

DDT and HCH Residues in the Blood Serum of Malaria Control Sprayers

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The widespread application of organochlorine pesticides for controlling agricultural pests has brought serious concern about the effects of these compounds on human health. The use of DDT and HCH is still permitted in Brazil for endemic disease vector control. These pesticides are widely recognized as neurotoxic substances, affecting the peripheral and central nervous system, respectively, and causing a hyper-excitability of nerves and muscles (Hassal 1983). In a recent work, Fleming et al. (1994) found p,p'-DDE, a metabolite of DDT, in the majority of postmortem brain samples from neurological disease cases. Among the population exposed to pesticides, the sprayers are subject to the highest risks due to insufficient knowledge about the long term toxic effects and carelessness in handling the products.

Residues of organochlorines have been detected in the blood of both exposed workers (Griffith and Duncan 1985, Mazzarri B. and Mazzarri de Lauschner 1989, Bouwman et al. 1991, Chandra et al. 1992) and unexposed people (Leoni et al. 1989, Sasaki et al. 1991, Lommel et al. 1992) all over the world. However, few studies on organochlorine pesticides have been carried out in Brazil (Fernicola and Azevedo 1982; Lara et al. 1987, Carvalho 1991). In the present study, the serum levels of DDT and HCH of exposed workers, engaged in intradomiciliary spraying for the vector control program, and unexposed workers, were determined to assess the extent of long term occupational exposure to these pesticides.

MATERIALS AND METHODS

Twenty-six workers engaged in spraying DDT and HCH for controlling disease carrying vectors in the city of São José do Rio Preto, in the State of São Paulo, Brazil, were selected for serum analyses. Sixteen unexposed workers were also selected for comparison with exposed workers. Their ages and working years are shown in Table 1.

Blood was collected by venipuncture and the serum was separated. Serum samples were stored frozen at - 18° C until analysis. The samples were analyzed using a method previously described (Minelli and Ribeiro 1996). The method involved:

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pesticide extraction of a serum silica suspension with n-hexane:acetone 9:1, extract purification on alumina column chromatography, and analysis using gas-liquid chromatography with electron capture detection.

All solvents used were of pesticide grade quality (Merck) and checked for interfering residues by GC after a forty fold concentration. Silica gel 60 (Merck, 70-230 mesh ASTM) and alumina 90 neutral (Merck, 70-230 mesh ASTM) were treated as described by Minelli and Ribeiro (1996). Reference standards of all pesticides used were provided as gifts from the U.S. Environmental Protection Agency, Research Triangle Park, NC. Standard solutions were made up in isoctane and stored at - 18°C.

Chromatographic analyses were carried out using a Varian 3300 gas chromatograph equipped with a glass column (2 m x 2 mm id.) packed with 1.5% OV-17 / 1.95% QF-1 on 80-100 mesh Chromosorb W-AW-DMCS, and a constant current ⁶³Ni electron capture detector (ECD). The signal from the ECD was monitored with a Varian 4290 Integrator. Operating conditions were as follows: injector temperature, 230°C; column oven temperature, 200°C; detector temperature, 300°C; and carrier gas (N₂) at 30 mL/min. Qualitative analyses were performed on a CG 35370 gas chromatograph equipped with a glass column (1.8 m x 2 mm id.) packed with 5% QF-1 on 80-100 mesh Chromosorb WHP, and a constant current ⁶³Ni electron capture detector. Operating conditions were as follows: injector temperature, 210°C; column oven temperature, 188°C; detector temperature, 260°C; and carrier gas (N₂) at 40 mL/min.

Table 1. Age and years of work of the groups studied

GROUP	Age (years)		Years of work	
	Range	Mean ± SD	Range	Mean ± SD
Exposed (n = 26)	50 - 65	56 ± 4.1	22 - 35	30 ± 2.9
Unexposed (n = 16)	42 - 69	54 ± 8.8	8 - 38	20 ± 8.2

RESULTS AND DISCUSSION

Serum levels of organochlorine residues for the exposed and unexposed workers are shown in Tables 2 and 3, respectively. Mean concentrations and range values of the total DDT and total HCH are presented in Table 4.

Table 2. Pesticide levels in serum of exposed workers

Sample	Concentration (µg/L)					
	α-HCH	γ-HCH	β-HCH	p,p'-DDE	o,p'-DDT	p,p'-DDT
1	0.6	<0.2 ^a	6.3	26.2	1.0	8.2
2	0.6	0.3	13.3	45.5	nd ^b	1.6
3	0.6	<0.2	27.1	35.4	1.9	10.8
4	<0.2	<0.2	48.6	62.5	2.4	23.2
5	0.4	<0.2	19.6	74.7	0.7	5.8
6	<0.2	nd	23.2	47.2	1.2	16.8
7	<0.2	<0.2	12.2	25.2	nd	4.1
8	0.6	0.6	14.6	56.4	nd	3.3
9	0.3	<0.2	27.2	55.6	nd	8.8
10	<0.2	<0.2	15.3	31.6	nd	5.0
11	0.3	<0.2	114.8	40.9	2.5	19.6
12	0.7	<0.2	43.9	106.0	4.4	23.7
13	0.5	<0.2	12.7	33.0	nd	5.5
14	0.4	<0.2	40.3	41.3	1.3	10.8
15	0.2	<0.2	12.3	11.5	<0.7	2.5
16	0.3	<0.2	75.7	48.2	<0.7	5.2
17	0.4	<0.2	13.1	57.7	nd	4.9
18	0.9	<0.2	129.0	24.2	1.9	15.0
19	0.4	<0.2	58.2	405.9	4.7	62.9
20	0.3	0.2	7.0	5.9	nd	1.6
21	<0.2	<0.2	32.9	54.5	nd	4.4
22	0.8	<0.2	20.8	99.0	1.4	13.2
23	0.3	<0.2	17.1	72.1	1.3	15.4
24	0.5	0.5	11.8	39.2	<0.7	7.7
25	0.9	0.7	30.5	157.0	2.1	21.5
26	0.4	nd	3.4	14.7	nd	1.9

^a below the limit of detection^b not detected

Residues of six different pesticides were found at detectable levels in the serum of the exposed group. Eleven samples showed p,p'-DDD, but the levels were below the detection limit. In 62% and 92% of the samples, o,p'-DDT and γ-HCH were detected. All serum samples were contaminated with α-HCH, β-HCH, p,p'-DDE and p,p'-DDT at mean concentrations of 0.4, 31.8, 64.3 and 13.5 µg/L, respectively. The values obtained for β-HCH, p,p'-DDE, and p,p'-DDT were considerably higher than those found, at mean concentrations of 3.3, 14.3 and 1.5 µg/L, respectively, in the unexposed workers. Mean levels of α-HCH for both groups were not significantly different (0.4 and 0.3 µg/L).

The levels of HCH and DDT residues for the exposed workers were high when compared to those reported from other countries. Griffith and Duncan (1985)

Table 3. Pesticide levels in serum of unexposed workers

Sample	Concentration (µg/L)					
	α-HCH	γ-HCH	β-HCH	p,p'-DDE	o,p'-DDT	p,p'-DDT
1	0.6	< 0.2	2.5	5.1	nd	nd
2	0.3	< 0.2	3.1	21.1	nd	1.8
3	0.2	< 0.2	7.5	16.3	nd	1.7
4	0.4	< 0.2	2.0	13.6	nd	1.2
5	0.4	< 0.2	3.3	7.1	nd	1.6
6	0.4	< 0.2	5.5	18.4	nd	2.0
7	0.7	< 0.2	3.6	13.0	nd	1.6
8	0.3	0.3	4.2	16.5	nd	2.3
9	< 0.2	< 0.2	3.0	10.7	nd	1.0
10	< 0.2	< 0.2	3.5	14.0	nd	2.5
11	0.3	< 0.2	3.3	14.4	nd	2.0
12	0.2	0.2	5.1	31.6	nd	1.3
13	0.3	0.2	3.4	15.6	nd	1.6
14	nd	nd	nd	12.2	nd	1.3
15	0.3	nd	2.1	12.0	nd	1.7
16	0.3	nd	1.6	7.0	nd	1.1

Table 4. Mean levels and ranges of total HCH and total DDT in serum of the two groups

	Exposed (n=26)		Unexposed (n= 16)	
	Mean* ± SD	Range	Mean* ± SD	Range
Total HCH	32.4 ± 31.5	3.8 - 129.9	3.7 ± 1.7	nd - 7.7
Total DDT	76.9 ± 89.1	7.5 - 473.5	16.1 ± 6.3	5.1 - 32.9

(*) non detectable and trace values excluded from calculations

reported HCH and DDT mean levels from 1.7 to 26.9 µg/L, in a study involving 567 Florida citrus fieldworkers. In a survey carried out in Honduras by Steinberg et al. (1989) β-HCH, p,p'-DDE and p,p'-DDT were found at mean concentrations of 0.44, 42.5 and 2.69 µg/L, respectively, in the serum of the exposed group. Chandra et al. (1992) observed mean serum levels of total HCH and total DDT of 15.9 and 4.4 µg/L among 52 sprayers from mango groves in Malihabad, India. As reported by Rosell et al. (1993), γ-HCH and p,p'-DDT were found at mean concentrations of 0.021 and 3.63 µg/L, respectively, in the serum of 71 agricultural workers from Spain.

However, serum DDT levels in exposed workers from São José do Rio Preto were much lower than those reported by Mazzarri B. and Mazzarri de Lauschner (1989) for malaria control sprayers from Venezuela, and for workers engaged in the vector control program of the State of Bahia, Brazil (Carvalho 1991). It was also

observed that these levels are four times lower than those reported for residents of areas surrounding a DDT manufacturing factory in Delhi (Saxena et al. 1987).

The unexposed group presented organochlorine levels much lower than those observed in samples from the general population in Ahmedabad, India (Bhatnagar et al. 1992, Kashyap et al. 1993) but were similar to the results reported for samples from Pakistan (Krawinkel et al. 1989). The concentrations of β -HCH and p,p'-DDE were found to be higher than those reported from developed countries such as Italy (Leoni et al. 1989) and Japan (Sasaki et al. 1991) but were comparable to the results reported by Lommel et al. (1993) for Elb River residents, Germany.

The occurrence of p,p'-DDE in all the samples and β -HCH in the majority of the samples of the unexposed population may be due to the widespread use in Brazil of DDT and HCH for crop protection, prior to the establishment, in 1985, of restriction laws. As was reported earlier in Brazil by Carvalho (1991), we also found that the distribution of HCH isomers in serum occurs in the following order: β -HCH > α -HCH > γ -HCH. In the present study it should be noted that p,p'-DDE is the long lasting residue of DDT due to occupational exposure or food contamination. These results are in good agreement with the report by Morgan et al. (1980) which indicates that these compounds are slowly excreted by the body, and also clearly indicates the bio-accumulation of these pesticides in the human body.

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