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The citrus reentry problem: Research on its causes and effects, and approaches to its minimization

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THE CITRUS REENTRY PROBLEM: RESEARCH ON ITS CAUSES AND EFFECTS, AND APPROACHES TO ITS MINIMIZATION

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The citrus reentry problem: Research on its causes and effects, and approaches to its minimization

Preface

That residues of pesticide and other contaminants in the total environment are of concern to everyone everywhere is attested by the reception accorded previous volumes of "Residue Reviews" and by the gratifying enthusiasm, sincerity, and efforts shown by all the individuals from whom manuscripts have been solicited. Despite much propaganda to the contrary, there can never be any serious questions that pest-control chemicals and food-additive chemicals are essential to adequate food production, manufacture, marketing, and storage, yet without continuing surveillance and intelligent control some of those that persist in our foodstuffs could at times conceivably endanger the public health, Ensuring safety-in-use of these many chemicals is a dynamic challenge, for established ones are continually being displaced by newly developed ones more acceptable to food technologists, pharmacologists, toxicologists, and changing pest-control requirements in progressive food-producing economies.

These matters are of genuine concern to increasing numbers of governmental agencies and legislative bodies around the world, for some of these chemicals have resulted in a few mishaps from improper use. Adequate safety-in-use evaluations of any of these chemicals persisting into our foodstuffs are not simple matters, and they incorporate the considered judgments of many individuals highly trained in a variety of complex biological, chemical, food technological, medical, pharmacological, and toxicological disciplines.

It is hoped that "Residue Reviews" will continue to serve as an integrating factor both in focusing attention upon those many residue matters requiring further attention and in collating for variously trained readers present knowledge in specific important areas of residue and related endeavors involved with other chemical contaminants in the total environment. The contents of this and previous volumes of "Residue Reviews" illustrate these objectives. Since manuscripts are published in the order in which they are received in final form, it may seem that some important aspects of residue analytical chemistry, biochemistry, human and animal medicine, legislation, pharmacology, physiology, regulation, and toxicology are being neglected; to the contrary, these apparent omissions are recognized, and some pertinent manuscripts are in preparation. However, the field is so large and the interests in it are so varied that the editors and the Advisory Board earnestly solicit suggestions of topics and authors to help make this international book-series even more useful and informative.

"Residue Reviews" attempts to provide concise, critical reviews of timely advances, philosophy, and significant areas of accomplished or needed endeavor in the total field of residues of these and other foreign chemicals in any segment of the environment. These reviews are either general or specific, but properly they may lie in the domains of analytical chemistry and its methodology, biochemistry, human and animal medicine, legislation, pharmacology, physiology, regulation, and toxicology; certain affairs in the realm of food technology concerned specifically with pesticide and other food-additive problems are also appropriate subject matter. The justification for the preparation of any review for this book-series is that it deals with some aspects of the many real problems arising from the presence of any "foreign" chemicals in our surroundings. Thus, manuscripts may encompass those matters, in any country, which are involved in allowing pesticide and other plant-protecting chemicals to be used safely in producing, storing, and shipping crops. Added plant or animal pest-control chemicals or their metabolites that may persist into meat and other edible animal products (milk and milk products, eggs, etc.) are also residues and are within this scope. The so-called food additives (substances deliberately added to foods for flavor, odor, appearance, etc., as well as those inadvertently added during manufacture, packaging, distribution, storage, etc.) are also considered suitable review material. In addition, contaminant chemicals added in any manner to air, water, soil or plant or animal life are within this purview and these objectives.

Manuscripts are normally contributed by invitation but suggested topics are welcome. Preliminary communication with the editors is necessary before volunteered reviews are submitted in manuscript form.

Department of Entomology
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March 1, 1977

F.A.G.
J.D.G.

Foreword

Worldwide concern in scientific, industrial, and governmental communities over traces of toxic chemicals in foodstuffs and in both abiotic and biotic environments has justified the present triumvirate of specialized publications in this field: comprehensive reviews, rapidly published progress reports, and archival documentations. These three publications are integrated and scheduled to provide in international communication the coherency essential for nonduplicative and current progress in a field as dynamic and complex as environmental contamination and toxicology. Until now there has been no journal or other publication series reserved exclusively for the diversified literature on "toxic" chemicals in our foods, our feeds, our geographical surroundings, our domestic animals, our wildlife, and ourselves. Around the world immense efforts and many talents have been mobilized to technical and other evaluations of natures, locales, magnitudes, fates, and toxicology of the persisting residues of these chemicals loosed upon the world. Among the sequelae of this broad new emphasis has been an inescapable need for an articlated set of authoritative publications where one could expect to find the latest important world literature produced by this emerging area of science together with documentation of pertinent ancillary legislation.

The research director and the legislative or administrative advisor do not have the time even to scan the large number of technical publications that might contain articles important to current responsibility; these individuals need the background provided by detailed reviews plus an assured awareness of newly developing information, all with minimum time for literature searching. Similarly, the scientist assigned or attracted to a new problem has the requirements of gleaning all literature pertinent to his task, publishing quickly new developments or important new experimental details to inform others of findings that might alter their own efforts, and eventually publishing all his supporting data and conclusions for archival purposes.

The end result of this concern over these chores and responsibilities and with uniform, encompassing, and timely publication outlets in the field of environmental contamination and toxicology is the Springer-Verlag (Heidelberg and New York) triumvirate:

Residue Reviews (vol. 1 in 1962) for basically detailed review articles concerned with any aspects of residues of pesticides and other chemical contaminants in the total environment, including toxicological considerations and consequences.

Bulletin of Environmental Contamination and Toxicology (vol. 1 in 1966) for rapid publication of short reports of significant advances and discoveries in the fields of air, soil, water, and food contamination and pollution as well as methodology and other disciplines concerned with the introduction, presence, and effects of toxicants in the total environment.

Archives of Environmental Contamination and Toxicology (vol. 1 in 1973) for important complete articles emphasizing and describing original experimental or theoretical research work pertaining to the scientific aspects of chemical contaminants in the environment.

Manuscripts for *Residue Reviews* and the *Archives* are in identical formats and are subject to review, by workers in the field, for adequacy and value; manuscripts for the *Bulletin* are not reviewed and are published by photo-offset to provide the latest results without delay. The individual editors of these three publications comprise the Joint Coordinating Board of Editors with referral within the Board of manuscripts submitted to one publication but deemed by major emphasis or length more suitable for one of the others.

March 1, 1977 Coordinating Board of Editors

The citrus reentry problem: Research on its causes and effects, and approaches to its minimization

By

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I. Introduction

The "reentry problems" arises from agricultural workers becoming ill as a result of entering and working in a field some time after a pesticide application has been made to a crop plant. Although sulfur, with its capacity to cause eye irritations, may be claimed to have caused the first reentry problem in agriculture, the problem as currently evaluated is limited to the use of cholinesterase (ChE)-inhibiting organophosphorus (OP) pesticides. The definition of the problem in the future will likely extend to other compounds and other biological effects, such as conjuntivitis and dermatitis. Table I lists the reported cases of post-treatment illnesses to farm workers in California, where most of the incidents in the United States have occurred. A few isolated incidents have been reported from some of the cotton-and tobacco-growing states in this country; there are no documented reports yet from any other country. It is evident from this table that the problem is not a new one and that it appeared hand-in-hand with the introduction of OP pesticides to agriculture. The use of OP pesticides has increased greatly and will probably continue to increase as the use of organochlorine pesticides becomes more restricted. Production and usage of OP pesticides in the United States are expected to continue at a high level for the forseeable future, even though other approaches to pest controls such as biological control techniques, pheromones, and new classes of pesticides will be added to pest-control methods. More intensive farming methods, including OP pesticides, are being introduced to many other countries, and the same problems experienced in the United States will probably be experienced by these other users. Although foods, fibers, and feedstuffs were grown successfully in the United States prior to the introduction of synthetic pesticides in the late 1940s, expectations have changed regarding both quality and quantity of agricultural production as a result of the effectiveness of chemical pest control. Increasing world population requires greater productivity and it is, therefore, unrealistic to halt the use of OP compounds as a solution to the reentry problem (TASK GROUP 1974).

QUINBY and LEMMON (1958) documented in detail 11 worker-poisoning episodes involving 70 workers engaged in thinning, picking, cultivating, or irrigating crops of apples, pears, grapes, oranges, and hops treated with one lb or more of parathion[1]/A. QUINBY *et al.* (1958) reported on health hazards, including reentry hazards, due to the use of OP pesticides, primarily methyl parathion and azinphosmethyl, in cotton culture in the delta area of Mississippi. Following an outbreak of illness among peach harvesters in 1963, MILBY *et al.* (1964) studied 186 peach orchard workers in relation to pesticide application practices and fruit harvesting procedures representative of the orchards in which they worked. Although parathion could be easily recovered from all elements of the orchard environment, it was not present in amounts deemed sufficient to account for the observed illnesses. The toxic parathion alteration product paraoxon was postulated as the prime cause of the outbreak. DAVIES *et al.* (1976) reported on the occurrence of systemic poisoning in 1970 when 20 workers became ill shortly

[1]Chemical designations of pesticides mentioned in text are listed in Table XXIII.

after entering a cornfield in Florida that had been sprayed the day before with a mixture of ethyl and methyl parathion. WARE and MORGAN (1976) have expressed concern for the cotton insect field checkers (cotton scouts) who may acquire up to ten hr of intermittent pesticide residue exposure/day and up to 40 hr/week.

Thus, reported problems involving injury or illness as a result of exposure to treated crops have been largely limited to tree fruits, grapes, tobacco, and cotton where hand labor is involved, resulting in continuous and extensive contact with treated foliage and other plant surfaces (PAYNTER 1976); however, one cannot rule out the possibility of risk in leafy vegetables such as cauliflower, Brussels sprouts, artichokes, cabbage, broccoli, celery, and lettuce (TASK GROUP 1974). This is supported by the 1970 Florida cornfield episode (DAVIES /et al. 1976). The principal hand labor operations requiring contact with treated foliage and other plant surfaces include harvesting, fruit thinning, summer pruning, and propping (grapes). In addition, "scouting" to determine the cotton pest situation and the need for pesticide treatment may result in considerable contact with treated foliage. Cotton scouts, however, could be considered a special group of field laborers requiring special methods of protection (TASK GROUP 1974). While some exposure is unavoidable in each of these operations, harvest normally results in the greatest exposure of workers to treated crops (PAYNTER 1976). With all crops, parathion is the compound most often involved when episodes of worker poisoning occur. This may be coincidental with its extensive use in pest control or may be related to some special property associated with the compound, such as its high dermal toxicity. The heart of the reentry problem is not that workers become ill after entering a pesticide-treated grove several hours or days after application, as this might be anticipated, but rather arises from the fact that episodes are reported in groves several weeks or months after the last known pesticide application, during which time toxic residues should have dissipated to a safe level.

a) Physiological effects of OP pesticides on workers

The cause of the worker illnesses is attributed to exposure to the OP pesticides and their alteration products. The primary mode of action of most of these pesticides is the inhibition of ChE enzymes throughout the body. Normally, acetylcholine liberated at the presynaptic endings and motor endplates by nerve impulses acts directly upon motor and autonomic effector cells to produce appropriate responses. The enzyme ChE terminates the response by hydrolyzing acetylcholine to choline and acetate ion. There is also a considerable quantity of the enzyme in the circulating blood, both in the red cells and in the plasma; physiologists have not yet given a satisfactory account of the role of these high concentrations in blood (GAGE 1967). OP compounds entering the bloodstream may undergo a number of reactions. They may be hydrolyzed to relatively nontoxic products. Compounds containing the P=S group may be converted to P=O compounds (oxons) by mixed-function oxidase enzymes, making them much more toxic, but generally also more susceptible to hydrolysis.

Table I. *Reported cases of post-treatment illness to farm workers in California, 1949 - September 10, 1976* (courtesy J. B. Bailey).

Location (California)	Date	Crop	Activity	Chemical(s) implicated (lb a.i./A)	Days since last sprayed	No. persons involved
Marysville	July 1949	pears	picking	parathion (2.5)	12	20-25
Highland	May 1951	citrus	cultivating	parathion (–)	8	1
Delano	June 1951	grapes	stripping & thinning	parathion (1.87)	33	16
Riverside	Aug. 1952	oranges	picking	parathion (12.00)	16-19	11
Riverside	July 1953	oranges	picking	parathion (–)	17	7
Entire state	1959	citrus	picking	parathion (–)	—	275
Hughson	Aug. 1963	peaches	picking	parathion (7.14)	14-38	94
Navelencia	Aug. 1966	oranges	picking	parathion (9.00)	—	11
				malathion (6.75)	28	—
Terra Bella	June 1966	oranges	picking	parathion (1.78)	15	8
Porterville	July 1966	oranges	picking	parathion (1.3)	32	6
Lindsay	July 1966	oranges	picking	parathion (2.0)	15	3
Terra Bella	Aug. 1966	oranges	picking	parathion (3.75)	—	9
				ethion (1.60)	46	—
Hughson	Aug. 1967	peaches	picking	azinphosmethyl (–)	—	22
				ethion (–), TEPP (–)	38-47	—
Lindsay	May 1968	oranges	picking	parathion (3.75)	22	19
Porterville	May 1970	lemons	pruning	dioxathion (6.0)	—	1
				naled (1.0)	1	—
Terra Bella	May 1970	oranges	picking	ethion (4.0)	—	8
				azinphosmethyl (1.5)	11-14	—
McFarland	Sept. 1970	oranges	picking	dioxathion (4.0)	120	35
				parathion (9.0)	34	—
Orosi	Oct. 1970	oranges	picking	parathion (3.0)	—	11
				malathion (4.5)	31	—

Tulare County	Aug. 1971	olives	pruning	parathion (1.5)	30	6[a]
Porterville	May 1972	oranges	picking	parathion (2.5)	21	3
Exeter	June 1972	oranges	picking	parathion (–)	12	9
Huron	Sept. 1972	lettuce	weeding	parathion (0.5)	0	2[a]
				methamidophos (0.5)	0	–
Corona	May 1973	oranges	picking	Bonamite® (2.2)[b]	52	13[c]
Fowler[d]	30 Aug. 1973	grapes	picking	ethion (1.0)	60	12-30[e]
				phosalone (3.0)	40	–
				phosmet (–)	60	–
				dialifor (1.0)	41	–
				carbophenothion (3.0)	60[f]	–
				naled (–)	7[f]	–
Richgrove[d]	23 July 1974	oranges	picking	parathion (–)	3	2
				malathion (–)	–	–
Kerman[d]	1 Sept. 1974	grapes	picking	phosalone (2.5)	64	2-5
				azinphosmethyl (1.0)	25	–
Lemoncove[d]	11 June 1975	oranges	picking	parathion (2.0)	14	3-15
Madera[g]	8-10 Sept. 1976	grapes	picking	dialifor (–)	–	115
				phosalone (–)	–	–
				ethion (–)	–	–
				methomyl (–)	–	–
				dimethoate (–)	–	–

[a] Report from Fresno Co. Agricultural Commissioner's Office, November 1971.
[b] Bonamite® 75 WP, U27, 415; use cancelled, dermatitis.
[c] Report from Riverside Agricultural Commissioner's Office.
[d] Reports furnished by Drs. J. Knaak and K. Maddy, California Department of Food and Agriculture.
[e] California Department of Food and Agriculture and California Department of Health.
[f] No residue found on foliage.
[g] Reports from Drs. J. B. Bailey and W. F. Spencer.

Through a two-stage process, the second of which is irreversible, OP compounds can attach a phosphoryl group to ChE and thereby render the enzyme unable to perform its function. Excessive concentrations of acetylcholine, therefore, accumulate in the endings of parasynaptic nerves to the smooth muscles of the iris, ciliary body, bronchial tree, gastrointestinal tract, blood and blood vessels, in the secretory glands of the respiratory tract, in cardiac muscle, and in the synaptic nerves of sweat glands. There is also increased accumulation of acetylcholine in motor nerves to voluntary muscles in the autonomic ganglia, and in the central nervous system (KRAMER 1972). SUMERFORD *et al.* (1953) postulated that the rate of fall in ChE rather than the level of ChE determines whether or not symptoms representing systemic effects of the toxicant could be expected.

The first definite symptoms of intoxication include nausea and loss of appetite which may be aggravated by smoking. Other effects such as vomiting, abdominal colic, diarrhea, sweating, and salivation then ensue. Central nervous system effects, which include restlessness, giddiness, apprehension, slurred speech, loss of coordination, and hyperventilation later become apparent (KRAMER 1972). QUINBY and LEMMON (1958) described symptoms reported for workers involved in poisoning episodes. Often-described complaints and observations, which varied with the degree of exposure and with individuals, were uncontrollable twitching of the eyelids, headache, vertigo, nausea, vomiting, retching, subnormal temperature, chills, excessive perspiration, pallor, weakness, fast pulse, twitching of arm and leg muscles, chest pains, abdominal cramps, visual disturbance, and miosis (pinpoint pupils). Administration of atropine gave dramatic relief. MILBY *et al.* (1964) reported that the most consistent complaints described by clinically ill peach pickers were nausea, vomiting, headache, profound weakness, and extreme malaise. Other manifestations of parasympathetic stimulation including miosis, blurred vision, dizziness, excessive sweating, salivation, diarrhea, and abdominal cramping were reported, but not consistently so.

Large doses of OP pesticides lead ultimately to death, but the field worker is extremely unlikely to accumulate such dosages. The onset of symptoms whose severity increases with continued exposure should force the individual to cease working and thus to terminate continued exposure. No fatal cases of poisonings from pesticide residues related to reentry have been verified (TASK GROUP 1974). Thus, the reentry problem is concerned with the debilitating effects on the workers with the added burden of loss of income during the illness to a group which can least afford it. Following human exposure to OP compounds, plasma ChE levels return to normal within a few days; red blood cell (RBC) ChE levels, however, only return to normal as new red blood cells are produced and require much longer for complete recovery to pre-exposure levels. For the overall safety of workers, it would be desirable also to know whether inhibition of ChE is the sole toxic action of the OP compounds. While no gross or microscopic pathologies were observed after two one-year chronic feeding studies at 100 ppm of parathion in the total diet of rats (WILLIAMS *et al.* 1958), certain effects may not have been observable with the sample size used. All OP pesticides are alkylating agents and thus, in addition to the hazards of acute toxicity to workers caused by the anti-ChE properties, there may be another hazard from the alkylating effects of these compounds on DNA (deoxyribonucleic acid). Several OP pesticides, including dichlorvos, trichlorfon, dimethoate, Bidrin, and oxydemeton-methyl, have been shown to be potentially mutagenic in some organisms (WILD *et al.* 1975), raising the possibility of germ cell mutations and carcinogenicity in man. It should be strongly emphasized that no evidence for mutagenic or carcinogenic activity of OP pesticides in man has ever been observed. All mammals have a variety of effective defenses against potentially mutagenic chemicals in various organs and within the cells themselves. However, both dimethoate and trichlorfon have been reported to be carcinogenic in rats and mice (GIBEL *et al.* 1973) when applied for long periods at high doses.

Little data exist on carcinogenicity and mutagenicity of OP pesticides in mammalian species, due at least partly to the time and expense of animal testing. KOLATA (1976) stated that an animal test program for carcinogenicity requires about $100,000 and three years' time. At least one other animal study, on dichlorvos, has been carried out, using mice and Chinese hamsters; no detectable effects were observed (DEAN and THORPE 1972). Other studies have been performed which indicate little or no effect from normally encountered concentrations of pesticides; WILD (1975) has observed that tissue concentrations in mammals following exposure to pesticides are lower by a factor of 10^3 to 10^5 than the lowest concentrations which are mutagenic for microbes *in vitro*. Although it may well be an insignificant hazard, the possibilities of mutagenicity and carcinogenicity are an argument for keeping exposure to OP pesticides as low as possible.

b) Routes of worker exposure to residues

There are three principal routes by which workers may be exposed to OP pesticides: respiratory, oral, and dermal. Pesticide applied to foliage can dissipate through volatilization and thus give a finite air concentration of pesticide; workers would then be exposed simply through breathing the air. Also, worker activity can disperse pesticide-bearing particulate matter into the air, and the workers could inhale the dust. Small particles ($<7\mu$) would penetrate into the respiratory tract, giving rise to respiratory exposure, while larger particles would be trapped by mucus which can be swallowed, resulting in oral exposure. The pesticide residing on the plant surface or sorbed to particulate matter on foliage or on the soil surface can be transferred to worker skin and clothing, resulting in dermal exposure.

CARMAN *et al.* (1952) were first to note that in Southern California, citrus tree surfaces are normally covered with dust and microdebris and that this condition coupled with the use of parathion formulated as wettable powders may lead to losses of parathion from the foliage as air dispersions of contaminated particulate matter; their analytical data supported this contention. The most hazardous exposure to parathion residues was postulated to result from mechanical dislodgment of pesticide-bearing dust during operations involving tillage, picking, pruning, and similar operations. Later, QUINBY and LEMMON (1958) also discounted vapor exposure to parathion; they attributed illnesses to extensive contact with fruit and less extensive contact with foliage, a conclusion now reversed. MILBY *et al.* (1964), using alcohol swabs, confirmed the presence of parathion on the arms and trunk as well as on the palms and hands of peach harvesters and suggested that contact with leaves and tree surfaces contributed to total exposure. GUNTHER *et al.* (1973) considered foliage to be a more important source of toxic material than fruit, due to the much greater area of foliage/tree; for the average 20-year-old orange tree, the leaf:fruit area ratio is 17:1 and for the average 10-year-old peach tree this ratio is 53:1 for freestone and 28:1 for cling peaches.

WARE and MORGAN (1976) estimated that field checkers working for 30 min in a just-treated cotton field would accumulate parathion residues of 3.5 mg on the hands, 1.1 μg by inhalation, and 18 mg on the clothing. Thus, the hands and clothing were the greatest sources of pesticide chemical exposure. Although the amount of trapped material was of sufficient quantity to have an effect on ChE activity if completely absorbed, exposure did not affect serum or RBC ChE levels and there was no detectable *p*-nitrophenol in urine collections from subjects up to 48 hr after exposure. WARE and MORGAN (1976) suggested that data on skin and clothing contamination by the toxicant are therefore inadequate for evaluation of pesticide absorption and effect. It is obviously difficult to estimate how much of the pesticide trapped by clothing ultimately penetrates the skin. MILBY *et al.* (1964) described peach harvesters' clothing as being light in weight due to the heat and often sweat-impregnated. Shirts were open at the collar and often shirt sleeves were short or rolled above the elbow. Exposure of greater skin area would, of course, facilitate percutaneous pesticide penetration. The use of protective clothing in warm weather is impractical (PAYNTER 1976). Thus, at temperatures which prevail during harvest seasons in most parts of the United States, workers engaged in strenuous physical activities will not wear rubber garments, respirators, and other types of protective devices. The risk of heat prostration may well be greater than the risk from pesticide residues. Also QUINBY and LEMMON (1958) pointed out that most of the laborers who do thinning and similar agricultural tasks may wear their work clothing for a week or longer without laundering. Prolonged wearing of contaminated clothing was thought to increase the likelihood of poisoning. Personal hygiene involving bathing and frequent clothing changes can only be recommended to workers since such a requirement cannot be practically enforced. In addition, studies have shown that washing of skin exposed to pesticides lowers but does not prevent percutaneous pesticide exposure (TASK GROUP 1974).

A complex lipid mixture known as sebum covers and permeates the outer horny skin layer known as the stratum corneum. Sebum contains many lipids and has a high affinity for lipid-soluble, water-insoluble substances and thus is ideal for the acquisition of most toxic pesticide residues. The average adult skin area is about 17,000 cm^2, and the area which is most likely to be directly exposed (face, neck, "V" of the chest, forearms, and hands) is about 2,100 cm^2. There is little literature to allow prediction of the relationship between surface concentration and penetration. In the case of parathion, by increasing the surface dose from four to 2,000 $\mu g/cm^2$, the percentage of the applied dose that was absorbed remained relatively constant (TASK GROUP 1974).