37		67	96–99	C, H, N	0.1	0.1		5/5
38	NEt_2	70	84–85	C, H, N	0.3	0.3		5/5
39	NMe_2	71	148–149	C, H, N	0.5	0.5	0.9	2/5
40	$NCH(CH_3)_2$	65	139–142	C, H, N	0.5	0.9		5/5
41	NH_2	63	136–138	C, H, N	0.1	0.1		5/5
42	OH[c]	68	174–176		...	...	...	

[a] See Table I, footnote *a*. [b] See Table I, footnote *b*. [c] O. K. Behrens, J. Corse, D. E. Huff, R. G. Jones, Q. F. Soper, and C. W. Whitehead, *J. Biol. Chem.*, **175**, 771 (1948). [d] Deaths at the 640-mg/kg dose level.

The Reformatsky addition of methyl γ-bromocrotonate to the corresponding diaryl ketones gave 30–50% over-all yields of the diphenyl, fluorenyl, and bis(4-chlorophenyl)dienoic acids (**21**, **28**, **14**). Acrylic acid **42** (Table II) was obtained in 68% yield *via* the Reformatsky addition of α-bromoacetate. The three-step Reformatsky procedure involved the initial Reformatsky addition to give a diarylcarbinol, acid-catalyzed dehydration to a dienoic ester, and hydrolysis of the ester. These steps were generally conducted without purification of the intermediate alcohol and ester. This reaction sequence gave only 10% of bis(4-fluorophenyl)pentadienoic acid (**35**) despite considerable manipulation of the reaction conditions.

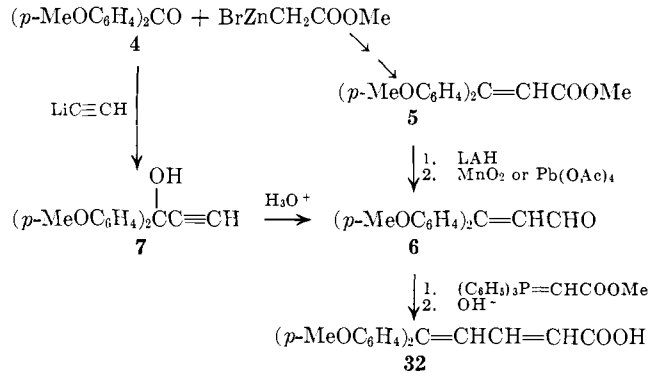
The addition of 4-phosphonocrotonic ester anion to bis(4-chlorophenyl) ketone gave a lower yield of the desired acid product (**14**) than was obtained by the Reformatsky sequence. This phosphono ester reagent was reported by Werbel, *et al.*, to give excellent results in the formation of monoaryldienoic acids from benzaldehydes.[18] The identity of the products obtained by the two procedures, along with uv absorption data, is considered as evidence in the present case against the possible occurrence of "abnormal" reactions on the α-carbon atom of the Reformatsky reagent.[19]

The use of the triphenylphosphorus ylide from methyl γ-bromocrotonate gave only very poor yields of dienoic esters in several cases when treated with diaryl ketones.

Preparation of bis(4-anisyl)pentadienoic acid (**32**) proved the most difficult of the dienoic acid syntheses reported. The Reformatsky procedure gave the desired acid in, at best, 5% yield although a variety of experimental conditions were employed. This reaction had been previously reported to fail entirely.[20] The ylide synthesis also failed in this case.

Two successful preparations of acid **32** are illustrated in Scheme I. Reduction and subsequent reoxidation of acrylic ester **5**, obtained from Reformatsky addition of methyl bromoacetate to ketone **4**, according to a reported general synthesis,[21] gave a very poor yield of the intermediate acrolein **6**. The reduction of the ester to the intermediate allyl alcohol even under mild conditions produced a major portion of hydrocarbon.

Scheme I



The intermediate acrolein could be converted to dienoic ester *via* an ylide reaction in good yield.

Bis(*p*-anisyl) ketone (**4**) was converted to the corresponding acrolein in much better yield by route **4** → **7** → **6**. Acetylenic carbinol **7**, prepared from the ketone and lithium acetylide, rearranged readily to **6** in ethanolic sulfuric acid. Aldehyde **6** was rather sensitive and was converted without purification to dienoic acid **32**. Before applying this acetylene–acrolein sequence to bis(*p*-anisyl) ketone, benzophenone was employed as a model. Results were equally good. Again, identity of products obtained by these two procedures with that from the Reformatsky method ruled against an "abnormal" Reformatsky reaction.

The amides have been conveniently prepared from the acids by the mixed anhydride method. No advantage was noted in either yield or ease of work-up of the reactions when isobutyl chloroformate was substituted for methyl chloroformate.

Biological Activity.—The compounds were assayed against lethal, blood-induced *P. berghei* infections in mice[17] as part of the Walter Reed Army Institute of Research malaria program. It can be seen from Tables I and II that the majority of the amides did not significantly increase the survival time of infected mice. The one interesting compound, 5,5-bis(4-chlorophenyl)-penta-2,3-dienoic acid diethylamide (**10**), shows both an appreciable activity and a distinct dose–response effect. None of the acrylic acid amides were active and all were toxic at the 640-mg/kg dose level.

Compounds **12**, **19**, **26**, and **36–40** did not inhibit the growth of *Staphylococcus albus*, *Escherichia coli*, *Serratia marcescens*, *Klebsiella aerobacter*, *Saccharomyces cerevis-*

(19) F. Bohlmann, *Chem. Ber.*, **90**, 1519 (1957).
(20) L. H. Klemm and G. M. Bower, *J. Org. Chem.*, **23**, 344 (1958).
(21) R. Heilman and R. Glenat, *Bull. Soc. Chim. France*, 1586 (1955).

iae, Penicillium notatum, or *Sporobolomyces salmoncolor*, when tested in a paper disk–agar diffusion assay. Compounds **12**, **13**, **19**, **20**, **26**, **27**, **37**–**39**, **40**, and **41** failed to significantly prolong the survival time of mice infected with lethal inocula of *Schistosoma mansoni* cercariae.[22]

Experimental Section

Most of the compounds prepared in this work have been analyzed by pmr using a Varian A-60A or HR-100 spectrometer. Typical uv spectra are given in the representative preparations below.

5,5-Bis(4-chlorophenyl)pentadienoic Acid (14).—A mixture of 20 g (0.08 mole) of 4,4′-dichlorobenzophenone and 5.9 g (0.09 mole) of activated 40-mesh Zn in 100 ml of C_6H_6 and 60 ml of Et_2O was refluxed under N_2. A portion of a solution of 14.3 g (0.08 mole) of methyl 4-bromocrotonate in 35 ml of C_6H_6 was added, and the reaction was initiated with the aid of a few drops of MeMgBr solution. The remainder of the RBr solution was then added. Reflux was maintained for 3 hr. The cooled mixture was treated with 60 ml of 2 *N* HOAc and stirred to yield a clear organic phase. The organic layer was washed (H_2O), twice with 5% $NaHCO_3$, and twice with H_2O) and then dried ($NaSO_4$). The solvent was removed *in vacuo*. The residue was heated on the steam bath for 35 min with 40 ml of 90% HCO_2H. The HCO_2H was removed *in vacuo*. The residue was taken up in 100 ml of boiling MeOH and treated with 4 g of KOH in a little MeOH–H_2O. The solution was refluxed for 1.5 hr while H_2O was gradually added (total 40 ml) to the cloud point. After 16 hr, water was added until no more solid precipitated. Filtration and recrystallization of the residue gave 8.8 g of starting ketone. The filtrate was acidified with concentrated H_2SO_4, and the collected solid was recrystallized from MeOH–H_2O to yield 13.6 g (53%) of the product: uv (EtOH), 243 mμ (ε 15,700), 258 (15,700), 320.5 (28,000).

5,5-Bis(4-chlorophenyl)penta-2,4-dienoic Acid Diethylamide (10).—Acid **14** (3 g, 0.0094 mole) was dissolved in 20 ml of dry THF. Et_3N (0.01 mole) was added at room temperature and the solution was stirred for 20 min. The solution was cooled to 0°, treated dropwise with 0.0103 mole of $ClCO_2Me$, and stirred 15 min more at 0°. Et_2NH (0.0105 mole) was added dropwise to the cold mixture, and the resulting solution was allowed to warm to 20° over 40 min. The resulting mixture was taken up in Et_2O–H_2O, and the Et_2O layer was washed (5% HCl, H_2O, twice with 5% $NaHCO_3$, and twice with H_2O). After drying (Na_2SO_4), the Et_2O was removed *in vacuo* and the solid residue was recrystallized from MeOH–H_2O to yield 2.12 g (60%) of product: uv (95% EtOH) 242 mμ (ε 18,200), 257 (18,700), 325 (35,900). Starting acid (30%) was recovered from the base washes.

4,4′-Dimethoxybenzophenone was prepared in 90% yield either by the reaction of anisic acid with anisol and PPA[23] or by the MeI–K_2CO_3 methylation of bis-4-hydroxybenzophenone.

Bis(4-anisyl)ethynylcarbinol was prepared by the reaction of 4,4′-dimethoxybenzophenone with lithium acetylide–ethylene diamine complex (Foote Mineral Co.) in DMF saturated with C_2H_2. The product was obtained in 90% yield, mp 89–91.5° (lit.[23] mp 93°).

3,3-Bis(4-anisyl)acrolein.—Bis(4-anisyl)ethynylcarbinol (15 g, 0.056 mole) was dissolved in 75 ml of EtOH under N_2. An instantaneous red-purple color developed when the first drops of 20% H_2SO_4 were added. The solution was an opaque dark brown color after a total of 3 ml of acid had been added. The reaction mixture was diluted with 20 ml of EtOH and stirred for 2 hr at room temperature. A copious black oil had precipitated at this time. The reaction mixture was taken up in Et_2O–H_2O. The Et_2O layer was washed (H_2O, 5% Na_2CO_3, H_2O). The Et_2O solution was dried and the Et_2O was removed *in vacuo* to yield the product as 15.4 g of a black oil: ir spectrum, λ_{max}^{film} 6.01 μ (C=O); tlc, silica gel ($CHCl_3$), R_f 0.35.

Several additional experiments gave poor yields and/or less pure product when less acid was used or the reaction period was shortened or the reactant concentration was decreased.

5,5-Bis(4-anisyl)penta-2,4-dienoic Acid.—To a 500-ml flask containing 100 ml of dry C_6H_6 and 7.27 g (0.0257 mole) of carbomethoxymethylenetriphenylphosphorane was added 5.0 g (0.0187 mole) of 3,3-bis(4-anisyl)acrolein in 30 ml of C_6H_6. The mixture was refluxed for 16 hr and the C_6H_6 was removed *in vacuo*. The resulting solid was dissolved in 30 ml of MeOH and treated with a mixture of 1.8 g of KOH in 20 ml of H_2O. The mixture was refluxed for 2 hr and cooled. A solid, neutral by-product was filtered off and the basic filtrate was acidified with 40% H_2SO_4. The resulting solid was filtered off and recrystallized from MeOH to yield 1.41 g (25%) of the desired acid, mp 186–189°.

(22) This test was performed under the auspices of the Walter Reed Army Institute of Research. We are grateful to Colonel William E. Rabe for providing this information.

(23) P. Cadiot, *Ann. Chim.* (Paris), **13**, 214 (1956).

Synthesis and Pharmacology of Some α-Oxy- and α-Hydroxy-1-benzyltetrahydroisoquinolines

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A series of α-oxy- and α-hydroxyl-1-benzyltetrahydroisoquinolines has been prepared and subjected to several pharmacological screening procedures. These include dose-range studies in mice and analgetic and antipyretic testing. A biological profile of these compounds derived from the results obtained is discussed.

The naturally occurring 1-benzyl and phthalideisoquinoline alkaloids exemplified by laudanosine (I) and narcotine (II) have often been the subject of chemical and pharmacological investigations.[1-3] More recently, synthetic 1-phenethyltetrahydroisoquinoline types such as III–V have undergone extensive chemical study[4] because of their interesting analgetic properties.[5]

In spite of the intense and continuing activity in this area, both the α-oxo- (VI) and α-hydroxy-1-benzyltetrahydroisoquinoline (VII) alkaloid types have not yet been thoroughly examined chemically or, more important, been adequately screened pharmacologically. Occasionally the former have been isolated as by-products in synthetic sequences;[6-8] there also are two

(1) A. Burger, *Alkaloids*, **4**, 29 (1954).

(2) J. Staněk, *ibid.*, **7**, 433 (1960), and references cited therein.

(3) H. G. Boit, "Ergebnisse der Alkaloid-Chemie bis 1960," Akademie-Verlag, Berlin, 1961.

(4) A. Brossi, H. Besendorf, L. A. Pirk, and A. Rheiner, Jr., in "Analgetics," G. deStevens, Ed., Academic Press Inc., New York, N. Y., 1965, p 281.

(5) A. Brossi, H. Besendorf, B. Pellmont, M. Walter, and O. Schneider, *Helv. Chim. Acta*, **43**, 1459 (1960).