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SYNTHESIS OF SOME (BENZOXAZOLYL-2)-ALKYL/ARALKYL SULPHIDES AND SULPHONES AS POTENTIAL PESTICIDES

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ABSTRACT

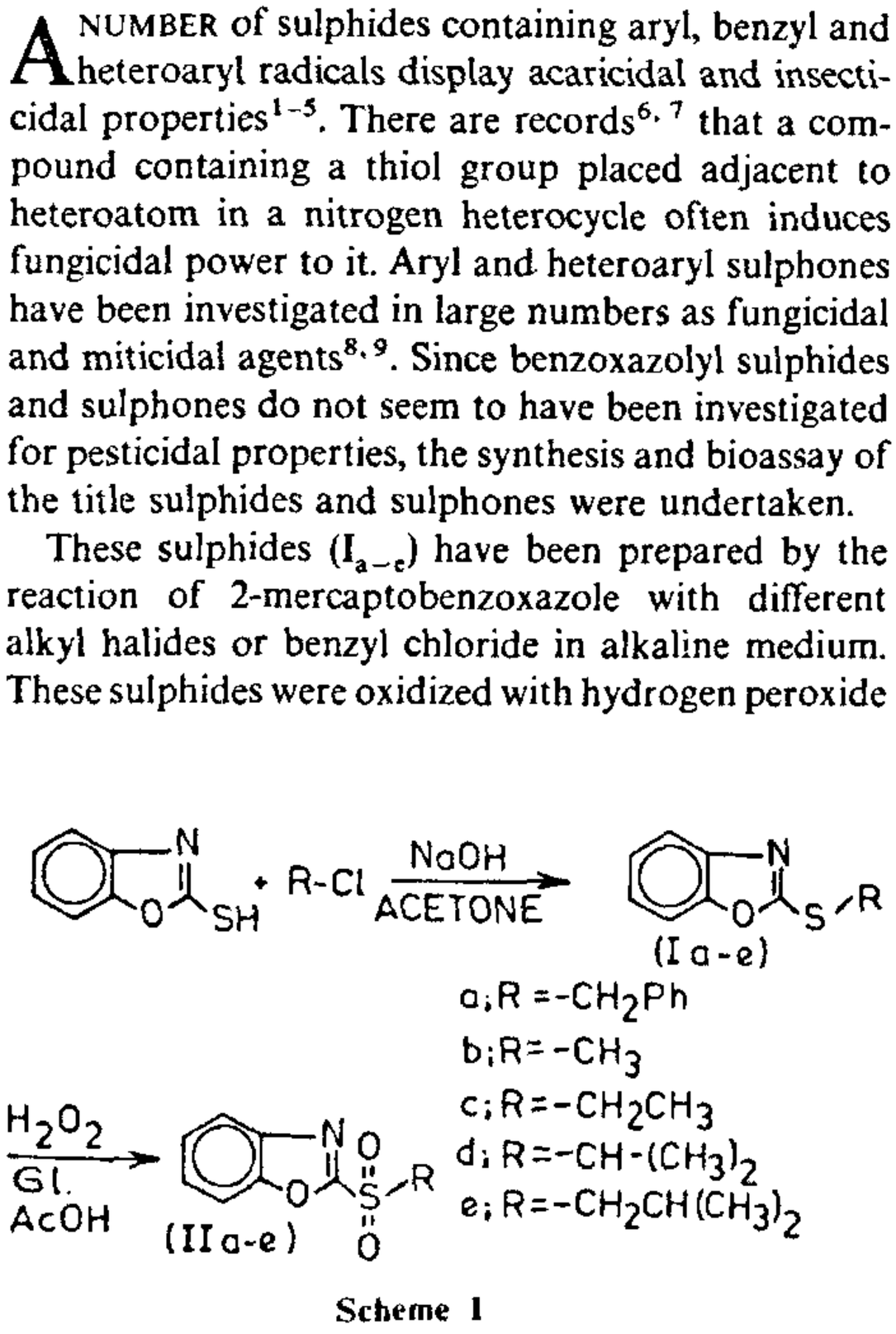
Several new (benzoxazolyl-2)-alkyl/aralkyl sulphides and their respective sulphones have been synthesised and screened for their fungicidal activity against *Alternaria tenuis* and *Helminthosporium oryzae*; and two of them have been tested for their molluscicidal activity against *Lymnea acuminata*. The sulphides synthesised herein are fluorescent compounds.

INTRODUCTION

A NUMBER of sulphides containing aryl, benzyl and heteroaryl radicals display acaricidal and insecticidal properties[1-5]. There are records[6, 7] that a compound containing a thiol group placed adjacent to heteroatom in a nitrogen heterocycle often induces fungicidal power to it. Aryl and heteroaryl sulphones have been investigated in large numbers as fungicidal and miticidal agents[8, 9]. Since benzoxazolyl sulphides and sulphones do not seem to have been investigated for pesticidal properties, the synthesis and bioassay of the title sulphides and sulphones were undertaken.

These sulphides (I_{a-e}) have been prepared by the reaction of 2-mercaptobenzoxazole with different alkyl halides or benzyl chloride in alkaline medium. These sulphides were oxidized with hydrogen peroxide in glacial acetic acid to yield the respective sulphones (II_{a-e}) (Scheme 1).

The authenticity of these compounds has been confirmed by their elemental analysis and IR spectral analyses. It is interesting to note that all sulphides synthesised emitted intense fluorescence.



Scheme 1

EXPERIMENTAL

IR spectra and elemental analyses (C, H, N and S) of the compounds are compatible with their structures. Melting points were taken in open capillaries and were uncorrected. IR spectra in KBr were recorded on Beckmann's spectrophotometer. TLC was performed on silica gel-G. 2-Mercaptobenzoxazole has been prepared following a method given in literature[10].

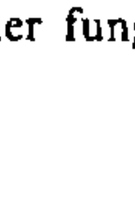
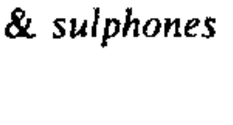
(Benzoxazolyl-2)-alkyl/aralkyl sulphides (I_{a-e}):

2-Mercaptobenzoxazole (1.1 M) dissolved in NaOH solution was refluxed with alkyl halides or benzyl chloride (1 M) for 4–6 hr. The reaction mixture was then poured into ice-water and the sulphide was taken into ether. After removal of the ether, the sulphides were obtained which were crystallised from ethanol. Yield 70–50%. I_a: m.p. 54° [IR spectrum reveals characteristic absorption frequencies at 1740 cm^{-1} (C–N=C stretching), 1280 cm^{-1} (C–O–C stretching), 720 cm^{-1} (C–S–C stretching) and 1590 cm^{-1},

1425 cm^{-1} (aromatic rings)]; I_b: gummy product; I_c: m p. 19 ; I_d: heavy liquid; I_e: heavy liquid.

The fluorescence spectrum of I_a was taken in UV light and it emits yellowish-green fluorescence at 5300 and 5600 Å. The rest of the sulphides were also fluorescent.

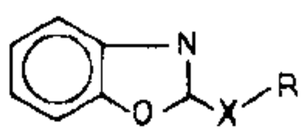
(Benzoxazolyl-2)-alkyl/aralkyl sulphones (II_{a-e}):

This was prepared by the method of Bombos[11]. (Benzoxazolyl-2)-alkyl/aralkyl sulphide (I) was refluxed with 30% H_2O_2 in glacial acetic acid for 2 hr. Excess acid was distilled off and the reaction mixture was poured in ice-water. The solid product obtained, was filtered, dried and crystallised from ethanol. Yield 65–50%, II_a: m.p. 128–30° [IR spectrum has significant peaks at 1750 cm^{-1} (5-membered imide cyclic ring). 1470 cm^{-1} ($-CH_2-$ bending). 1610 cm^{-1}, 1395 cm^{-1} (aromaticity), and 1300 cm^{-1} ( asymmetric stretching), 1250 cm^{-1} ( stretching)]; II_b: m.p. 135–6°; II_c: m.p. 150–1°, II_d: m p. 156°, II_e: m.p. 149–50°.

PESTICIDAL STUDIES

Fungicidal activity of the title compounds was evaluated by agar growth technique at 1000, 100 and 10 ppm concentrations against *Alternaria tenuis* and *Helminthosporium oryzae* and compared with Bavistin and Dithane M-45. The fungicidal data (table 1) clearly indicate the sulphones to be better fungicides than

Table 1 *Fungicidal activity of (Benzoxazolyl-2)-alkyl/aralkyl sulphides & sulphones*



Compound no.	X	Percentage inhibition after 96 hours: *Alternaria tenuis* at 1000 ppm	100 ppm	10 ppm	*Helminthosporium oryzae* at 1000 ppm	100 ppm	10 ppm
I_a	S	86 9	79 9	71.4	82.1	72.7	60.2
I_b	S	83.3	75 0	68.3	62.6	45 5	26.9
I_c	S	88.9	81 9	77.8	85.0	72.7	63.6
I_d	S	95.2	92.7	87 8	89.5	78.1	67.8
I_e	S	97.0	95.2	87.0	95.8	89.0	77.3
II_a	SO_2	88.9	82.4	76.1	84 3	78.9	66.7
II_b	SO_2	94.1	82.9	70.2	66.6	50.6	37.2
II_c	SO_2	87.3	78 9	71 4	87.0	80.5	58.0
II_d	SO_2	92.7	88.9	77.8	85.7	76.5	59.1
II_e	SO_2	95 5	90.1	77.8	94.1	85.7	63 6
Bavistin		98.9	96.6	94.2	94.2	91.2	84.3
Dithane M-45		99.2	98.5	96 7	96.7	93.2	85.7

Table 2 *Molluscicidal activity* No. of animals = 60

Compound no.	Doss mg/litre	Percentage mortality (M ± SE) at indicated time intervals after treatment: 24 hr	48 hr	96 hr
I_a	1	13.33 ± 2.31	20.0 ± 2.84	28.33 ± 3.37
	3	23.33 ± 2.32	31.67 ± 3.37	40.0 ± 4.01
	5	43.33 ± 5.26	50.0 ± 4.01	56.67 ± 5.43
II_b	1	18.33 ± 3.37	26 67 ± 3.36	36.67 ± 3.66
	3	28.33 ± 4.41	35.0 ± 3.75	46.67 ± 3.66
	5	51 67 ± 5.24	56.67 ± 3.36	71.67 ± 3.37

their sulphide counterparts at all concentrations against both the fungal species. All the compounds are quite toxic even at low concentrations except compound nos I_b and II_b. As the length of alkyl chain increases, the fungicidal activity of the compounds is also enhanced. These results reveal that all these sulphides and sulphones are very active and their activity appears to be governed by the nature of alkyl chain.

Molluscicidal activities of two of them have been tested against a mollusc–*Lymnea acuminata*. The results (table 2) clearly indicate that the activity is both dose-dependent and time-dependent. The compound no. II_a causes considerable mortality of snails.

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EFFECT OF AUTONOMIC DRUGS ON THE ISOLATED MELANOPHORES OF THE WALL LIZARD, *HEMIDACTYLUS FLAVIVIRIDIS*

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ABSTRACT

The nature of autonomic receptors present in the isolated skin melanophores of *Hemidactylus flaviviridis* was investigated by measuring the mean melanophore size index (MMSI). It was found that adrenaline and isoprenaline caused the dispersion of the melanophores, whereas phenylepherine caused aggregation. The melanophores of the wall lizard may have alpha as well as beta adrenergic receptors which mediate the aggregation and dispersion respectively. Acetylcholine, atropine and eserine, all caused melanophore aggregation in varying degree. 5-HT and histamine, both, caused a clear aggregation of melanophores.

INTRODUCTION

THE physiology, pharmacology and endocrinology of the melanophores of fish, amphibians and mammalian melanocytes have been extensively studied[1,2]. However, similar studies on the reptilian species are scanty[3]. In *Anolis carolinensis* the only extensively studied reptilian species, Hadley and Goldman[4] described a mosaic population of melanophores, having both alpha and beta adrenergic receptors, and others having only beta receptors. Unfortunately, pharmacological characterization of the receptors in melanophores of other species has not been done, needless to say, of any Indian species. Therefore, we have selected this common wall lizard