The Chemical Control of Powdery Mildew by Fumigant Redistribution

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ABSTRACT

An experimental fungicide, 4-phenyl-4-(3-pyridyl)-3-oxatricyclo[4.2.1.$0^{2,5}$] nonane (U.C. 23271), controls several foliar and soil-borne plant diseases and is an unusually effective eradicant of bean powdery mildew. Soil, stem, and leaf applications of U.C. 23271 resulted in insufficient systemic movement to eradicate mildew. A unique redistribution of U.C. 23271 resulting in mildew eradication was discovered. One hundred µg of U.C. 23271 localized on a small area of leaf surface completely eradicated powdery mildew over the entire leaf surface by fumigation. The long residual life of U.C. 23271, coupled with its excellent mildewcidal properties, suggests that its fumigant characteristics could have practical importance. Two structural analogues, diphenyl-3-pyridylmethane and diphenyl-3-pyridylmethanol, were mildew eradicants equivalent to U.C. 23271 in spray applications. The former was equivalent to U.C. 23271 as a fumigant and the latter was inactive. Thus, while the property of fumigant redistribution may have a practical significance, it alone does not explain the mildew eradication which was observed. Phytopathology 60:1062-1064.

An unusually effective eradication of bean powdery mildew, *Erysiphe polygoni* DC., resulted from spray applications of 4-phenyl-4-(3-pyridyl)-3-oxatricyclo[4.2.1.$0^{2,5}$] nonane (U.C. 23271), an experimental fungicide which controls several foliar and soil-borne plant diseases (3, 4). Disease eradication at very low chemical dosages (ED_{50} = 1 ppm) is rare and indicates an efficient route to contact with the pathogen. These investigations explore several alternative pathways by which U.C. 23271 may distribute itself in or on the plant and their effect on the fungicidal efficiency of this compound.

MATERIALS AND METHODS.—Bean plants, *Phaseolus vulgaris* L. 'Tendergreen', in small clay pots were inoculated when the primary leaves were half expanded by dusting them with conidia from older, heavily mildewed plants. Inoculations were made on both the upper and lower surfaces of the primary leaves. One day after inoculation, the plants were sprayed, drenched, or otherwise treated with chemicals. Thus, our data are based upon the eradication of powdery mildew disease, since the pathogen had infected the host. Inoculated plants were incubated in a growth room at 20 C and 50% relative humidity for 7 days. Similar untreated, inoculated plants were used as controls. When the primary leaves of the untreated plants were covered with mildew in 7 days, the degree of infection on the primary leaves of treated plants was evaluated as follows: 1 = no control; 2 = many, but discrete spots; 3 = 4 to 10 spots/leaf; 4 = 1 to 3 spots/leaf; 5 = no mildew. Our experience over many years has shown this evaluation system to be highly reliable and reproducible (5). For spray and drench applications, aqueous formulations were prepared by dissolving the chemical in acetone containing 0.1% Triton X-155 (a nonionic alkyl aryl polyether alcohol surfactant, Rohm & Haas Co.) for serial dilution with water.

RESULTS.—Although our results are based upon the eradication of powdery mildew 24 hr after inoculation, we were interested in determining the full potential of U.C. 23271 for the control of established infections. To this end, heavily mildewed plants of the type used for making inoculations were sprayed with U.C. 23271 at rates of 500, 1,000, and 2,000 ppm, and control plants were sprayed with the aqueous formulation without this mildewcide. Conidia from these plants were used to inoculate young, healthy plants 1 hr and 72 hr after this spray treatment. The plants which were inoculated after 1 hr became infected with mildew and did not differ from the untreated controls at any dosage of U.C. 23271. After 72 hr, no infection was produced from any of the inoculum from plants sprayed with U.C. 23271.

When the upper surfaces of primary bean leaves were sprayed using a Beltsville sprayer, U.C. 23271 eradicated the mildew on both upper and lower leaf surfaces; Dinocap was effective only on the upper leaf surface (Table 1). In another experiment, one of two infected primary leaves on each plant was covered with aluminum foil during the spray application to prevent contact with the chemical. After spraying, the aluminum foil was removed. Mildew eradication was effected on sprayed and unsprayed leaves by U.C. 23271 (Table 1). In order to effect complete control on one of the paired leaves, slightly higher dosages had to be applied to the adjacent leaf. In these experiments, U.C. 23271 moved either systemically and/or by fumigation to provide disease control on unsprayed leaf surfaces.

Spot applications of U.C. 23271 dissolved in acetone (50 mg/ml) were made to the upper surface of the leaf tip or the upper surface of the leaf base of primary leaves or to the stem directly below the cotyledons at concentrations from 0.01 mg (0.2 µliters) to 5 mg (100 µliters). The solution was also applied to the surface of 22 mm^2 glass microscope cover slides and allowed to dry. The cover slides with treated surfaces up were then taped to the upper tips of inoculated primary leaves. These applications of U.C. 23271 resulted in effective eradication of powdery mildew and a dosage response was observed (Table 2). This was the first clear evidence of fumigant redistribution by U.C. 23271 resulting in effective eradication of disease symptoms (Fig. 1). The application to the leaf base was slightly more effective than to the leaf tip, but

TABLE 1. Bean powdery mildew eradication on upper and lower leaf surfaces effected by spray applications of chemical to the upper surface only and average eradication on both leaf surfaces of leaves covered during the spray application[a]

Treatment	Leaf surface evaluated	Rate ppm				
		1,000	500	100	20	4
U.C. 23271	Upper	5	5	5	5	5
	Lower	5	5	5	2	2
	Both (avg)[c]	4	3	3	1	
Dinocap[b]	Upper	5	5	5	5	3
	Lower	1	1	1		
	Both (avg)[c]	1	1	1		

[a] Evaluation: 5 = complete control; 1 = no control.
[b] 2-(1-Methylheptyl)-4, 6-dinitrophenyl crotonate.
[c] Leaf covered with aluminum foil during the spraying of the plant.

control resulted from both treatments. The stem application was effective also, but required higher dosages. The area of the leaf to which the chemical was applied showed mild phytotoxic symptoms in the form of white, desiccated tissue. The dosage response from the cover slides was equivalent to that from the spot applications to the leaves (Tables 2, 3). In another experiment, eradication of mildew was observed on unsprayed plants which were surrounded by, but not touching plants sprayed with U.C. 23271 at 500 ppm. These experiments established the effectiveness of fumigant redistribution to eradication but did not eliminate the possibility of some systemic redistribution.

Systemic movement as an effective means of redistribution of U.C. 23271 was ruled out by the following observations. When the upper leaf surfaces were sprayed with a 1,000 ppm of U.C. 23271, mildew developed *only* under pieces of cellophane tape stuck to the lower leaf surfaces. Aqueous soil drenches (50 ml of 1,000 ppm of U.C. 23271/pot) and immersion of stems cut just above the soil line in aqueous solutions containing 1,000 ppm of U.C. 23271 did not eradicate mildew. Therefore, systemic movement of U.C. 23271 through leaves or from the roots or stems is insufficient to effect disease control.

The long residual effect and the excellent activity for bean powdery mildew eradication of U.C. 23271 was emphasized when cover slides containing 5, 10, and 20 μg of U.C. 23271 were taped to leaves at 7-day intervals up to 28 days after being prepared. The 28-day-old slides controlled mildew as well as the freshly prepared slides, indicating that volatilization or decomposition of U.C. 23271 under ambient conditions is very slow. This is in agreement with a study of physical properties which showed that U.C. 23271 sublimed slowly without decomposing under reduced pressure at 120 C (R. L. Hinman, *personal communication*).

TABLE 2. Bean powdery mildew eradication resulting from spot applications of U.C. 23271 dissolved in acetone[a]

Application spot	Rate mg (upper surface evaluated)					
	5	1	0.5	0.1	0.05	0.01
Leaf base	5	5	5	5	5	4
Leaf tip	5	5	5	4	4	4
Stem by cotyledon	4	3	3	1	2	1

[a] Evaluation: 5 = complete control; 1 = no control.

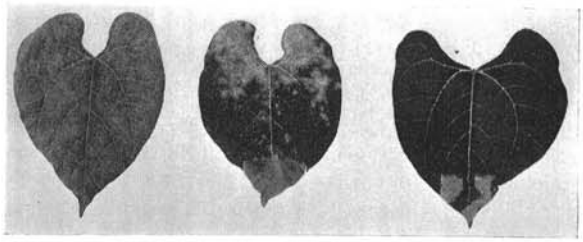
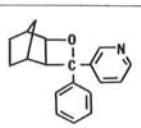
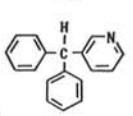
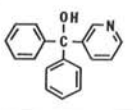


Fig. 1. Powdery mildew eradication from primary bean leaves 7 days after the spot application of (left to right) 0, 1, and 10 μg of U.C. 23271 to the glass cover slides taped to the upper leaf tips.

Two active analogues of U.C. 23271 were evaluated using the cover slide fumigant test. One analogue, diphenyl-3-pyridylmethane, was equivalent to U.C. 23271 in fumigant activity (Table 3). The second analogue, diphenyl-3-pyridylmethanol, was inactive as a fumigant. Although fumigant activity is a property of U.C. 23271 and diphenyl-3-pyridylmethane, it is not a property of diphenyl-3-pyridylmethanol, which is equivalent to the other two compounds in effectiveness when applied as a spray. Thus, fumigant activity alone does not explain the excellent mildewcidal effectiveness observed with these compounds.

DISCUSSION.—Although U.C. 23271 is a highly effective bean powdery mildew eradicant, our results demonstrate it is not systemic in plants in amounts sufficient to be fungitoxic. It does have a unique property in redistribution by fumigation. The importance of chemical redistribution to disease control has been recognized

TABLE 3. Comparative efficacy of three chemical analogues for bean powdery mildew eradication via two application techniques: (i) spraying the leaves (spray); (ii) applying the chemical to glass cover slides taped to the upper leaf tips (fumigate)

U.C. No.	Structure	Spray concn (ppm)[a]		Fumigate concn (μg)[b]	
		ED_{50}[c]	ED_{95}	ED_{50}	ED_{95}
23271 (MP 132-134)		1	4	13	100
Diphenyl-3-pyridyl-methane (MP 77-80)		2	24	14	100
Diphenyl-3-pyridyl-methanol (MP 115-116)		1	2	>500	

[a] Concn of solution sprayed on the leaves.
[b] Amount of chemical placed upon the glass cover slides.
[c] ED = effective dosage (50% control, 95% control).

by other researchers (1, 2). These other investigations, however, have centered largely on the redistribution of chemicals in water, either dew or rain, and to a lesser extent on systemic movement within the plant. Fumigation considerations have dealt largely with soil and greenhouse applications and with the loss of protective action or lack of persistence of certain foliage fungicides. Thus, it appears that the fumigant redistribution observed with U.C. 23271 is unique. The low volatility and high intrinsic toxicity for the bean powdery mildew fungus are indicative of the practical potential of this compound as a persistent type of fumigant.

When the fumigant property of U.C. 23271 was recognized, we were interested in determining its relationship to the eradication of bean powdery mildew. The use of the two analogues, diphenyl-3-pyridylmethane and diphenyl-3-pyridylmethanol, which were equivalent to U.C. 23271 in bean powdery mildew eradicant activity when applied in a spray was helpful in this respect. Since diphenyl-3-pyridylmethanol proved inactive as a fumigant, fumigant redistribution does not explain the excellent mildewcidal activity of U.C. 23271. The laboratory spray applications were precise, and so it is logical to assume that the initial distribution of chemical by this application technique was very good and the resulting data reflected inherent toxicity of a chemical.

Thus, while fumigant redistribution is interesting, unique, and perhaps important under practical situations, it is not restricted to U.C. 23271 alone, and does not explain the excellent mildewcidal activity of U.C. 23271.

LITERATURE CITED

1. Burchfield, H. P. 1960. Performance of fungicides on plants and in soil—physical, chemical and biological considerations, p. 477-520. *In* J. G. Horsfall & A. E. Dimond, [ed.] Plant Pathology, An advanced treatise, Vol. III. Academic Press, N. Y.
2. Courshee, R. J. 1967. Application and use of foliar fungicides, p. 239-286. *In* D. C. Torgeson [ed.] Fungicides, an advanced treatise. Vol. I. Academic Press, N. Y.
3. Sousa, A. A., H. W. Spurr, Jr., & D. R. Arnold. 1968. A new fungicide: 4-phenyl-4-(3-pyridyl)-3-oxatricyclo [4.2.1.$0^{2,5}$] nonane (U.C. 23271). Phytopathology 58:108-110.
4. Spurr, H. W., Jr., & E. L. Chancey. 1968. Unique bean powdery mildew eradication by fumigant redistribution of 4-phenyl-4-(3-pyridyl)-3-oxatricyclo [4.2.1.$0^{2,5}$] nonane (U.C. 23271). Phytopathology 58:1068-1069 (Abstr.).
5. Torgeson, D. C. 1961. The fungicidal activity of (aminoalkyl) pyrenes. Contrib. Boyce Thompson Inst. 21:21-26.