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Organochlorine Pesticide Residues in Black Tea, Camomile, and Linden

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Organochlorine pesticides still exist as pollutants in countries like Spain, despite the limit which was put on their use more than ten years ago. These compounds are highly persistent and they are capable of spreading stuck to dust and rain particles. The purpose of the present study is to give some data on the levels of organochlorine pesticides in infusion plants (black tea, camomile and linden). These infusion plants are taken by a lot of people, of different ages and social classes, so it is important to know the risk that their consumption supposes to health.

MATERIAL AND METHODS

A total of 66 samples of infusion bags (black tea, camomile and linden) were obtained from different supermarkets in León, Spain. The bags selected for analysis were from 4 of the most popular makes in Spain, and they expired in 1992, 1993, 1994, 1995 and 1996. We were not able to find a sample from each year as some of the makes had not yet produced them. The weight of the bags ranged from 1 to 2 grams. We used the whole content of the bag for analysis. Two bags of each expiry date were analyzed.Subsequent extraction and clean-up were carried out according to methods described by Richardson *et al.* (1971) and Sthar (1977). A column containing Florisil and anhydrous sodium sulphate conditioned for 24 hrs. at 130°C was used to remove interfering substances and to separate the pesticides in two fractions.

The clean-up extracts were completely evaporated under a gentle stream of nitrogen; 0,5 ml of hexane were added for reconstitution. The analysis was carried out by electron capture gas chromatography using a Hewlett Packard gas- chromatograph fitted with a Ni^{63} electron capture detector and a computer. The operating conditions were as follows: Columns were 6 feet long and 1/4 inch inside diameter containing: a) a 1,5 % coating of OV-17/1,95 % of QF-1on 100-120 mesh Chromosorb W(AW/DMCS) and b) 3,8% SE-30 on 80-100 mesh Chromosorb W(AW/DMCS).The operating temperatures were: inlet 250°C; detector 300°C; column a)200°C and column b)175°C. The carrier gas was 5% argon-methane at a flow rate of 23 ml/min and sample volume 3 µl.

The samples were tested for the residues of organochlorine pesticides: αHCH, βHCH, lindane, heptachlor-epoxide, aldrin, dieldrin, endrin, o,p'-TDE, p,p'-TDE, p,p'-DDE, and p,p'-DDT. Reference standards of these pesticides were used to identify and quantify the levels of residue.

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Recovery from fortified samples of plants were : 97,1 % for αHCH; 95,3 % for βHCH; 87,2 % for lindane; 96,9 % for heptachlor-epoxide; 98,4 % for aldrin; 90,2 % for dieldrin; 90,3 % for endrin; 82,4 % for o,p'-TDE; 84,4 % for p,p'-TDE; 88,4 % for p,p'-DDE and 88,5 % for p,p'-DDT.

RESULTS AND DISCUSSION

The mean residue levels and ranges of organochlorine pesticides in the three types of samples (black tea, camomile and linden), are summarized in Tables 1, 2 and 3, expressed in parts per billion (ppb) by weight of sample as received. The incidence percentage of organochlorine pesticides is also summarized in the same Tables.

In order to compare the results obtained with the maximum levels allowed by the EEC in black tea and similar substances (published in Spain in the BOE-4th November 1989), we have included the values of Σ HCH, Σ dieldrin and Σ DDT in Figures 1, 2 and 3. Σ HCH means the addition of αHCH and βHCH residues. Likewise, Σ dieldrin means the amount of aldrin plus dieldrin and ΣDDT refers to the addition of o,p'-TDE, p,p'-TDE, p,p'-DDE and p,p'-DDT.

None of the analyzed samples exceeds the maximum levels established by the EEC (0.01 ppm for Σ HCH; 0.5 ppm for lindane; 0.1 ppm for heptachlor; 0.1 ppm for Σ dieldrin; 0.1 ppm for endrin and 1 ppm ΣDDT) for tested pesticides.

In spite the fact that none of them exceeds the allowed limits, we want to point out the high percentage incidence of αHCH, which is present in all the samples analyzed. We have also obtained a high percentage incidence for αHCH in other biological samples (organs and tissues from raptors and trouts) analyzed by us in earlier studies (Sierra *et al.* 1984; 1987; Sierra and Santiago 1987; Terán and Sierra 1987). The frequences of βHCH and p,p'-DDT were also considerable in the three types of samples analyzed, as well as heptachlor-epoxide in black tea and linden samples.

We have not found studies on the percentage incidence and levels of organochlorine pesticides in tea and similar substances in Spain. Our results agree with those found by Buchholz in Germany, in 1988 in a study carried out on black tea from various regions. The author reports that the major contaminants were HCHs and DDT and its isomers and metabolites particulary in Chinese teas.

Likewise, Niimura *et al.* (1989) found a high incidence of HCHs in tea leaves in Japan. We do not have any data about organochlorine residues in camomile and linden.

In relation to residue levels, in general, camomile and linden samples were more contaminated than black tea samples. The most contaminated camomile samples were those that expired in 1996, with DDT levels of 260.3 ppb and aldrin levels of 78.3 ppb. This fact can indicate recent direct application of aldrin.

The highest levels in linden samples were found in samples that expired in 1993, where 352.8 ppb of lindane, 51.0 ppb of o,p'-TDE and 27.8 ppb of αHCH appeared.

Our levels are clearly lower than those found by De Silva and Thiemann, 1991

Table 1. Average, range (ppb/wet weight) and incidence percent of organochlorine pesticides in black tea.

	1992 [b]	1993 [d]	1994 [a]	1995 [b]	1996 [a]
1	11.8 1.8-25.8 100	20.4 4.5-37.4 100	14.4 11.4-17.4 100	10.2 7.5-13.5 100	8.1 4.4-11.7 100
2	54.0 33.1-78.6 100	29.9 21.3-45.9 62.5	53.6 44.4-62.8 100	34.1 1.7-93.8 100	65.8 56.6-75.0 100
3	36.4 5.9-66.8 50	347.8 53.9-651.6 25	195.9 168.6-223.4 100	15.5 15.5 25	11.7 11.7 50
4	--- --- 0	27.7 4.7-67.5 75	10.5 7.7-13.4 100	24.4 7.1-42.8 75	6.9 6.9 50
5	10.2 10.2 25	4.7 4.7 12.5	--- --- 0	12.7 12.6-12.7 50	--- --- 0
6	5.3 5.3 25	0.55 0.07-1.0 25	--- --- 0	--- --- 0	--- --- 0
7	--- --- 0	--- --- 0	0.19 0.19 50	--- --- 0	--- --- 0
8	--- --- 0	29.7 8.5-73.2 37.5	--- --- 0	7.9 7.8-8.0 50	6.3 6.3 50
9	--- --- 0	10.4 3.6-19.3 50	--- --- 0	6.0 1.8-10.3 50	8.0 3.0-13.1 100
10	3.3 1.8-4.9 50	4.0 2.2-6.8 62.5	7.5 5.8-9.2 100	8.3 7.6-9.0 50	11.1 11.1 50
11	7.4 1.3-13.5 75	6.5 2.1-9.3 37.5	29.9 9.7-50.0 100	5.3 2.2-9.1 75	163.5 21.1-305.9 100

1: αHCH; 2: βHCH; 3: Lindane; 4: Heptachlor-epoxide; 5: Aldrin; 6: Dieldrin; 7: Endrin; 8: o,p'-TDE; 9: p,p'-TDE; 10: p,p'-DDE; 11: p,p'-DDT. a: one make analyzed; b: two makes analyzed; d: four makes analyzed.

in Sri Lanka tea and similar to those reported by Niimura *et al.*, 1989 for BHC (0.002-0.375 ppm), p,p'-DDT (0.036-0.140 ppm) and p,p'-DDE (0.003-0.056 ppm).

Table 2. Average, range (in ppb/wet weight) and incidence percent of organochlorine pesticides in camomile.

	1992 [c]	1993 [c]	1994 [c]	1995 [c]	1996 [a]
1	6.7 2.7-9.1 100	8.3 0.35-24.4 100	10.3 5.8-12.6 100	9.2 6.5-12.9 100	23.4 14.7-32.1 100
2	3.8 1.4-8.6 83.3	1.2 0.06-2.3 100	13.3 1.1-19.4 66.7	7.8 0.67-12.4 66.7	--- --- 0
3	8.6 5.9-11.3 50	10.5 10.5 16.7	32.9 31.6-34.1 33.3	--- --- 0	--- --- 0
4	22.0 22.0 16.7	--- --- 0	--- --- 0	6.1 6.1 16.7	--- --- 0
5	15.8 0.38-39.5 66.7	10.7 8.1-13.2 33.3	3.6 3.6 16.7	5.9 5.6-6.2 33.3	78.3 78.3 50
6	--- --- 0	0.22 0.22 16.7	--- --- 0	--- --- 0	--- --- 0
7	--- --- 0	--- --- 0	6.1 6.1 16.7	--- --- 0	--- --- 0
8	12.7 6.1-16.2 66.7	7.0 3.3-9.6 83.3	9.3 2.5-15.2 50	10.5 2.0-16.3 83.3	17.4 17.4 50
9	3.7 0.79-15.5 100	--- --- 0	2.5 1.2-3.8 33.3	3.4 3.0-3.9 33.3	--- --- 0
10	5.7 2.1-9.3 50	3.9 3.7-4.0 50	21.9 4.8-34.7 50	3.8 1.0-6.6 33.3	4.5 4.5 50
11	10.0 0.14-22.0 83.3	4.7 1.5-7.9 50	42.7 10.2-75.2 33.3	26.4 8.0-10.5 66.7	270.3 218.6-321.9 100

1: αHCH; 2: βHCH; 3: Lindane; 4: Heptachlor-epoxide; 5: Aldrin; 6: Dieldrin; 7: Endrin; 8: o,p'-TDE; 9: p,p'-TDE; 10: p,p'-DDE; 11: p,p'-DDT. a: one make analyzed; c: three makes analyzed.

This study shows that contamination by organochlorine pesticides is low in the infusion plants sutied by us (black tea, camomile and linden).

Table 3. Average, range (in ppb/wet weight) and incidence percent of organochlorine pesticides in linden.

	1992 [a]	1993 [d]	1994 [b]	1995 [b]	1996 [a]
1	1.9 1.8-1.9 100	2.8 1.0-9.8 100	3.9 3.0-5.2 100	15.4 3.5-39.9 100	6.4 1.6-11.2 100
2	1.1 1.1-1.2 100	4.2 0.29-10.6 100	21.1 1.8-40.5 50	9.4 5.7-13.2 50	15.4 15.4 50
3	3.0 3.0 50	6.4 1.6-16.8 50	--- --- 0	9.1 4.3-23.4 75	13.6 13.6 50
4	1.7 1.7 50	13.0 1.9-43.1 62.5	8.1 2.8-18.4 100	16.8 7.9-36.7 100	--- --- 0
5	1.0 0.37-1.7 100	2.4 1.4-3.6 50	2.8 2.3-3.4 50	11.3 11.3 25	--- --- 0
6	--- --- 0	--- --- 0	--- --- 0	--- --- 0	--- --- 0
7	--- --- 0	--- --- 0	0.66 0.66 50	--- --- 0	--- --- 0
8	5.0 2.9-7.1 100	3.3 3.3 25	4.6 3.4-5.8 50	8.7 3.0-13.6 75	16.7 16.7 50
9	--- --- 0	2.6 0.10-7.1 50	0.06 0.06 25	1.5 0.51-2.2 50	7.1 2.0-12.1 100
10	--- --- 0	4.8 0.83-6.9 62.5	2.0 1.0-3.0 50	8.0 8.0 25	2.0 2.0 50
11	4.7 4.7 50	4.8 0.25-10.2 87.5	45.7 2.2-89.1 50	14.2 10.0-18.3 50	--- --- 0

1: αHCH; 2: βHCH; 3: Lindane; 4: Heptachlor-epoxide; 5: Aldrin; 6: Dieldrin; 7: Endrin; 8: o,p'-TDE; 9: p,p'-TDE; 10: p,p'-DDE; 11: p,p'-DDT. a: one make analyzed; b: two makes analyzed; d: four makes analyzed.

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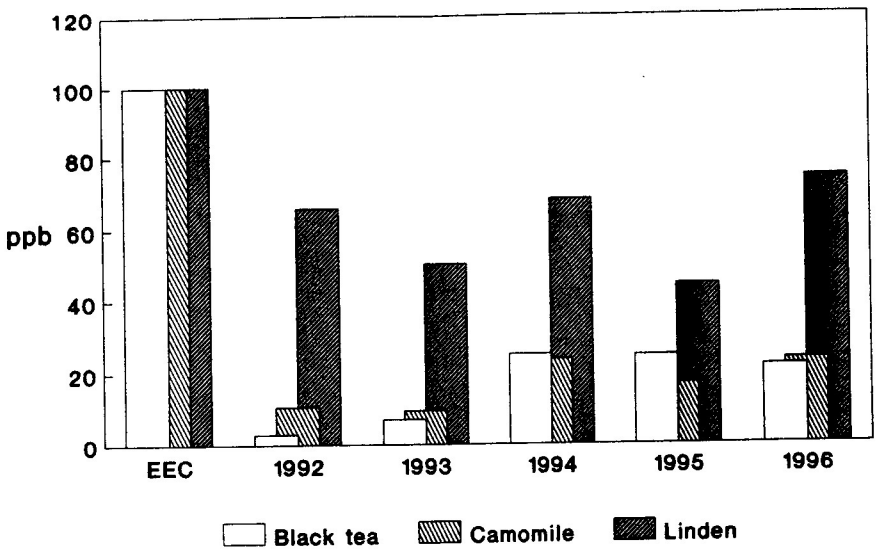

Figure 1. Average of ΣHCH detected in black tea, camomile and linden compared to EEC limits.

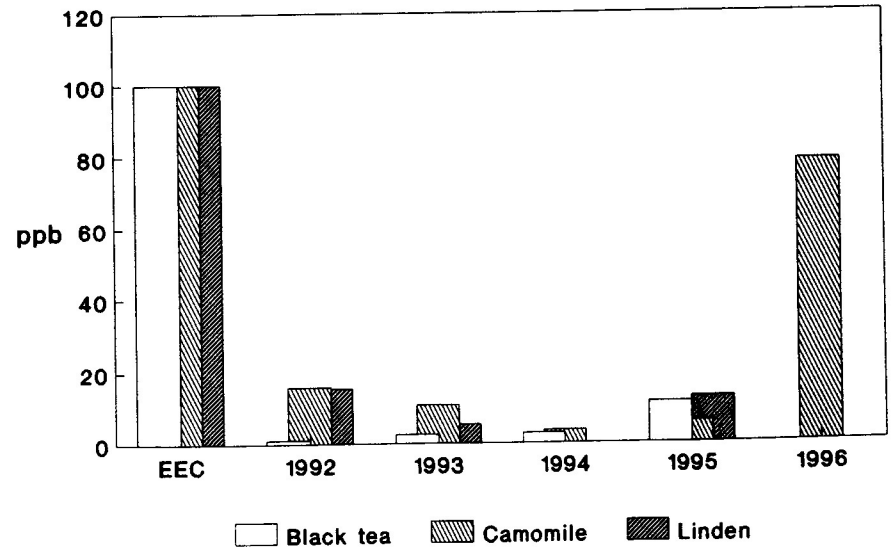

Figure 2. Average of Σdieldrin detected in black tea, camomile and linden compared to EEC limits.

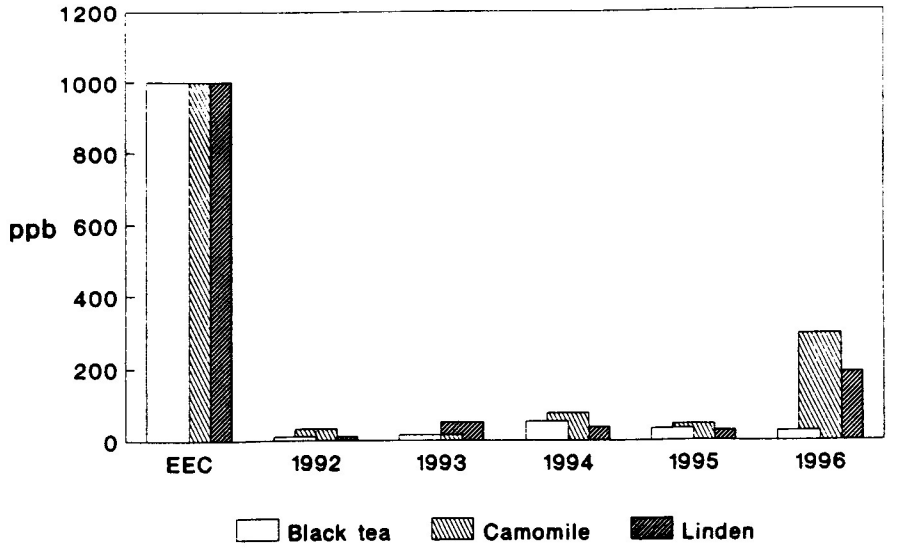

Figure 3. Average of ΣDDT detected in black tea, camomile and linden compared to EEC limits.

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