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Pesticide Residues in Imported Spices. A Survey for Chlorinated Hydrocarbons

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Pesticide analyses for residues of chlorinated hydrocarbons were performed on 28 spices from 25 producing countries. Several shipments were examined during several crop years. In all, 226 samples were analyzed. Low levels of DDT and BHC were detected consistently, the level being generally below 0.5 ppm. Residues of other chlorinated hydrocarbons such as Dieldrin, Endrin, and HCB were detected sporadically at a very low level. Except for oregano from Mexico, some of which is grown in an area where DDT is actively used, there is no relationship between pesticide residues and country of origin or individual spice. In view of the low level detected and the uniformity of detection, there does not appear to be any cause for concern.

The increased statutory and regulatory attention given to the use of pesticides and the subsequent reduction of residue tolerances has been of considerable interest to the membership of the American Spice Trade Association. As a group, it represents the major importers of spices which are grown throughout the world, especially in the developing nations where laws and practices are substantially different from ours.

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Of particular interest are residues of the chlorinated hydrocarbons which continue to be used in some countries for a variety of reasons. In Mexico, for example, DDT is used for malaria control in the same region in which oregano is collected after harvest.

The significance of continued worldwide use of the chlorinated hydrocarbons was brought to our attention early in 1973 when several shipments of paprika imported from Spain were detained by the Food and Drug Administration (FDA) for having residues of benzene hexachloride (BHC) in excess of the tolerance of 1 ppm ap-

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Table I. Summary of DDT Residues (ppm)

item	source	N	average	range
allspice	Guatemala	6	0.38	0.03-0.96
	Jamaica	6	0.02	ND[a]-0.05
anise	Spain	6	0.04	0.02-0.07
basil	Hungary	5	0.58	0.50-0.73
	France	3	0.25	0.23-0.27
bay leaves	Turkey	6	0.06	0.03-0.08
black pepper	Brazil	5	0.23	0.10-0.26
	Indonesia	5	0.11	ND-0.32
	Malaysia	5	0.25	0.15-0.37
	India	4	0.10	0.06-0.18
caraway	Poland	3	0.23	0.14-0.29
	Holland	4	0.01	ND-0.01
cardamom	Guatemala	2	0.03	0.02-0.03
cassia	China	6	0.12	0.02-0.52
	Indonesia	3	0.04	0.02-0.06
	Seychelles	5	0.03	0.01-0.06
celery seed	India	11	0.43	0.08-1.67
chillies	Japan	3	0.01	ND-0.01
	Turkey	3	0.81	0.53-0.96
	China	4	0.80	0.02-2.64
	Mexico	10	0.15	0.02-0.33
cloves	Madagascar	10	0.32	0.05-0.88
coriander	Romania	3	0.05	0.01-0.11
	Morocco	4	0.15	0.08-0.20
cumin	Iran	6	0.03	0.02-0.04
dill seed	India	3	0.49	0.46-0.55
fennel	India	11	0.15	0.10-0.45
ginger	India	8	0.28	0.01-0.80
	Nigeria	1	0.08	
mace	Indonesia	3	0.03	0.01-0.07
marjoram	Egypt	3	0.12	0.10-0.14
	France	5	0.13	0.02-0.24
nutmeg	Indonesia	1	0.02	
oregano	Mexico	3	1.71	1.24-2.42
	Greece	6	0.08	0.01-0.16
poppy seed	Holland	4	<0.005	
rosemary	Yugoslavia	5	0.09	0.05-0.21
	Spain	3	0.11	0.09-0.14
sage	Albania	9	0.32	0.10-0.68
	Yugoslavia	5	0.12	0.06-0.22
sesame	Guatemala	4	0.15	0.07-0.21
	Mexico	2	0.02	0.01-0.02
tarragon	France	3	ND	
thyme	Spain	3	0.08	0.04-0.10
	France	3	0.11	0.10-0.12
turmeric	India	4	0.03	0.01-0.05
white pepper	Brazil	6	0.25	0.02-1.14
	Malaysia	3	ND	

[a] ND = nondetectable.

Table II. Summary of BHC Residues (ppm)

item	source	N	average	range
allspice	Guatemala	6	0.04	<0.005-0.09
	Jamaica	6	0.03	<0.005-0.13
anise	Spain	6	0.12	0.02-0.21
basil	Hungary	5	0.06	0.04-0.10
	France	3	0.08	0.07-0.73
bay leaves	Turkey	6	0.03	0.01-0.05
black pepper	Brazil	5	0.01	0.01-0.02
	Indonesia	5	<0.005	
	Malaysia	5	0.19	0.13-0.33
	India	4	0.11	0.02-0.22
caraway	Poland	3	0.01	0.01-0.02
	Holland	4	<0.005	
cardamom	Guatemala	2	<0.005	
cassia	China	6	0.24	0.06-0.68
	Indonesia	3	0.02	<0.005-0.05
	Seychelles	5	0.01	<0.005-0.02
celery seed	India	11	0.37	0.08-0.68
chillies	Japan	3	0.01	0.01-0.02
	Turkey	3	0.21	0.17-0.25
	China	4	0.34	0.20-0.67
	Mexico	10	0.06	ND[a]-0.53
cloves	Madagascar	10	0.17	ND-1.27
coriander	Romania	3	0.13	0.02-0.09
	Morocco	4	0.04	0.01-0.02
cumin	Iran	6	0.01	0.01-0.02
dill seed	India	3	0.14	0.14-0.14
fennel	India	11	0.51	0.01-3.52
ginger	India	8	0.29	0.02-1.19
	Nigeria	1	0.01	
mace	Indonesia	3	0.01	<0.005-0.03
marjoram	Egypt	3	0.06	0.03-0.07
	France	5	0.06	0.01-0.11
nutmeg	Indonesia	1	0.02	
oregano	Mexico	3	0.10	0.06-0.18
	Greece	6	0.09	0.02-0.27
poppy seed	Holland	4	<0.005	
rosemary	Yugoslavia	5	0.04	0.01-0.13
	Spain	3	0.05	0.04-0.07
sage	Albania	9	0.23	0.04-0.65
	Yugoslavia	5	0.06	0.03-0.11
sesame	Guatemala	4	<0.005	
	Mexico	2	<0.005	
tarragon	France	3	0.04	0.04-0.05
thyme	Spain	3	0.04	0.01-0.07
	France	3	0.09	0.09
turmeric	India	4	0.28	0.04-0.60
white pepper	Brazil	6	0.05	0.01-0.21
	Malaysia	3	<0.005	

[a] ND = nondetectable.

plicable to peppers in the United States. Previously, the regulated tolerance was 5 ppm. These detentions led us into a major project to evaluate the need for BHC application in Spain and the definition of appropriate residue levels. Subsequently, a petition for a tolerance of 5 ppm in dehydrated peppers (paprika) was submitted and published in the *Fed. Regist.* (1975) by the Environmental Protection Agency (EPA).

With spices other than paprika, little information has been published. In 1974, Lutomski et at. (1974) reported levels of DDT up to 0.47 ppm on eight spices and herbs including allspice, basil, and black pepper. Gerhardt and Roth (1975) found a maximum of 0.19 ppm of DDT and also detected Lindane on eight major spices while phosphoric acid esters were not detected. Corvi and Vogel (1976) and Illes et al. (1976) reported similar levels of the chlorinated hydrocarbons in several herbs and confirmed the absence of organophosphate residues.

Based on this, we established a program to survey all spices from all major source countries for the purpose of identifying potential problem item source countries with respect to residues of the chlorinated hydrocarbons.

PROCEDURE

Since we are concerned with 28 items from some 25 source countries, many with different growing seasons and each with its own agricultural practices and shipping schedules, our samples were taken from several selected shipments of each item/source received during the entire shipping season for a 2–3-year period. The survey encompassed 226 individual samples.

Samples representing a shipment as imported were taken at the wharf. This was begun in Oct 1973 and continued through April 1978. All the analyses were done by the Wisconsin Alumni Research Foundation using the method for pesticide residues published by FDA. This is a widely used procedure which after isolation and purification with Florisil columns uses a gas chromatographic assay. For these analyses, a Barber Coleman Model 5380 pesticide analyzer equipped with a ^{63}Ni electron capture was employed.

Some items, but not all, were checked for recovery and

Table III. Residues of PCB, Dieldrin, Endrin, and HCB (ppm Average)

item	source	N	est PCB	Dieldrin	Endrin	HCB
allspice	Guatemala	6	<0.010	<0.005	<0.005	<0.005
	Jamaica	6	<0.010	<0.005	<0.005	<0.005
anise	Spain	6	<0.010	<0.005	<0.005	<0.005
basil	Hungary	5	<0.010	<0.005	<0.005	<0.005
	France	3	ND[a]	<0.005	0.16	<0.005
bay leaves	Turkey	6	ND	<0.005	<0.005	<0.005
black pepper	Brazil	5	<0.05	<0.005	ND	ND
	Indonesia	5	<0.01	<0.005	ND	ND
	Malaysia	5	ND	0.01	ND	ND
	India	4	ND	<0.005	<0.005	ND
caraway	Poland	3	ND	<0.005	<0.005	<0.005
	Holland	4	<0.01	<0.005	ND	<0.005
cardamom	Guatemala	2	<0.01	0.03	<0.005	<0.005
cassia	China	6	0.04	<0.005	<0.005	ND
	Indonesia	3	0.01	<0.005	<0.005	ND
	Seychelles	5	ND	<0.005	<0.005	<0.005
celery seed	India	11	<0.010	<0.005	0.01	<0.005
chillies	Japan	3	<0.01	<0.005	<0.005	ND
	Turkey	3	ND	0.01	<0.005	ND
	China	4	0.03	ND	ND	ND
	Mexico	10	<0.010	0.03	0.03	<0.005
cloves	Madagascar	10	<0.010	<0.005	<0.005	<0.005
coriander	Romania	3	ND	<0.005	<0.005	<0.005
	Morocco	4	ND	<0.005	0.10	0.01
cumin	Iran	6	ND	<0.005	<0.005	<0.005
dill seed	India	3	ND	<0.005	<0.005	<0.005
fennel	India	11	<0.01	<0.005	<0.005	<0.005
ginger	India	8	<0.01	<0.005	<0.005	<0.005
	Nigeria	1	ND	0.012	ND	ND
mace	Indonesia	3	<0.01	0.01	<0.005	<0.005
marjoram	Egypt	3	ND	0.020	0.04	<0.005
	France	5	0.03	<0.005	<0.005	<0.005
nutmeg	Indonesia	1	0.39	0.019	ND	ND
oregano	Mexico	3	ND	<0.005	ND	ND
	Greece	6	ND	<0.005	<0.005	0.02
poppy seed	Holland	4	ND	<0.005	<0.005	<0.005
rosemary	Yugoslavia	5	ND	<0.005	<0.005	<0.005
	Spain	3	ND	<0.005	<0.005	<0.005
sage	Albania	9	0.08	<0.005	<0.005	<0.005
	Yugoslavia	5	0.29	<0.005	ND	<0.005
sesame	Guatemala	4	ND	<0.005	<0.005	ND
	Mexico	2	ND	<0.005	<0.005	<0.005
tarragon	France	3	<0.01	<0.005	<0.005	0.01
thyme	Spain	3	0.01	ND	ND	0.02
	France	3	ND	<0.005	<0.005	<0.005
turmeric	India	4	0.02	ND	<0.005	ND
white pepper	Brazil	6	ND	<0.005	ND	ND
	Malaysia	3	ND	<0.005	ND	ND

[a] ND = nondetectable.

interfering substances. In general the recoveries ranged from 75 to 100%.

RESULTS AND DISCUSSION

The results of the analysis for DDT summarized in Table I reflect the sum of the DDT residues. With the exception of oregano from Mexico, all but three average below 0.5 ppm and only an additional eight exceed 0.5 ppm at the upper limit of the range.

Basil from Hungary and red pepper from Turkey, which average 0.58 and 0.81 ppm, respectively, appear to do so consistently. With the red peppers from China, three of the four lots were below 0.5 ppm, while one was 2.64 ppm. The present tolerance for DDT on peppers is 7 ppm.

For those eight items which exceeded 0.5 ppm at the upper limit of the range, in each set of samples one or two are abnormally high as compared with the others. With allspice from Guatemala, for example, five lots gave a maximum result of 0.42 ppm (range 0.03–0.42 ppm), while one registered 0.96 ppm. Similarly, five samples of white pepper from Brazil were below 0.14 ppm while one contained 1.14 ppm.

The relatively high and consistent values in excess of 1.0 ppm for oregano from Mexico strongly suggest that an active program with DDT is underway in that country.

Since an intensive investigation was beyond the objectives of this survey, a separate program was undertaken in cooperation with growers in Mexico. Our suspicions were confirmed to the extent that DDT is used in some of the areas where oregano is grown for mosquito control, and there is some incidental and unavoidable contamination of the oregano. This program resulted in the submission of a petition to EPA to establish a tolerance of 5 ppm of DDT on oregano from Mexico. The detection of these higher levels of confirmation of actual use of DDT in Mexico adds confidence to the validity of the sampling plans and analyses for all spices.

Residues of BHC, summarized in Table II, appear to follow the same general pattern as DDT. Only one item, fennel seed from India, exceeds an average of 0.50 ppm of total BHC, and that is due entirely to one of the eleven samples which contained 3.52 ppm. The remainder ranged

Table IV. Average Residues (ppm) of DDT and BHC by Source

source	item	DDT	BHC
Albania	sage	0.32	0.23
Brazil	black pepper	0.23	0.01
	white pepper	0.41	0.01
China	red pepper	0.81	0.34
	cassia	0.12	0.24
Egypt	marjoram	0.12	0.06
France	tarragon	ND[a]	0.04
	marjoram	0.13	0.06
	basil	0.25	0.08
	thyme	0.11	0.09
Greece	oregano	0.08	0.09
Guatemala	allspice	0.38	0.04
	cardamom	0.03	<0.005
	sesame	0.15	<0.005
Holland	poppy seed	<0.005	<0.005
	caraway	0.01	<0.005
Hungary	basil	0.62	0.05
India	black pepper	0.10	0.11
	celery seed	0.43	0.37
	fennel seed	0.17	0.62
	ginger	0.28	0.29
	dill seed	0.49	0.14
	turmeric	0.03	0.28
Indonesia	black pepper	0.11	0.01
	cassia	0.04	0.02
	mace	0.03	0.01
	nutmeg	0.02	0.02
Iran	cumin	0.03	0.01
Jamaica	allspice	0.02	0.03
Japan	red pepper	0.01	0.01
Madagascar	cloves	0.32	0.17
Mexico	oregano	1.71	0.10
	red pepper	0.15	0.06
	sesame	0.02	<0.005
Malaysia	white pepper	ND	0.01
	black pepper	0.25	0.19
Morocco	coriander	0.15	0.04
Nigeria	ginger	0.08	0.01
Poland	caraway	0.23	0.01
Romania	coriander	0.05	0.13
Seychelles	cassia	0.03	0.01
Spain	thyme	0.08	0.04
	anise	0.04	0.12
	rosemary	0.11	0.05
Turkey	bay leaf	0.06	0.03
	red pepper	0.81	0.21
Yugoslavia	sage	0.12	0.06
	rosemary	0.09	0.04

[a] ND = nondetectable.

from 0.01 to 0.48 ppm.

An additional six items exceeded 0.5 ppm at the top of the range, but, as with DDT, an individual result or two was substantially higher than the others. For example, nine shipments of cloves from Madagascar ranged from a nondetectable level to 0.19 ppm, while one sample contained 1.27 ppm.

Results of the analysis for Dieldrin, Endrin, and hexachlorobenzene (HCB) and an estimate of polychlorinated biphenyl (PCB) are tabulated in Table III. Except for an infrequent detection, such as 0.16 ppm of Endrin in basil from France, most are below the sensitivity of the method.

An examination by country of origin (Table IV) does not suggest that residues occur more frequently or at a higher level in spice products from one country over another. As discussed previously, oregano from Mexico is higher than other sources in DDT residues but this is not reflected in two other items grown in Mexico. Similarly, there is a need for a 5 ppm of BHC tolerance on paprika from Spain but three other crops, thyme, anise, and rosemary, do not appear to have the same requirement.

SUMMARY AND CONCLUSIONS

While the presence of chlorinated hydrocarbons, particularly DDT and BHC, can be detected on spices imported into the United States, the levels in general are sufficiently low to be of no cause for alarm. Occasionally greater than normal levels are detected. Sampling variability aside, this may very well stem from the accidental exposure during the complex warehousing/distribution chain which spices undergo before they are imported into the United States.

The consistent detectability of these pesticides is also of interest. The persistent nature of these pesticides is, of course, well-known, and it is quite possible their presence in spices is the result of previous unrelated applications.

In addition, since our purpose was to conduct a general survey to identify potential problem areas, confirmatory analyses were not done with all items. The complex composition of the essential oils contained in spices may very well interfere with the analytical method despite the lengthy cleanup procedures and the selective gas chromatographic detectors. However, in view of the low levels found, it did not appear that extensive methods development work would be justified.

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