Further work will ascertain whether all 3 compounds operate through the same mechanism, and whether DNA is the target of the photosensitized reactions described in this report.

The ovicidal properties of natural products, both in dark reactions and in the presence of light, may be an important part of the plant-insect relationship. The results described here with *D. melanogaster* suggest the need for experiments in which egg survival should be scrutinized, particularly during the interaction between herbivorous insects and plants and their constituents in a natural environment.

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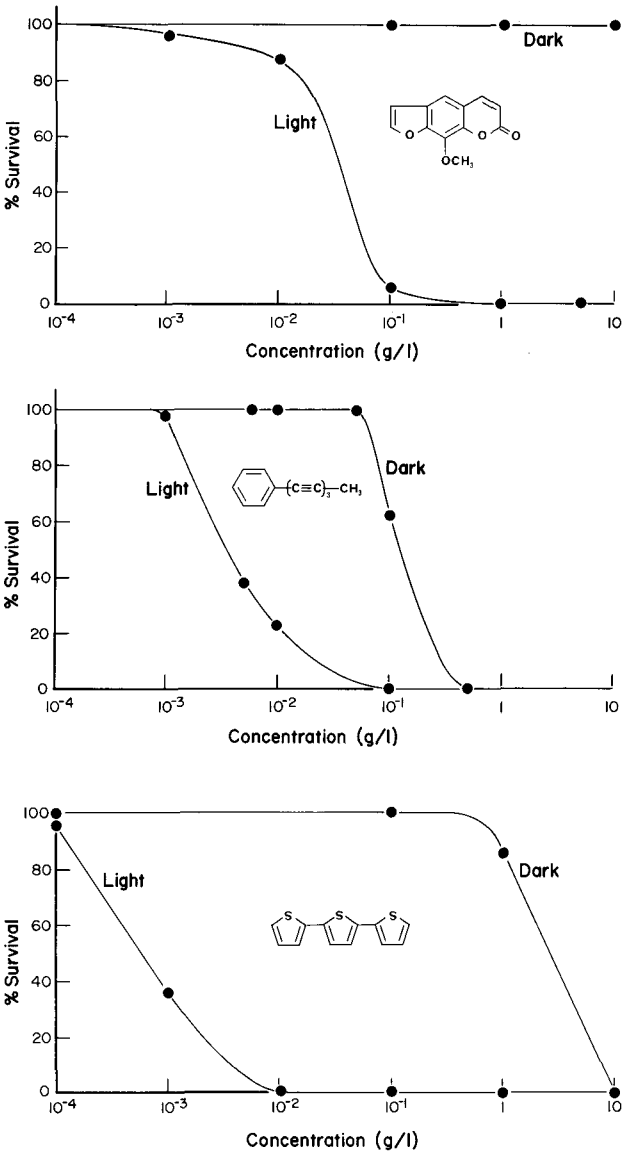

The ovicidal activity of PHT, αT, and 8-MOP. The survival of *D. melanogaster* is expressed as a function of the concentration of sensitizer used, plotted on a logarithmic scale. The concentrations in $\mu g/cm^2$ can be obtained by multiplying the values shown by 11.5.

Antifeedant activity of some ajugarin derivatives in three lepidopterous species

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Summary. The trans decalin unit of clerodendrin has antifeedant activity in larvae of *Pieris brassicae,* but not in larvae of *Spodoptera littoralis* and *S. exigua.* It is suggested that antifeedant activity of clerodanes for *Spodoptera* spp. is based on the combination of a furofuran ring and epoxy diacetate groups in the decalin unit.

Naturally occurring antifeedants may occasionally be rendered much more deterrent to insects by minor modifications of their molecular structure. Thus, the antifeedant activity of clerodendrin (fig., A), a bitter principle isolated from the Indian bhat tree *(Clerodendron infortunatum),* was raised 15-fold by transforming it into its methanol adduct[2]

when *Spodoptera litura* was used as experimental insect. On the basis of this observation Kojima et al.[3] suggested that the perhydrofuro-2,3-b-furan ring represents the active part of the clerodendrin molecule. However, none of 25 perhydrofuro-2,3-b-furan derivatives could reach the antifeedant level of the complete clerodendrin molecule[3]. Alternatively, the antifeedant activity could reside in the decalin unit of the clerodane molecule. This supposition is corroborated by the finding that ajugarin (fig., B), containing the same trans decalin unit, inhibits feeding in *Spodoptera exempta* and *S. littoralis*[4]. To test this hypothesis 2 stereoisomers of the decalin moiety (fig., C and D) as well as the cyclohexane derivative (E) have been synthesized[5] and were tested for their antifeedant activity. The present report describes the effect of C, D and E on food intake in an oligophagous lepidopterous species (*Pieris brassicae* L.) and 2 polyphagous insects (*Spodoptera littoralis* and *S. exigua*).

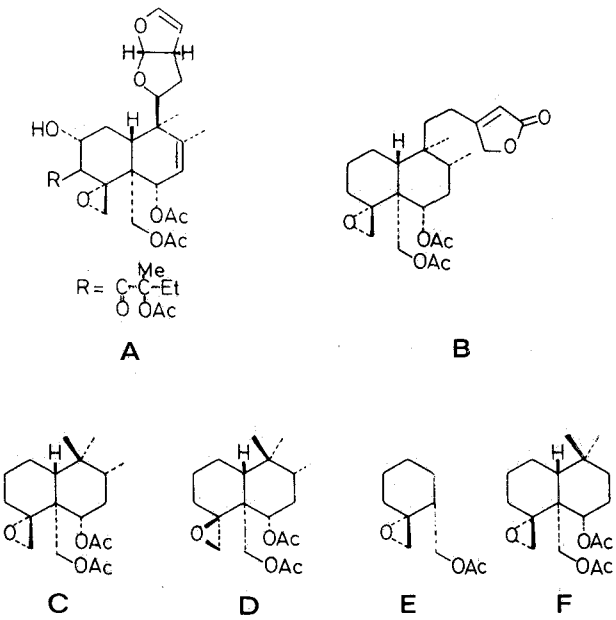
Newly moulted 5th instar larvae, raised on artificial diets, were tested on foamed polystyrene (p15) lamellae 0.7 mm thick[7]. The compounds to be tested were dissolved in ethanol 96%. Each solution was mixed with an equal volume of a 0.25 M sucrose in water solution, which served as a feeding stimulant. Each lamella, measuring 18 cm^2, was treated with 0.25 ml of the test solution, dried for 30 min at 50 °C (C-treated lamellae were dried for 24 h at room temperature) and offered to a single insect for 24 h. The insects were starved for 3 h before the beginning of the experiments. During the tests they had access to free water. After the tests the remains of the lamellae were dried again for 30 min (at 50 °C or room temperature) and weighed. Each compound was tested in a nonchoice situation and at various concentrations. Each concentration was tested on 20 larvae. Each control series involved 10 larvae.

As shown in the table compound C inhibits feeding in *P. brassicae*, but not in the 2 *Spodoptera* species, even when tested at high concentrations. *P. brassicae* responds also to compound D, albeit only at a 10-fold higher concentration. Compound E, tested only in *P. brassicae*, is inactive, despite the presence of the spiro epoxide and one acetate group. Interestingly compound C fails to inhibit feeding in *S. littoralis*, whereas ajugarin shows complete feeding inhibition in this species at 300 ppm[4]. *S. littoralis* is known to be inhibited by clerodendrin[6]. Obviously the presence of a butanolide containing side chain is required to extend the anti-feeding activity of the epoxy diacetate group.

The following conclusions may be drawn. *P. brassicae* larvae react to low concentration of compound C, since 95–100% feeding inhibition is obtained at concentrations where 1 molecule of antifeedant neutralizes the feeding stimulating effect of 370 molecules of sucrose. Furthermore, it is interesting to note that the trans decalin unit, lacking the methyl group at C-8 (see F), shows anti-feeding activity in *Locusta migratoria*. A 70% inhibition has been observed at a concentration of F of 100 ppm[8].

It is interesting to note that the oligophagous species *P. brassicae* is inhibited by 2 substances which do not affect the *Spodoptera* species tested. This would fit the idea that food specialists are more sensitive to antifeedants than generalists[9]. On the other hand, Kato et al.[2] have reported that clerodendrin inhibits the polyphagous larvae of *S. litura* from feeding, but not 3 other insect species which are monophagous. These observations stress the fact that different species may show different reactions to certain substances. This agrees with the finding that deterrent receptors in various insects show large interspecific variations[10].

In conclusion it may be envisaged that the antifeedant activity of the clerodanes to *Spodoptera* spp. is neither located exclusively in the furofuran unit of the molecule, as Kojima and Kato[3] conjectured, nor in the decalin moiety. The present results support the view that the furofuran ring and the epoxy diacetate groups of the trans decalin act synergistically, thus producing the striking antifeedant effect of clerodendrin in, for instance, *S. litura*.



A Clerodendrin A; *B* ajugarin I; *C* and *D* stereoisomers of decalin moiety; *E* cyclo hexane derivative, and *F* decalin derivative.

Antifeedant activities for compounds C, D and E

	Concentration (ppm)	1000	500	100	50	25
C	*P. brassicae*	++++	++++	++++	+++	++
	S. littoralis	0				
	S. exigua	0				
D	*P. brassicae*	++++	+++			
	S. littoralis	0				
E	*P. brassicae*		0			

Degree of antifeedant activity: ++++ = 100–95%; +++ = 95–75%; ++ = 75–50%; + = 50–25%; 0 = 25–0%.

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