Chemical defense in the seaweed *Ochtodes secundiramea* (Montagne) Howe (Rhodophyta): effects of its monoterpenoid components upon diverse coral-reef herbivores

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Abstract: Many species of tropical red algae are known to produce halogenated secondary metabolites that have been hypothesized to function in chemical defense. The tropical fleshy red alga *Ochtodes secundiramea* (Montagne) Howe is a typical example known to produce the halogenated monoterpenoids ochtodene and chondrocole A. In this study, the susceptibility of *O. secundiramea* to grazing by herbivorous fishes was examined by offering an array of 11 red, green, and brown seaweeds to herbivores in field assays in the Grenadine Islands of the Caribbean. *O. secundiramea* was among the least susceptible to grazers, with no transplants being lost to herbivory during our assay. To examine the possible rôles of the monoterpenoids in reducing herbivory, the metabolites ochtodene and chondrocole C were isolated and purified by high-performance liquid chromatography (HPLC). These monoterpenoids were tested in field assays for their feeding deterrent effects toward herbivorous fishes in the Grenadine Islands and on Guam in the tropical western Pacific. In both sets of experiments, ochtodene was an effective feeding deterrent and chondrocole C was not. These compounds, and a nonpolar mixture (more than three compounds) of halogenated monoterpenes from *Ochtodes*, were also tested for their feeding deterrent effects toward herbivorous amphipods collected in the Grenadine Islands. Only the unresolved monoterpene mixture was an effective feeding deterrent toward the amphipods. Herbivory on coral reefs is intense and the diversity of herbivore types is great. It appears that the variety of secondary metabolites synthesized by seaweeds such as *O. secundiramea* may be effective in deterring different reef herbivores.

Key words: Amphipod; Chemical defense; Coral-reef fish; Herbivory; *Ochtodes secundiramea*; Plant–herbivore interaction; Seaweed

INTRODUCTION

Although 100s of unique secondary metabolites have been isolated from marine algae, the ecological importance of these compounds is not well-studied. Tropical algae, in particular, have been shown to produce a diverse array of structurally unique and

biologically active compounds (Norris & Fenical, 1982; Faulkner, 1984). Many species
of red algae (Rhodophyta) have the distinctive ability to synthesize organic halogen-
containing compounds that incorporate halides from sea water. Many novel and
complex halogenated terpenoids are known from tropical red algae, including species
of the genera *Laurencia, Desmia, Plocamium,* and *Ochtodes* (Fenical, 1975; Faulkner,
1984).

The importance of plant secondary metabolites in reducing herbivory in terrestrial
communities is well-documented and generally accepted as one of the most effective
means of herbivore defense (Rosenthal & Janzen, 1979; Crawley, 1983; Coley *et al.,*
1985). Herbivory is known to be intense on tropical reefs (Randall 1961, 1974; Hatcher
& Larkum, 1983; Hay, 1981a,b, 1984, 1985; Lewis, 1985, 1986; Carpenter, 1986) and,
therefore, constitutes a potentially important evolutionary pressure selecting for algal
chemical defenses. For these reasons, most natural products isolated from tropical algae
have been presumed to function as chemical defenses against herbivores (Ogden &
Lobel, 1978; Lobel & Ogden, 1981; Norris & Fenical, 1982; Hay, 1984). Only recently,
however, have these assumptions been evaluated with ecologically relevant experiments
(Targett *et al.,* 1986; Paul, 1987; Hay *et al.,* in press a; Paul & Van Alstyne, in press).

In this study, we examined the susceptibility of a fleshy red alga, *Ochtodes
secundiramea* (Montagne) Howe, to grazing by the diverse group of herbivorous fishes
occurring on reefs near where the plants were collected in the Grenadine Islands of the
Caribbean. The susceptibility of this alga was compared with that of 10 other species
of red, green, and brown algae. *O. secundiramea* is known to produce the halogenated
monoterpenoids ochtodene and chondrocole A (McConnell & Fenical, 1978) (Fig. 1);
it lacks calcification, toughness, or other obvious morphological defenses. Because we
observed very low susceptibility of this alga to grazing by coral-reef fishes, we asked the
following questions. (1) Are the halogenated terpenoids produced by *O. secundiramea*
effective feeding deterrents toward the diverse assemblages of herbivorous fishes that
encounter this alga on Caribbean reefs? (2) Are these compounds effective defenses
toward tropical mesograzers (Hay *et al.,* in press b) such as amphipods? (3) Is the
feeding deterrence of these compounds against tropical herbivores a general property?
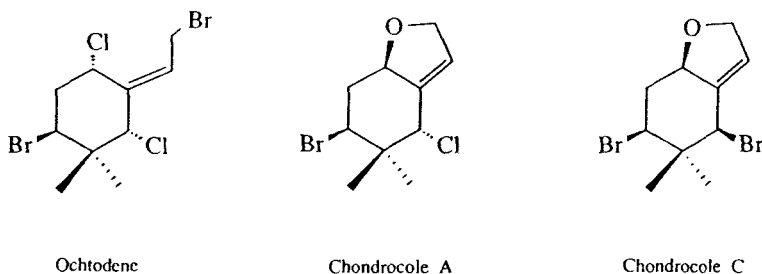That is, are these compounds effective defenses toward natural populations of

Ochtodene   Chondrocole A   Chondrocole C

Fig. 1. Chemical structures of the halogenated monoterpenoids isolated from *Ochtodes secundiramea.*

herbivorous fishes in the western Pacific that may not encounter *Ochtodes* but may encounter related seaweeds such as *Desmia* that produce similar halogenated monoter-penoids?

STUDY SITES

COLLECTION

Ochtodes secundiramea was collected subtidally (7 m depth) at Point Dunkerque near St. Anne on the Island of Martinique in the Caribbean on 19 July 1986. The habitat was characterized by flat carbonate rock with a heavy growth of many species of alga and relatively low numbers of herbivorous fishes compared with other nearby reef habitats.

FIELD EXPERIMENTS

Experiments examining the susceptibility of *O. secundiramea* to grazing were perform-ed at a depth of 3–6 m on a coral reef off Canouan Island in the Grenadine Islands. Numerous parrotfishes, including *Scarus vetula, S. croicensis, S. taeniopterus, Sparisoma aurofrenatum, S. viride*, and *S. chrysopterum*, as well as the surgeonfishes *Acanthurus coeruleus, A. bahianus*, and *A. chirurgus* (Randall, 1983) were common on this reef.

Experiments examining the feeding deterrent effects of purified metabolites were performed near Crab Louse Cay, a small island near Petite St. Vincent in the Grenadine Islands. This reef habitat (5–10 m depth) was characterized by many of the same species of herbivorous fishes as the Canouan Island site. The parrotfish *Sparisoma aurofrenatum* was particularly abundant in this habitat and was observed to graze during our experiments. This species and other parrotfishes probably accounted for most of the grazing during these experiments.

Experiments were also conducted on the island of Guam in the tropical western Pacific. The study site was a shallow patch reef inside Cocos Lagoon (3–6 m depth). Many species of herbivorous fishes were observed to graze during the experiments and common species included *Scarus sordidas, S. schlegeli, Acanthurus nigrofuscus, Naso literatus, Siganus argenteus, S. spinus, Zebrasoma flavescens*, and *Z. scopas* (Amesbury & Myers, 1982).

METHODS

CHEMICAL EXTRACTION AND STRUCTURAL DETERMINATION

Ochtodes secundiramea was homogenized in a blender and extracted with a 2:1 mixture of methylene chloride and methanol. The solvents were evaporated under reduced pressure to yield a dark viscous oil (0.8 g, $\approx 5.5\%$ of algal dry mass). Thin-layer chromatographic analysis (TLC) of the extract showed the presence of major amounts

of nonpolar secondary metabolites. The metabolites were isolated by high-performance liquid chromatography (HPLC) of the nonpolar mixture with 15% ethyl acetate in isooctane to yield three fractions in order of increasing polarity: a halogenated monoterpenoid mixture (more than three compounds) (15% of organic extract, 0.8% of algal dry mass), ochtodene (27% of organic extract, 1.5% of dry mass), and chondrocole C (26% of organic extract, 1.4% of dry mass) (Fig. 1). These metabolites were identified by high-resolution proton and ^{13}C nuclear magnetic resonance (NMR) spectroscopy by comparing spectral data for our isolates with published values. Details of the isolation and structural determination of these compounds have been reported (Burreson *et al.*, 1975b; McConnell & Fenical, 1978). The halogenated monoterpenoid mixture, which was eluted as a single peak by HPLC, was later illustrated to be composed of at least three undefined monoterpenoids by proton NMR.

ALGAL SUSCEPTIBILITY ASSAYS

Susceptibility of 11 species of algae to grazing by herbivorous fishes was examined by weaving small (3–4 cm long) pieces of the different thalli, at 5-cm intervals, into a 0.5-m length of three-strand, 7-mm thick, polypropylene line that was then fastened to the reef slope (n = 23 ropes). The 11 species of alga were haphazardly arranged on the lines and should have been equally apparent and available to herbivorous fishes encountering the lines. Grazing of the thalli on the lines was allowed to continue for 3.5 h until there was clear distinction between the most and least susceptible plants. At the end of the experiment, each species on each line was recorded as either still present or absent. Small portions of consumed thalli often remained between the strands of the ropes, but these portions almost always showed the crescent-shaped bite marks of herbivorous fishes. We did not observe the fishes during the assays; thus, it is possible that some algal material was bitten and rejected instead of being completely consumed. Similar methods have been used in other studies of algal susceptibility to grazers (Hay, 1984; Paul & Hay, 1986).

FEEDING DETERRENT ASSAYS

For grazing assays in the Grenadine Islands, blades of the palatable seagrass *Thalassia testudinum* Banks ex König were used as the bioassay food. *T. testudinum* is eaten by many species of Caribbean fishes, especially the parrotfishes (Randall, 1967). On Guam, we used the palatable green alga *Enteromorpha clathrata* (Roth) J. Agardh, which is eaten by almost all herbivorous fishes on Guam. For both sets of experiments, the seagrass blades or *Enteromorpha* were coated with a solution of the purified halomonoterpenoid in diethyl ether. The final metabolite concentration on the *Thalassia* blades was 1.2% of the dry mass for ochtodene and 2% of the dry mass for chondrocole C. These concentrations approximate the natural concentrations of 1.5 and 1.4% of the dry mass of *Ochtodes* for each major metabolite, respectively. The *Enteromorpha* was coated with each metabolite at 1% of the algal dry mass, which is slightly below natural

concentrations, because of a limited amount of the purified metabolites. Dry mass of the wet *Thalassia* and *Enteromorpha* was calculated by a previously determined wet mass : dry mass ratio. Natural concentrations of ochtodene and chondrocole C were determined during the extraction and isolation process. Control *Thalassia* blades and *Enteromorpha* were coated only with diethyl ether.

For both sets of experiments, four pieces of treated *Thalassia* or *Enteromorpha* were woven into a 0.5-m length of polypropylene line. The same number of control pieces were woven into another line and these lines were placed on the reef as a matched pair (0.25–0.5 m apart). Replicate pairs were placed several metres apart. The duration of the assays varied in the two habitats. Coated *Thalassia* remained on the reef for 3 h at Crab Louse Cay in the Grenadines. All ropes ($n = 21$ for ochtodene, $n = 18$ for chondrocole C) were then recovered and grazing was measured as the decrease in length of the *Thalassia* blades (Hay *et al.*, in press a). Coated *Enteromorpha* remained on the reef only 10–15 min at Cocos Lagoon sites since grazing occurred very rapidly; ropes were collected when at least half of the algae was eaten ($n = 12$ for ochtodene, $n = 6$ for chondrocole C). Grazing was measured as the number of pieces of algae consumed (Paul, 1987; Paul & Van Alstyne, in press). Results of both Caribbean and Pacific assays were analyzed by a paired-sample t test.

Since the halomonoterpenoids are nonpolar and lipid soluble, they adhere to the surface of the *Thalassia* and *Enteromorpha* after the ether evaporates and can then be placed in sea water for the feeding experiments. McConnell *et al.* (1982), Targett *et al.* (1986), Paul (1987), and Hay *et al.* (in press a) used similar methods and found that these kinds of metabolites were not rapidly lost to sea water. Following each of the field assays, all remaining *Thalassia* and *Enteromorpha* that had been treated with a metabolite were extracted in ether and the extract analyzed by thin-layer chromatography (TLC). The TLC result was then compared with the TLC result of the pure metabolite to determine if the compound was still present or had decomposed to some other compound. These TLC comparisons are not quantitative but, in all cases, showed no decomposition of the metabolites, although some material may have been lost to sea water.

AMPHIPOD GRAZING ASSAYS

Amphipod grazing assays were conducted on board the *R. V. Columbus Iselin* with natural mixed-species assemblages of herbivorous amphipods collected in the Grenadine Islands. The amphipods were collected by shaking the animals out of intertidal seaweeds (primarily *Dictyopteris delicatula* Lamouroux and *Sargassum hystrix* J. Agardh) collected on Mayreau and Canouan Islands. The animals were held in buckets of aerated sea water on board the ship until used in experiments. The actual individuals used in the assays were not identified; however, subsamples (totalling 955 individuals) taken from the same holding bucket had the following composition: *Hyale macrodactyla* Stabbing (84%); *Elasmopus pectenicrus* Bate (6%); Isopoda (3%); *Tethygenia longleyi*

Shoemaker (2%); *Ampithoe marcuzzi* Ruffo (2%); *Audulla chelifera* Chevreux (1%); and *Ampithoe* sp., *Stenothoe minuta* Holmes, *Lembos* sp., *Lysianassa alba* Holmes (<1%). *Hyale* is a common genus worldwide and is known to be herbivorous (McBane & Croker, 1983; J.D. Thomas, pers. comm.). Given its numerical dominance in our samples, *H. macrodactyla* was probably responsible for most or all of the grazing in these experiments. However, because these experiments used the natural mixed-species assemblages of amphipods found on plants in the field, they are comparable to our field tests of how compounds affected grazing by the guilds of reef fishes that consume *Thalassia* and *Enteromorpha*.

For amphipod grazing assays, halomonoterpenoids were coated onto the green alga *Ulva* sp., which was readily eaten by these grazers. Small thin 10-mm disks were punched out of the *Ulva* thallus and metabolites were coated onto these *Ulva* disks at 1% of plant dry mass for the halogenated monoterpenoid mixture, 1.2% for ochtodene, and 2% for chondrocole C. The monoterpenoid-coated disks were offered to the amphipods with solvent-coated (diethyl ether) control disks for 36–48 h. The amount of each disk consumed by the amphipods was determined by measuring the surface area lost to grazing using a dissecting microscope. A point-intercept method was used to calculate the amounts of treated and control *Ulva* that were eaten. Results were analyzed by a paired-sample *t* test.
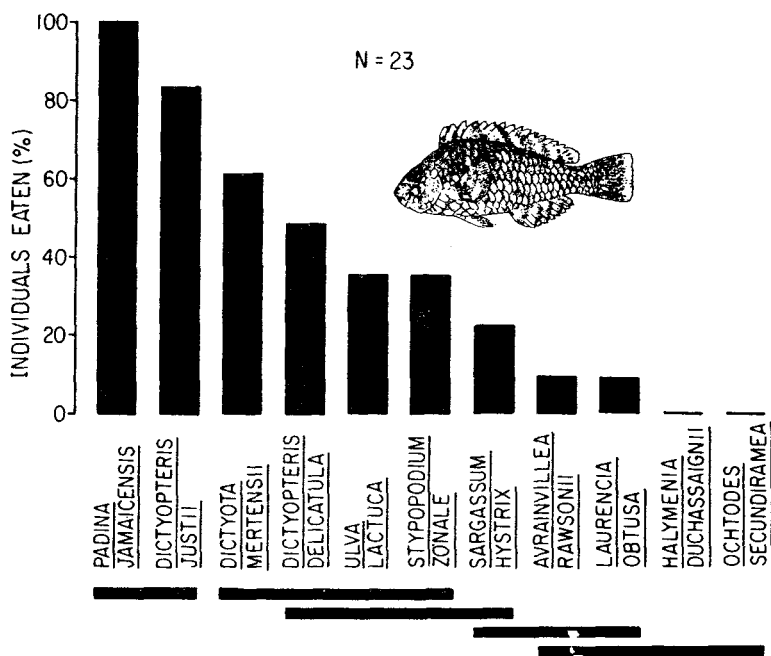

Fig. 2. Percentage of individuals completely consumed when transplanted into a reef habitat with high rates of herbivory for 3.5 h. Bars beneath histograms indicate species that were not significantly different ($P > 0.05$). Significant differences were evaluated by contingency table analysis supplemented with Fisher's exact test for comparisons with some unacceptably small cell sizes.

RESULTS

The susceptibilities of *Ochtodes secundiramea* and 10 other species of tropical algae to herbivorous fishes in the Grenadine Islands are shown in Fig. 2. *Ochtodes* was among the least preferred of these seaweeds which included several other species known to produce secondary metabolites. No transplants of *Ochtodes* were lost to grazing fishes during our assays (Fig. 2).

The feeding deterrent effects of the *Ochtodes* metabolites toward natural populations of herbivorous fishes and amphipods in the Caribbean are shown in Fig. 3. Ochtodene
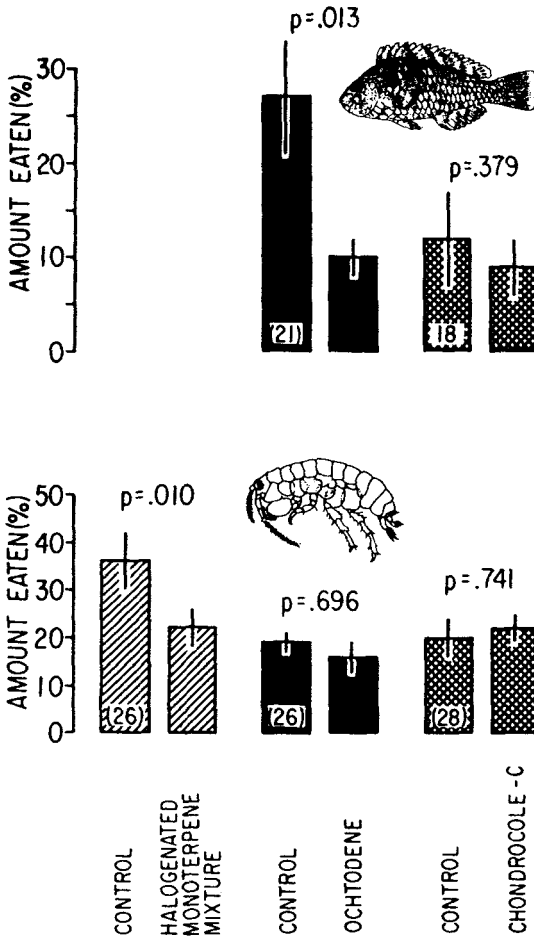
Fig. 3. The effects of ochtodene and chondrocole C on feeding by herbivorous reef fishes in the Grenadine Islands, and the effects of ochtodene, chondrocole C, and a halogenated monoterpene mixture on feeding by herbivorous amphipods in the laboratory. Vertical bars through each histogram show ± 1 SE. Numbers inside each histogram indicate the number of paired samples used in the assays. Significance values are from the paired-sample *t* test.

was an effective feeding deterrent toward herbivorous fishes ($P = 0.013$, grazing reduced 63% relative to controls); however, chondrocole C caused no significant effect. The halogenated monoterpenoid mixture was tested only against the amphipods because of the lesser amount of material we isolated. This mixture was the only significant ($P = 0.010$) feeding deterrent toward the amphipods and decreased grazing by 45% relative to controls. Both ochtodene, which was an effective deterrent toward fishes, and chondrocole C were not effective feeding deterrents toward the amphipods.

Similar results were observed for the herbivorous fishes on Guam (Fig. 4). Ochtodene was an effective feeding deterrent ($P = 0.007$) and reduced grazing by 57% relative to controls. Chondrocole C had no significant effect on grazing.
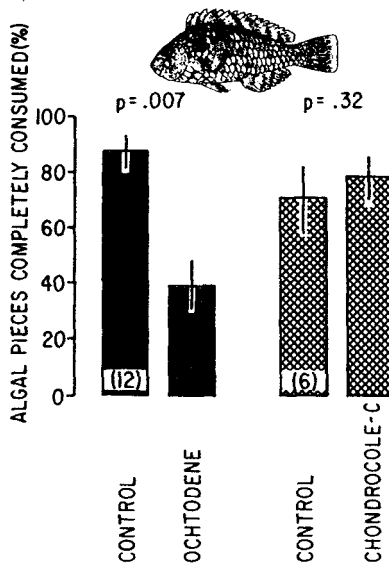

Fig. 4. The effects of ochtodene and chondrocole C on feeding by herbivorous reef fishes on Guam. Symbols and statistical tests are as in Fig. 3.

DISCUSSION

Chemical defense appears to explain the low susceptibility of *Ochtodes secundiramea* to grazing by herbivorous fishes in our assays in the Grenadine Islands of the Caribbean. *Ochtodes* ranked with *Laurencia obtusa* (Hudson) Lamouroux and *Avrainvillea rawsonii* (Dickie) Howe, which are also uncalcified and known to produce secondary metabolites (Paul & Hay, 1986), as among the least preferred seaweeds we tested. *Stypopodium zonale* (Lamouroux) Papenfuss, *Avrainvillea rawsonii*, *Laurencia obtusa*, and many species of *Dictyota* and *Dictyopteris* are all known to produce bioactive secondary metabolites that have been hypothesized to function in chemical defense (Norris &

Fenical, 1982; Hay, 1984; Paul & Hay, 1986). The metabolites stypotriol, from *Stypopodium zonale*, elatol, from *Laurencia obtusa*, and pachydictyol A, from several species of *Dictyota*, have all been demonstrated to function as effective feeding deterrents toward populations of herbivorous fishes in the Caribbean (Hay *et al.*, in press a). However, *Dictyota mertensii* (Martius) Kuetzing, *Stypopodium zonale*, and two species of *Dictyopteris* were significantly more susceptible to grazing fishes than *Ochtodes secundiramea* in our field assays (Fig. 2).

O. secundiramea lacks obvious morphological defenses and is known to produce secondary metabolites. McConnell & Fenical (1978) studied *O. secundiramea* from Belize and isolated the major metabolite ochtodene, minor amounts of ochtodiol, and smaller amounts of a compound, chondrocole A, isolated originally from *Desmia* (syn. *Chondrococcus*) *hornemanni* Lyngb. (Burreson *et al.*, 1975a) (Fig. 1). In the population of *Ochthodes secundiramea* from Martinique, we isolated ochtodene and chondrocole C (also previously known from *Desmia hornemanni*, Burreson *et al.*, 1975b) as the major metabolites (Fig. 1), along with a mixture of at least three nonpolar halogenated monoterpenoids as minor metabolites. This variation in the production of secondary metabolites by different populations of algae has been documented for several other species of seaweeds (Fenical, 1976; Mynderse & Faulkner, 1978; Gerwick *et al.*, 1985; Paul & Fenical, 1986).

Only one of the two major metabolites, ochtodene, was a significant feeding deterrent toward fishes in our experiments. Chondrocole C was not an effective deterrent toward fishes or amphipods (Fig. 3), although other defensive rôles for this compound may be possible. The unresolved mixture of minor halomonoterpenoids was not tested toward fishes in the Caribbean or on Guam.

The deterrent effects of the *Ochtodes* metabolites were similar for both the Caribbean and Pacific assemblages (Fig. 4) of herbivorous fishes. In both cases, ochtodene was deterrent and chondrocole C was not. Different species of fishes occur in these areas, and different species grazed during our assays. Based on our observations, parrotfishes were the major grazers on the *Thalassia* used in our Caribbean assays. A wider variety of fishes including parrotfishes, surgeonfishes, and rabbitfishes, grazed on the *Entero-morpha* which was used in assays on Guam. This shows that, on average, ochtodene deters grazing by a variety of herbivorous fishes and suggests that these metabolites may have similar physiological effects toward different fish species.

Ochtodes does not grow on Guam and is not reported from any of the islands of Micronesia (Tsuda & Wray, 1977). *Ochtodes crockeri*, from the Galapagos Islands, has been chemically investigated and found to produce halogenated cyclic monoterpenoids similar to ochtodene and chondrocoles A and C (Paul *et al.*, 1980). The related alga *Desmia hornemanni* is common on Guam and throughout the Pacific, and is known to produce chrondocoles A and C and related halogenated monoterpenoids in Hawaii and Japan (Ichikawa *et al.*, 1974; Burreson *et al.*, 1975a,b). In recent studies of *D. horne-manni* from Guam, ochtodene was found to be a major secondary metabolite and several acyclic monoterpenes were minor metabolites (Paul, work in progress). Populations of

Desmia on Guam do not produce chondrocoles A and C. *Desmia* is susceptible to grazing by natural populations of herbivorous fishes in some habitats on Guam, but not in reef habitats where the alga is abundant (Paul & Potter, unpubl. data). *Desmia* is rapidly and completely eaten by the rabbitfish *Siganus spinus* (Paul & Nelson, in prep.) but is avoided by the surgeonfish *Zebrasoma flavescens*. Extracts of *Desmia* and the isolated metabolite ochtodene are also deterrent toward *Zebrasoma flavescens* (Wylie & Paul, unpubl. data). The secondary metabolites produced by *Desmia* appear to function as feeding deterrents toward some herbivorous fishes on Guam but not others. Ochtodene has been tested, but the other compounds produced by *Desmia* have not yet been isolated and tested on Guam.

The effects of the *Ochtodes* compounds on the herbivorous amphipods were different from their effects on fish. Ochtodene was not deterrent toward the amphipods, and only the monoterpenoid mixture was an effective amphipod feeding deterrent. The pattern of amphipods often being resistant to compounds that deter fishes has been documented previously and suggests that amphipods may be ecologically similar to terrestrial insects in their generally greater ability to breach plant chemical defenses (Hay *et al.*, in press b). Since the monoterpenoid mixture was composed of at least three related compounds, we could not determine whether some or all of these compounds were the deterrents.

It appears that *Ochtodes* produces a variety of related secondary metabolites as defenses toward diverse coral reef herbivores. The responses of the herbivorous fishes and amphipods were different. No defensive rôle of chondrocole C, a major metabolite produced by this alga, against fishes or amphipods was found. This compound may function in chemical defense against other species of herbivores (e.g., sea urchins or other fishes), toward pathogens or fouling organisms, or may not have a defensive rôle.

This study illustrates a clear relationship between low susceptibility to herbivores and the production of secondary metabolites by a tropical seaweed. At least one halogenated metabolite was an effective feeding deterrent toward fishes, and a mixture of other metabolites was effective toward amphipods. However, not all of the secondary metabolites produced by *Ochtodes* could be shown to be effective feeding deterrents. This result emphasizes the importance of experimentally evaluating predictions about the defensive rôle of algal secondary metabolites. Not all secondary metabolites appear to function as feeding deterrents, and compounds that may be effective toward one species of herbivore may be ineffective toward other species (Paul, 1987; Hay *et al.*, in press a, b). Lubchenco & Gaines (1981) have noted that any particular defense is likely to become less effective as a plant is subjected to attack by an increasing diversity of herbivore types. Given the high diversity of tropical reef herbivores, this prediction may explain why so many tropical seaweeds are known to produce a variety of secondary metabolites, why chemical variation exists among individuals and populations within many species of seaweeds, and why many tropical algae utilized multiple defenses such as secondary chemistry and calcification (Norris & Fenical, 1982; Faulkner, 1984; Hay, 1984; Paul & Hay, 1986; Paul & Fenical, 1986; Paul & Van Alstyne, in press). Thus, on tropical reefs where herbivores are numerous and diverse, many seaweeds such as

Ochtodes produce a variety of secondary metabolites which may be effective toward some, but not necessarily all, species of herbivores.

ACKNOWLEDGEMENTS

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