TOXICITY OF PLANT OILS AND THEIR MAJOR CONSTITUENTS AGAINST STORED PRODUCT INSECTS

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Abstract

The fumigant toxicity of 26 essential oils extracted from various spices and herb plants and some of their major constituents was assessed for adult *Rhyzopertha dominica*, *Oryzaephilus surinamensis*, *Tribolium castaneum* and *Sitophilus oryzae*. Three groups of active materials were distinguished according to their activity against the insects tested. In addition, it was possible to select a compound ZP51 with a potency 5-10 fold greater, relative to all other compounds assayed. Total mortality was obtained at concentrations of 1.5-3µg/l air for all four insects tested. In addition, this compound was most active against *Sitophilus Oryzae*.

The biological activity of a large number of fatty acids and their derivatives was also studied, against *Callosobruchus chinensis*. It was possible to show a close correlation between the chain length of the acids and their biological activity. The compound DB211 was found to be the most potent of all the other compounds tested. Full protection was obtained at a concentration of 50-100mg/kg seeds. It seems that these compounds act as repellents by preventing the females from laying eggs on the treated seeds.

Introduction

At present, insect control in stored food products relies heavily upon the use of gaseous fumigants and residual insecticides. Both pose serious hazards to warm-blooded animals and the environment. Searching for alternative ways for insect control not involving insecticides, we found that fractionation of various edible plant seed extracts yielded products possessing insecticidal properties (Shaaya et al.,1976; Shaaya and Ikan, 1979). Fatty acids were found the possible chemical structure of part of the active products (Shaaya and Ikan, unpublished data). With the present state of the art, finding and developing new compounds which have the potential to replace the toxic fumigants and eliminate insecticide residues, while being less harmful, more suitable, simple and convenient to use, would be of significant benefit for the preservation of stored products.

The present study was conducted to investigate further the potential use of essential oils extracted from various spices and herb plants in Israel and some of their major constituents as fumigants, for the control of stored-product insect pests. In addition, the biological activity of potential fatty acids for the control of these insects was also studied.

Materials and Methods

The insects were bred at 27°C and 70% RH. Sitophilus oryzae and Rhyzopertha dominica were reared on wheat; Oryzaephilus surinamensis on ground wheat with the addition of glycerin and yeast; Tribolium castaneum on wheat flour; and Callosobruchus chinensis on chickpeas. The essential oils were obtained from herbs and spices by steam distillation (Marcus and Lichtenstein, 1979). Major oil constituents and fatty acids were purchased commercially except for those prepared in the laboratory. The fumigation chambers consisted of 3.4 liter glass flasks with a round bottom, closed with a glass stopper fitted with a hook. The test material was applied on a small square piece of filter paper which, together with the insect cages, was suspended, by means of the hook, in the fumigation chamber. To obtain even distribution of the essential oil during the treatment, a magnetic stirrer was placed on the bottom of each flask. Twenty insects of each species, aged 5-10 days, were placed in each of the four cages (4 cm in length and 1.5 cm in diam), which were perforated with small holes enabling penetration of the gas. Small amounts of ground wheat was placed in each cage. Treatments were carried out for 24 h. Percent mortality was recorded 24 h after treatment. The English name of the essential oil was used, and in cases where no English name was available - the Latin name was used. The bioassay of the fatty acids was conducted as follows: 50µl of ice cold ether solution containing the desired amount of the compound being tested was distributed evenly on the surface of each seed. The solvent was allowed to evaporate for 3 h in a stream of cold air. Five seeds were used for each test, which was infested with five males and females aged 0-4 h for a period of one day. Numbers of eggs laid, egg mortality and number of F_1 progeny were recorded.

Results

A. Fumigant toxicity of essential oils and their major constituents

In this study the fumigant toxicity of 26 essential oils and 11 of their principal components (Table 1) was tested against four major stored-product insect pests. S. oryzae, R. dominica, O. surinamensis and T. castaneum. At a concentration of 15-5μl/l air, only 11 oils and five of

Table 1- Essential oils and some of their major constituents which were tested as fumigants against four major stored -product insects.Group A was found to be very active, and group B had low or no activity.

Group A
The essenstial oils of oregano, basil, syrian marjoran, thyme, three lobed sage, sage, bay laurel, rosemary, lavender, anise, peppermint.The compounds linalool,α- terpineol, carvacrol, terpinen 4- ol, 1, 8- cineole

Group B
The essential oils of celery, savory, lemon grass, rue, clary sage, cumin, caraway, lemon , grapefruit , orange , coriander, *Salvia dominica*, fennel, camomile, *Artemisia arboresence*, parsley, *Artemisia judaica*. The compounds β- caryophyllen, peymene, γ-terpinene, salinene, myrcene .

their major components were found to be highly active (Table 1). Among the active materials three groups could be distinguished according to their activity. Group (i): The compounds terpinen 4-ol, 1,8-cineole, and the essential oils of three-lobed sage, sage, bay laurel, rosemary and lavender, were most active against R. dominica. All of them gave 100% mortality at a concentration of 15μl/l. O. surinamensis was less susceptible than R. dominica, and at a concentration of 15μl/l, only terpinen 4-ol, 1,8-cineole and rosemary gave 100% mortality (Fig. 1). Group (ii): The compounds linalool, α-terpineol, carvacrol and the

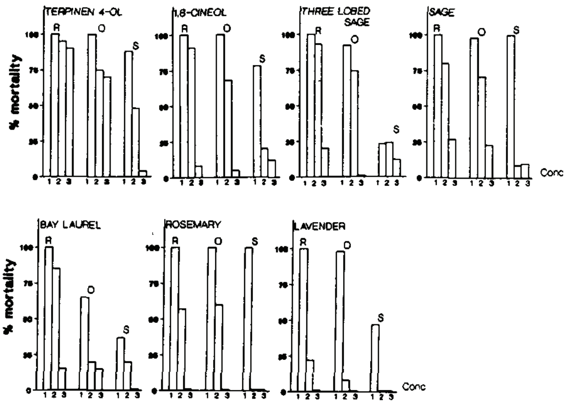

Fig. 1. Essential oils and compounds active against *R. dominica*. Percent mortality of *R*=*R. dominica*, *O*=*O. surinamensis*, *S*=*S. oryzae*, *T*=*T. castaneum*, using various concentrations (1=15,2=10,3=5µl/l air) of the test material. The results are the average of 5-10 replicates.

essential oils of oregano, basil, Syrian marjoram, and thyme were most active against O. surinamensis. The first five produced 100% mortality at a concentration of 15µl/l and the first two gave 100% mortality at a concentration of 10µl/l (Fig. 2). S. oryzae was much more resistant than O. surinamensis and R. dominica against all the essential oils

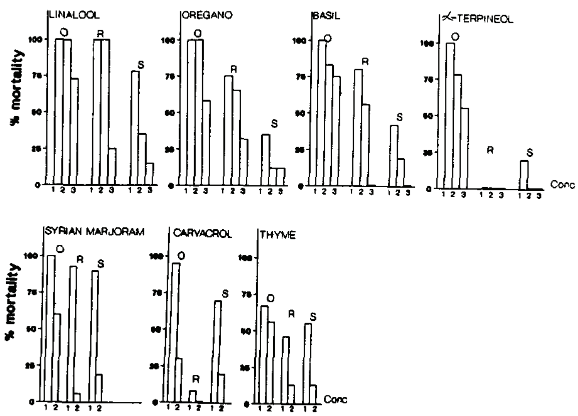

Fig. 2. Essential oils and compounds active against *O. surinamensis*. Percent mortality of *R*=*R. dominica*, *O*=*O. surinamensis*, *S*=*S. oryzae*, *T*=*T. castaneum*, using various concentrations (1=15,2=10,3=5µl/l air) of the test material.

and their major components tested. 100% mortality of S. oryzae was obtained only with sage oil at a concentration of 15µl/l (Figs. 1,2,3). Group (iii): The compound 1,8-cineole

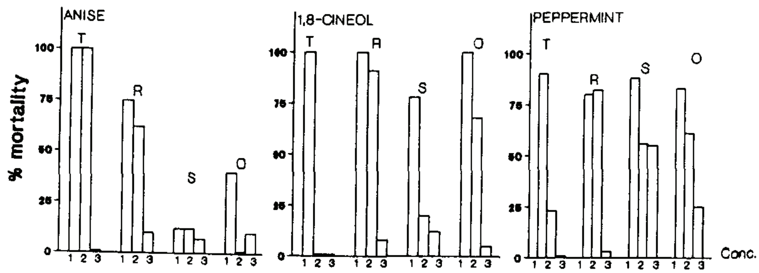

Fig. 3. Essential oils and compounds active against *T. castaneum. Persent* mortality of *R=R. dominica, O=O. surinamensis, S=S. oryzae, T=T. castaneum,* using various concentrations (1=15,2=10,3=5µl/l air) of the test material.

and the oils anise, and peppermint were the only active materials as fumigants against T. castaneum of all those tested. Anise and cineole gave 100% mortality at a concentration of 10 and 15µl/l, respectively.

By testing a large number of compounds and correlating the chemical structure of the molecules and their activity,

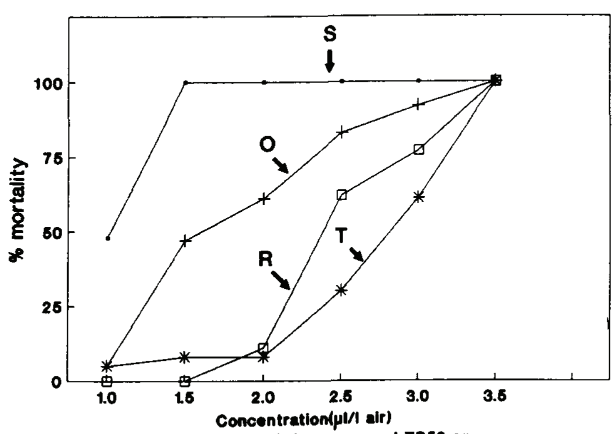

Fig. 4. The fumigant toxicity of the compound ZP52 on four stored product insects. The results are the average of 5-10 replicates, *S= S. oryzae, O= O. surinamensis, R= R. dominica, T= T. castanaum.*

it was possible to select a compound, ZP51, which had greater activity than all the essential oils and compounds tested (Fig. 4). A concentration of 3µl/l was enough to achieve 100% mortality of the four test insects. In addition, this compound was very potent against S. oryzae, with 100% mortality obtained by 1.5µl/l (Fig. 4).

From the other oils of low activity Artemisia judaica, parsley, Salvia dominica and clary sage were found to have activity mainly against O. surinamensis. At a concentration of 15µl/l, 80, 75, 55, 52 and 45% mortality, respectively was recorded. On the other hand, fennel oil, and the compounds pinene and γ-terpinene, were active only against R. dominica. At a concentration of 15µl/l, 83, 56 and 49% mortality, respectively, was recorded (data not shown).

B. Studies with fatty acids

Figure 5 shows the results obtained by applying a concentration of 2.5g/kg seeds of fatty acid C5-C18 carbon

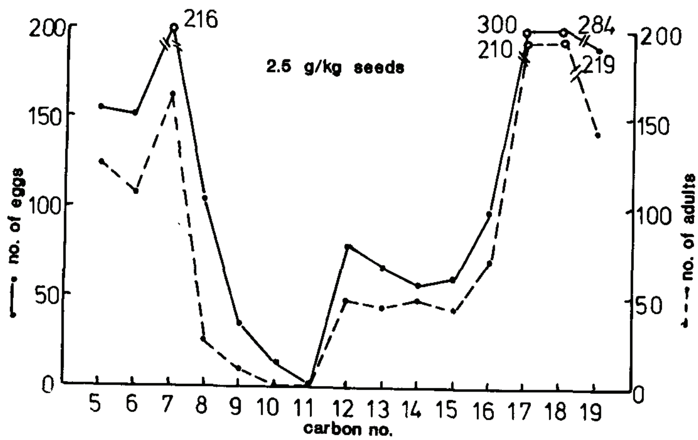

Fig.5 The activity of straight chain fatty acids from C5-C18 at a concentraion of 2.5 mg/kg seeds on *Callosobruchus chinensis*. The results are the average of three replicates each 5 tests.

atom. It was found that C10 (capric) and C11 (undecanoic) acids were the most effective in preventing oviposition of C. chinensis. The activity was remarkably decreased by lower (C5-C7) and higher (C17-C18) acids. The C12-C16 showed weak activity. To gain insight into the specificity of these molecules, first we tested the activity of C6-C12 methyl esters of the fatty acids. These compounds were found to possess no activity (Shaaya and Ikan, 1979). Second, we tested the activity of n-aliphatic acetates C6-C18 carbon atoms, prepared from aliphatic alcohols and acetic

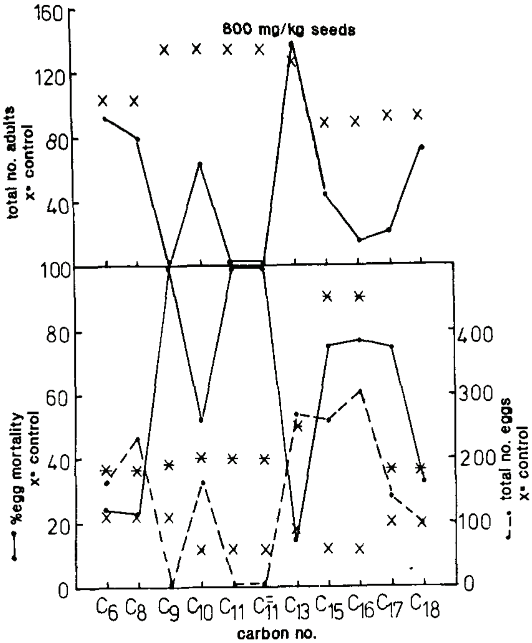

Fig.6 The activity of C6-C18 n-aliphatic acitates at a concentration of 800 mg/kg seeds on Callosobruchus chinensis. The results are the average of three replicates each 5 tests.

anhydride, against C. chinensis. Figure 6 shows clearly that C9, C11 and C11* at a concentration of 800mg/kg seeds were highly active, no eggs or very few eggs were laid and no adults were developed. A comparison between the activity of C11 and C11 acetate show that the former is twice as potent as the latter (Fig. 7). Respective concentrations of 200mg/kg seeds compared with 400mg/kg were needed to prevent adult development (Fig. 7).

By screening a large number of compounds, it was possible to select a potent compound, DB211, which prevented oviposition at a concentration of 100mg/kg seeds. At a lower concentration of 50mg/kg seeds, only 35 eggs were laid and 10 adults developed compared with 210 eggs and 110 adults found in the control seeds (Fig. 8).

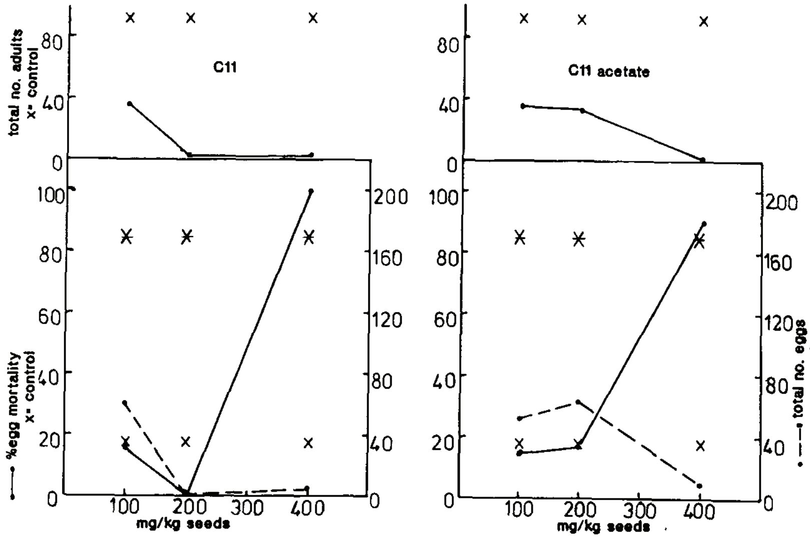

Fig.7 The activity of the straight chain fatty acids C11 and C11 acetate at various concentrations on *Callosobruchus chinensis.*

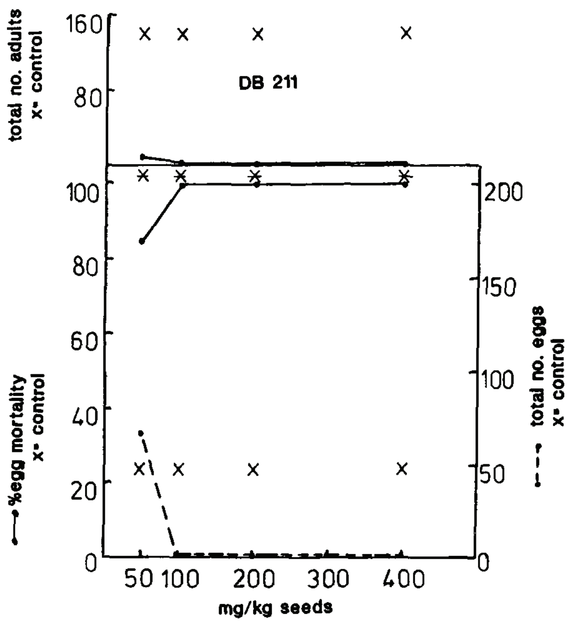

Fig.8 The activity of a potent compound DB 211 at various concentrations on *Callosobruchus chinensis.*

Discussion and conclusions

Three groups were selected from the 26 essential oils and some of their major constituents as active fumigants against four major stored product insects: Group (i), most active against O. surinamensis, group (ii) against R. dominica and group (iii) against T. castaneum. In addition, compound ZP51 was most active against S. oryzae. The activity of ZP51 was 5-10 fold more potent than any essential oil or major constituents tested. A concentration of 1.5µl/l was enough to achieve 100% mortality against S. oryzae and 3µl/l against the other three insects. It should be noted that a concentration of 20-30mg/l of methyl bromide is recommended for control of stored product insects in the field.

By screening a large number of fatty acids and their derivatives, we were able to demonstrate a close correlation between the chain length of the acids and their biological activity. These compounds seem to act as repellents by preventing the female from laying eggs on the tested seeds. DB211 was the most active of all the compounds tested, and full protection was obtained at a concentration of 50-100mg/kg seeds.

The existence of naturally occurring insecticidal plant components has been known for centuries. However, relatively few of these compounds are actually used in crop protection today (Marcus and Lichtenstein, 1979). Increasing problems concerning the use of modern synthetic insecticides, such as persistence of residues, resistance and damage to the environment and human health have produced interest in naturally occurring compounds. Biologically active compounds of food plants are assumed to be environmentally more acceptable and less hazardous to humans than synthetic chemicals. The results presented in this study suggest that some of the compounds tested might be efficient fumigants or repellents and also could be integrated with other pest management procedures for the control of stored product insects.

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TOXICITE DE PLANTES A HUILE ET DE LEURS CONSTITUANTS MAJEURS SUR LES INSECTES DES DENREES STOCKEES

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RESUME

La toxicité par effet de tension de vapeur de 26 extraits d'huiles essentielles provenant d'herbes et d'épices, ainsi que celles de quelques uns de leurs composés majeurs ont été testées sur des adultes de *Rhyzopertha dominica, Oryzeaphilus surinamensis, Tribolium castaneum* et *Sitophilus orysae*. Trois groupes de principes actifs ont été définis en fonction de leur efficacité sur les insectes testés. En outre, il a été possible de sélectionner un composé (ZP 51) possédant une puissance 4 à 10 fois supérieure aux autres composés essayés. La mortalité complète a été obtenue à des concentrations de 1,5 à 3 µg/l pour ces 4 espèces. En outre, ce composé a été le plus actif contre *Sitophilus orysae*.

L'activité biologique d'un grand nombre d'acides gras et de leurs dérivés a aussi été étudiée contre *C. chinensis*. Il a été possible de montrer une forte corrélation entre la longueur de la chaîne des acides et leur activité biologique. Le composé DB 211 s'est révélé le plus puissant de tous les produits testés. Une protection totale a été obtenue à une concentration de 50 à 100 mg/kg de grain. Il semble que ces composés agissent comme répulsifs en empêchant les femelles de pondre leurs oeufs sur les graines traitées et n'ont pas d'effet par contact.