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MERCURY CONTAMINATION IN MAN AND HIS ENVIRONMENT

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FOREWORD

In May 1967, at a Symposium organized by the International Atomic Energy Agency in Amsterdam, the special problems of food and environmental contamination by mercury were discussed by world experts on the subject and by representatives of FAO, WHO and IAEA. One of the recommendations made by this meeting was that the international organizations of the United Nations family should assist in the collection and distribution of information on environmental mercury. Subsequently, the organizations concerned agreed that a handbook on mercury contamination would be especially useful. This would deal with sources of mercury in relation to man and his environment; with physical and biological transfer processes that determine its distribution; with analytical methods for determining mercury and its compounds as environmental contaminants; with actual concentrations of mercury found in the environment, in living organisms and in man; and with its toxicology in animals and man.

The present monograph, which is published under the auspices of ILO, FAO, WHO and IAEA, represents the result of these earlier deliberations, up-dated in the light of recent developments in this rapidly advancing field. The international organizations concerned wish to express their gratitude to all those who have taken an active interest in the publication and especially to those who have contributed directly to the technical content of the monograph itself.

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1. INTRODUCTION

F.P.W. WINTERINGHAM

The metallic element mercury has been known since ancient times and was certainly mentioned by Aristotle (350 B.C.). Its unique and curious properties of being a mirror-like liquid of high density (13.596 g^{+1} cm^{-3} at 0°C), its relatively high vapour pressure (8×10^{-3} Torr at 40°C; 270×10^{-3} Torr at 100°C) and its consequent volatility (e.g. many times greater than that of the insecticide DDT), its ready ability to dissolve (amalgamate with) certain metals such as copper, zinc, lead and gold, sometimes with the formation of definite compounds (e.g. $NaHg_2$), and its obtuse angle of contact with glass and other "non-wettable" surfaces (depression of the internal mercury surface by a glass capillary) have attracted the attention of philosophers and scientists for hundreds of years. The elusive behaviour of spilled mercury globules is known to almost every schoolchild. The toxic properties of mercury and its compounds have also been known since ancient times, and mercuric chloride solution was one of the first antiseptics used in clinical medicine.

The element occurs in nature in its native elemental form. Indeed, it was the discovery of the mirror-like liquid at the bottom of local drinking wells at Idrija in Yugoslavia which led to its mining and extraction there more than three centuries ago (personal communication from Professor L. Kosta). It occurs in mineral forms especially as cinnabar (mercuric sulphide, HgS). Cinnabar is found in widely separated regions such as Almaden (Spain), Idrija (Yugoslavia), Monte Amiatra (Italy) and in lesser quantities in Peru, California, Mexico, Mainland China and Japan. According to Partington (Textbook of Inorganic Chemistry, Macmillan & Co. Ltd., London, 1933), Pliny reported that 10 000 lb of cinnabar came annually from Spain in his day. Its volatility and occurrence in minerals account for its presence throughout the environment and it can be detected in all living tissues. It is, therefore, a clear example of a natural constituent of the environment which only becomes a pollutant in virtue of the special conditions of concentration or location as a result of man's activities.

The total global production of mercury in 1966 (excluding USSR, China and CSSR) was 7260 metric tons and no significant trend was discernible during the preceding quinquennium (UN Statistical Yearbook, New York, 1968). However, this does not represent the upper limit of potential pollution or environmental contamination as a consequence of man's activities. If the average mercury content of crude oil is taken to be about 0.1 ppm then some 200 tons of mercury may be expected to be released annually into the atmosphere from the combustion of petroleum fuels alone. From the known average mercury content of snow or rainwater it has been estimated that the surface of the earth receives annually some 100 000 tons through precipitation from all mercury sources including natural ones (SAHA, J.G., Significance of mercury in the environment, Residue Reviews, 1972, in press).

Mercury in one form or another lends itself to a wide variety of uses in addition to its well-known use in thermometers and barometers. Major applications include those as an industrial chemical catalyst, in the manufacture of electrical batteries, switches, relays and rectifiers, in the chlorine-alkali industry, in marine paints, in paper and pulp manufacture, as a fungicide in agriculture, in dentistry, pharmacy and in medicine.

Its powerful inhibitory action against basal metabolic processes combined with special conditions of location or concentration as a result of man's activities have had tragic consequences from time to time in terms of human health. Notable were the many confirmed cases of mercury poisoning, a considerable number of which were fatal, in Minamata and Niigata, Japan, as a result of eating fish or shellfish caught in coastal and river waters contaminated as a result of an industrial discharge. Such instances have emphasized the growing problems of environmental contamination by undesirable chemicals, both in fact and in the public mind, and to which the forthcoming UN Conference on the Human Environment stands witness.

The particularly extensive studies of mercury have demonstrated clearly the importance of an overall and balanced approach to such problems. This so-called integrated approach must consider all possible sources of the contaminants, including any natural ones, their physical and chemical fate, behaviour in ecological systems and food webs, and their possible acute and chronic effects not only on human health but on wildlife and the complex of living organisms constituting the exposed ecosystem. Finally, it must consider benefits in terms of human health, comfort and welfare. Thus, in the case of mercury the relatively high mercury concentrations found in some aquatic samples led to an early suspicion that the use of agricultural mercury fungicides was mainly responsible. However, further study of the sources revealed that industrial discharges were the principal causes of contamination. Similarly, careful study of mercury concentrations in tuna fish, including samples of fish caught as long as 90 years ago, as well as studies of natural mercury in the marine environment have shown that the relatively high levels of mercury in tuna fish are unlikely to be due to man's activities. They are more likely to be due to the natural mercury content of the marine environment and the physiology of the fish species concerned (HAMMOND, A.L., Science 171 (1971) 788).

The element mercury lends itself to neutron radioactivation analysis with great sensitivity so that unequivocal identification of the element and its determination in a wide range of biological and environmental samples at concentrations of the order of 1 part in 10^9 have been greatly facilitated. In addition, the availability of radioactive isotopes of mercury such as mercury-203 has enabled the movement, fate, metabolism and excretion of the element to be studied in living organisms and in food chains with unrivalled specificity and sensitivity. Notable applications of these techniques have been made by Professor Miettinen and his colleagues in Finland, by Professor Kosta and his colleagues in Yugoslavia, and by Professor Jervis and his colleagues in Canada.

Such studies have provided a wealth of information on the behaviour of mercury in the environment and in living organisms including man. Of particular interest and significance has been the demonstration that inorganic mercury may undergo biological methylation with the formation of methyl mercury and this organic metabolite is, in fact, more toxic and more stable in vivo than some inorganic forms of the element. Mercury can become concentrated in contaminated seed-bird-mammalian predator food chains, and analyses of fish-muscle tissues have indicated that concentration factors of 1000 or more may obtain in marine and aquatic systems. The inhibition of photosynthesis of phytoplankton by low concentrations of mercury has also been indicated. Despite the wealth of information now available on the occurrence and significance of mercury in the environment many outstanding problems remain to be solved as usefully indicated in Dr. J. G. Saha's comprehensive review of the subject (Significance of mercury in the environment, Residue Reviews, 1972, in press).

The International Atomic Energy Agency, through its co-ordinated research programs on food and environmental contamination and collaborative programs with other UN Agencies, was placed in a good position to encourage an overall appraisal of environmental mercury problems and their study, as shown in the authoritative contributions which follow.

To conclude: The problems of mercury in the environment and their study have demonstrated the great importance of the so-called integrated approach and of taking fully into account the benefits as well as the risks associated with man's exploitation of this important element. For example, this approach has strongly suggested that some uses such as those in dental medicine or the prescribed uses as agricultural fungicides are without significant risk and provide considerable benefits. On the other hand, certain industrial uses and discharges require very careful monitoring, control and possible restriction. The elaboration of maximum permissible concentrations in food and drink, acceptable daily intakes by man and "action levels" must also take fully into account the natural occurrence of this element and its variations. The studies of the natural distribution of mercury in food and environment have also shown how man must always have been, and forever must be, exposed to this element. It is only on the basis of balanced and comprehensive studies that any necessary counter-measures and controls can be identified and developed on a rational basis.

The views and conclusions of this introduction are not necessarily those of the International or UN Agencies concerned.

2. USE OF MERCURY COMPOUNDS IN AGRICULTURE AND ITS IMPLICATIONS

Kristina RISSANEN and J.K. MIETTINEN

2.1. INTRODUCTION

Mercury fungicides have been used for treatment of seeds against fungi for more than half a century. In view of the large scale of use, their safety record is notably good, except for the alkyl mercury compounds, especially methyl mercury, whose agricultural use has been recently discontinued in several countries because of its accumulation by terrestrial food-chains. High levels of methyl mercury in the fish of industrialized regions, as well as severe accidents in which hundreds of people in developing countries have died due to the consumption of cereals treated by methyl mercury or to the consumption of animals which have consumed such treated cereals, have also recently focused attention on mercury fungicides.

The present review of the agricultural use of mercury fungicides is concerned with their persistance in nature, their concentration into foods and their effect on living things, man included.

2.2. DEVELOPMENT AND SCALE OF SEED TREATMENT BY ORGANOMERCURIALS

2.2.1. Development of methods

Inorganic mercury compounds had been in use for seed treatment against fungi since the end of the previous century, but the use of organomercurials was introduced in 1914 in the form of seed treatment with water solutions of hydroxyphenyl mercury chloride (trade name Uspulun). This process, called "wet dressing", was difficult since water had to be removed in order to prevent premature germination. To avoid this problem, the "dry dressing" was developed in the middle of the 1920s. By this technique, the fungicide was mixed with an inert dry powder and in this form further mixed with the seeds in a special mill. Because of the resulting toxic dust, the handling of seeds was unhygienic. Today, however, modern technology has eliminated the hazard of toxic dust. The so-called "short wet" and "slurry" methods were developed in 1927 in which only 2 - 3 litres of liquid agent were necessary for 100 kg of seed. However, the moisture content was still so high that the seed had to be sown within a few days after treatment. This difficulty was overcome in the 1930s when "liquid dressing", with methyl or ethyl mercury compounds, was developed. This dressing requires only 100 to 200 ml of a 1% solution of the agent per 100 kg of seed. In Sweden, methyl mercury dicyandiamide was introduced in 1938 under the trade name Panogen, and the "liquid dressing" technique soon replaced other methods of seed treatment in that country because of its efficacy, safety and technical ease. Later, it also became popular in other European countries, and in the USA and Canada as well. Mixtures of organomercurials and chlorinated hydrocarbons have also been on the market, but their use is now forbidden in many countries, e.g. Finland and Sweden.

The direct treatment of growing plants with organomercurials is a relatively new method. The use of phenyl mercury acetate in the prevention of rice blast began in Japan as late as 1952, but grew very rapidly until it was terminated around 1969. (See also FAO/WHO, Evaluation of Some Pesticide Residues in Food, FAO/PL; 1967/M/11/1, Rome 1968, p. 200 et seq., and FAO/WHO, Evaluation of Some Pesticide Residues in Food, FAO/PL-CP/15, Rome 1967, p. 188 et seq.)

2.2.2. Mercury compounds

Inorganic mercury (HgCl, $HgCl_2$, HgO) has been little used. On the other hand, hundreds of organic derivatives have been used, all of the type R-Hg-X, in which R is an organic radical and X originates from an organic or inorganic substance possessing a dissociable hydrogen atom. Examples of X, or substances which may produce X, are:

Inorganic acid radicals: chloride, bromide, cyanide, phosphate;
Organic acid radicals: acetate, benzoate, salicylate;
Amides: urea, thiourea, formamide, dicyandiamide;
Phenolates: pentachlorophenolate, 8-hydroxyquinolinate;
Mercaptides: 2, 3-dihydroxypropylmercaptide.

Although the anion can modify the properties of the compound, it is of less importance than the radical R. The classification of mercury fungicides is therefore based upon R, according to which the organomercurials are usually divided into the following three groups:

(1) Alkyl mercuric compounds:
    Methyl mercury (CH_3Hg-),
    Ethyl mercury (C_2H_5Hg-);
(2) Alkoxyalkyl mercuric compounds:
    Methoxyethyl mercury ($CH_3OC_2H_4Hg$-),
    Ethoxyethyl mercury ($C_2H_5OC_2H_4Hg$-);
(3) Aryl mercuric compounds:
    Phenyl mercury (C_6H_5Hg-),
    p-Tolyl mercury ($CH_3C_6H_4Hg$-).

In each group, the two most common radicals in use are mentioned.

2.2.3. Chemical and physical properties

The solubility of organomercurials in water and organic solvents is primarily determined by the anionic (amidic) radical, although the radical R also has some influence. In general, the aryl derivatives are less soluble in water than the alkyl derivatives, given the same anion.

Volatility is an important property of seed dressings. Generally, the alkyl compounds are the most volatile. However, volatility can be regulated to a certain extent by choosing the proper anion. In Table I, which is taken from Swensson and Ulfvarsson (1963), the volatilities of some organomercurials and of metallic Hg are presented as saturated vapour concentrations (for references, see original).

TABLE I. SATURATED VAPOUR CONCENTRATION OF SOME ORGANO-MERCURY SALTS AT 20°C (SWENSSON AND ULFVARSSON, 1963)

Cation	Anion	Vapour concn ($\mu g/m^3$)
Methyl-Hg	Chloride	94 000
	Bromide	94 000
	Iodide	90 000
	Acetate	75 000
	Hydroxide	10 000
	Toluenesulphonate	15 000
	Benzoate	2 000
	Dicyandiamide	300
Ethyl-Hg	Chloride	8 000
	Bromide	7 000
	Iodide	9 000
	Dicyandiamide	400
	Monohydrophosphate	50
Methoxyethyl-Hg	Chloride	2 600
	Acetate	2
Phenyl-Hg	Acetate	17
	Chloride	5
	Nitrate	1
	Methanedinaphthyldisulphonate	2
Metallic Hg		14 000

The C-Hg bond of the R-Hg- radical is chemically stable. It is neither split in water nor by weak acids or bases. However, as will be described later, only the alkyl-Hg bond is stable in soil and in most organisms. The C-Hg bond in alkoxyalkyl- and aryl-mercury compounds is easily broken, even in animals, thus producing inorganic mercury.

2.2.4. Dressing techniques

The techniques used for seed dressing in the middle of the 1960s were "liquid dressing", "dry dressing" and the "slurry method" (Ulfvarsson, 1969). They require different properties of the dressing agent, for the same agent may be excellent in one technique but poor in another, as was shown by Gassner (1951). "Liquid dressing" requires some volatility (at least 100 $\mu g/m^3$ at 20°C; Lindström, 1958; Bombach, 1963) in order to guarantee

TABLE II. CHEMOTHERAPEUTIC INDEX (c/t) ACCORDING TO GASSNER (1951)

Compound	Dosis curativa, c (ppm Hg)	Dosis toxica, t (ppm Hg)	Chemotherapeutic index, c/t
Methyl-Hg	15	72	0.21
Ethyl-Hg	23	56	0.41
Propyl-Hg	37	12	3.0
Butyl-Hg	44	10	4.4
Octyl-Hg	77	54	1.4
Methoxyethyl-Hg	36	73	0.50
Ethylene-Hg	42	80	0.52
Phenyl-Hg	34	37	0.92
Cyclohexyl-Hg	46	26	1.8
$HgCl_2$	200	380	0.53

its homogeneous distribution via the vapour phase during storage. Higher volatility increases the risks. The less moisture involved, the less damage to the seeds. In "dry dressing", the homogeneity of distribution is aided by the inert carrier. The agents are usually rather non-volatile and have a fungicidal effect only in the moisture of the soil.

2.2.5. Formulation and fungicidal effectivity

Gassner (1951) compared the effectivity of a number of mercury fungicides in protecting wheat against Tilletia tritici by the "short wet" and the "dry dressing" methods. In the process he determined the so-called chemotherapeutic index, which is the ratio dosis curativa/dosis toxica, dosis curativa being the lowest concentration of the dressing that prevents germination of the fungi and dosis toxica being the lowest dressing concentration that noticeably inhibits growth of the wheat. The effectivity of the dressing agent was determined by the structure of the R group, the X group being relatively insignificant. Some of Gassner's results are presented in Table II. As may be seen, methyl-Hg is the most effective dressing, (i. e. the index is lowest), while ethyl-Hg, ethylene-Hg and methoxyethyl-Hg are about two times and phenyl-Hg about five times less effective. Only these compounds are sufficiently effective in the "dry dressing" method.

Ishiyama (1969) determined the effectivity of 58 R-Hg-X type compounds against the rice blast Piricularia oryzea and established the following order: phenyl > o- , p-tolyl > ethyl, methoxyethyl, dimethylphenyl > naphthyl. The anion (-X) had little influence, iodide being the most effective when

TABLE III. DISEASES OF CEREALS CONTROLLED BY SEED TREATMENT WITH MERCURY FUNGICIDES

Cereal	Disease, common name	Disease, scientific name
Wheat	Bunt	Tilletia tritici
	Bunt	Tilletia foetida
	Snow mould	Fusarium nivale
	Seedling blight	Fusarium spp.
Barley	Covered smut	Ustilago hordei
	Leaf stripe	Helminthosporium gramineum
	Net blotch	Helminthosporium teres
	Seedling blight	Fusarium spp.
Rye	Snow mould	Fusarium nivale
	Stripe smut	Urocystis occulta
	Seedling blight	Fusarium spp.
Oats	Loose smut	Ustilago avenae
	Leaf spot	Helminthosporium avenae
	Covered smut	Ustilago levis
	Seedling blight	Fusarium spp.
Maize	Leaf spot	Helminthosporium spp.
Rice	Blast	Piricularia oryzae[a]
	Stem rot	Helminthosporium sigmoideum[a]
	Brown spot	Cochliobolus miyabeanus[a]

[a] Also sprayed during growth.

the radical R- was phenyl. The fungicides were thought to act at three stages of fungi development:

(1) Conidial germination;
(2) Formation of the appressoria; and
(3) Penetration of the plant surface.

Thus, for instance, phenyl mercury acetate prevents the latter two stages, but does not prevent conidial germination.

TABLE IV. USE OF MERCURY FUNGICIDES FOR SEED DRESSING IN FIFTEEN EUROPEAN COUNTRIES IN SPRING 1965 ACCORDING TO GRANHALL (1965)

Country	Area 1000 ha	Arable land 1000 ha	Sown with cereal 1000 ha	Mercury dressed 1000 ha	Percentage that is mercury dressed of:		
					The cereal	The arable land	The total area
Sweden	41.000	3.290	1.470	1.180	80	36	3
Norway	30.900	850	240	120	50	14	0.4
Finland	30.500	2.680	1.020	300	30	11	1
Denmark	4.300	2.820	1.570	1.290	82	46	30
Great Britain	24.100	7.270	3.400	2.630	77	36	11
Ireland	6.900	1.360	390	250	64	18	4
Netherlands	3.400	1.030	510	370	73	36	11
Luxemburg	260	75	45	30	67	40	12
West Germany	24.300	8.500	4.990	2.740	55	32	11
Poland	30.400	16.180	8.630	2.460	29	15	8
Austria	8.300	1.750	860	670	78	38	8
Hungary	9.300	5.620	3.000	1.700	57	30	18
Switzerland	4.000	420	160	90	56	21	2
France	54.700	21.410	7.910	2.850	36	13	5
Turkey	77.700	25.200	10.700	5.780	54	23	7

2.2.6. Plant disease and rate of use

The purpose of the seed dressing is to prevent the diseases which are spread with the seeds and to protect the germinating seeds against secondary infections. The most important cereal diseases which can be prevented by seed dressing are presented in Table III.

According to an announcement made by the US Department of Agriculture, (USDA, March 9, 1970), alkyl mercury fungicides are registered in the USA for the treatment of seeds of the following plants: barley, wheat, oats, rye, corn, millet, milo, sorghum, rice, beans, peas, cotton, flax, peanuts, soybeans, sugar beets, tomatoes and safflower.

Mercury fungicides are also used in the spraying of fruit trees against various types of scab, and rice against blast *Piricularia oryzea*, stem rot *Helminthosporium sigmoideum* and brown spot disease *Cochliobolus miyabeanus*. In countries with a snow-cover lasting at least three months, the protection of stands of winter cereals against *Typhula*, *Sclerotinia borealis* and especially *Fusarium nivale* is important. Mercury fungicides are effective for this purpose but, at least in Finland, only mercury-free compounds are used (Jamalainen, 1964).

Fungus diseases of turf can be controlled by alkyl-Hg and phenyl-Hg compounds (Ulfvarsson, 1969). Their use seems to be especially wide in Canada. In 1967, 2.8 metric tons of mercury were used in this form for the treatment of golf courses and home lawns (Fimreite, 1970a). Smart (1968) presented a list of about 40 plants and 50 corresponding diseases, and also mentioned the stage and the rate of fungicide application most effective in control of the disease. The amounts used vary greatly, but are usually between 0.02 and 0.3 g Hg/kg seed. In fruit treatment, the amounts vary from 10 to 400 g Hg/ha[1], and in soil treatment (root crops), they are an order of magnitude greater (0.5 - 10 kg Hg/ha). Until 1950, only inorganic compounds seem to have been used in soil treatment.

2.2.7. Concentrations and extent of use

The concentration of organomercurials in the differing fungicidal formulations varies greatly. Smart (1968) sets the following limits (all figures are percentages of Hg in formulation) for seed dressings: "dry" 0.2 - 10, "wet" 0.4 - 6, "slurry" 1.3 - 6; for bulb dips: "liquid" 1.5 - 6; for seed-potato dips: "liquid" 0.5 - 6, "dry" 3 - 9; for sugar-can dip: 6; for glasshouse aerosols: 0.35; for orchard canker paint: 2; for orchard spray: "dispersible powder" 0.6 - 40, "emulsifiable concentrate" 0.4 - 40; and for lawn: 3.4 - 6.7.

The inorganic mercury fungicides (HgCl or HgO) used in the treatment of potato soil contain 10% Hg, while those for lawn treatment contain 45% Hg.

Granhall (1965) reviews well the extent of mercury fungicide use in 15 European countries as of spring 1965. Table IV is taken from this review and it shows that a large percentage of all the cereals grown in Europe in 1965 was dressed. Healthy seeds were also preventively treated. These figures probably represent the highest level of use, at least in Scandinavia. In 1965, much attention was directed there to the death of birds caused by dressed seeds, and farmers accordingly reduced their use of mercury

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[1] 1 hectare (ha) = 2.48 acres.

TABLE V. QUANTITIES OF MERCURY COMPOUNDS USED IN OR SOLD TO AGRICULTURE IN 1965 ACCORDING TO SMART (1968)

Country	Metric tons of Hg compounds	Country	Metric tons of Hg compounds
USA	400	Norway	0.4
Denmark	3.5	Portugal	0.2
Germany	41	Turkey	22.5
Great Britain	20	Spain	7.1
Bulgaria	5	Sweden	2
Finland	1	Morocco	1
Italy	26	Israel	0.2
Poland	9	New Zealand	0.5
Austria	4	Japan	1 600 (approx.)

fungicides. In Sweden in 1965, treatment was restricted to contaminated lots of seed. Dressing was also reduced in other Scandinavian countries. Lihnell (1969) stated the following figures for the use of mercury fungicides in 1968 (as percentage of seed sown): Sweden 25 - 30, Norway 35 - 40, Denmark 80 and Finland 30. Smart (1968) gave figures for the agricultural use of mercury fungicides in 1965 in a number of countries (world total: 2100 t). These are presented in Table V. In the USA, only 120 t were used in 1968 (Nelson et al., 1971), while in Japan, in the same year, about 400 t were used (personal communication, 1969). About 16 t were used in Canada in 1965, and only 9 t in 1968 (Fimreite, 1970a). In Scandinavia, the use of mercury fungicides in 1968 was on about the same level as in 1965 (Lihnell, 1969).

2.2.8. Control of mercury residues

Some countries have established "tolerances" [maximum concentration of pesticide residue that is permitted in or on food at a specified stage of harvesting, storage, transport, marketing, etc. .... (see FAO/WHO, Pesticide Residues in Food, Report of the 1969 Joint Meeting of the FAO Working Party of Experts on Pesticide Residues and the WHO Expert Group on Pesticide Residues, FAO Agricultural Series, 84, Rome 1970, p. 40)] which have been reported to vary from 0 to 0.1 ppm (FAO/WHO, Evaluation of Some Pesticide Residues in Food, FAO/PL; 1967/M/11/1, Rome, 1968, p. 208). In view of the lack of adequate data and toxicological information the 1967 Joint FAO/WHO Meeting on Pesticide Residues was unable to recommend tolerances for organo-mercury compounds but "By way of guidance, however, practical residue limits [maximum unintentional residues (FAO/WHO Pesticide Residues in Food, Report of the 1969 Joint Meeting of the FAO Working Party of Experts on Pesticide Residues and