

Serum levels of organochlorine pesticides and polychlorinated biphenyls among inhabitants of Greater Metropolitan Rio de Janeiro, Brazil

Isabella Fernandes Delgado ¹
 Heloisa H.C. Barretto ²
 Teresa A. Kussumi ²
 Irene Baptista Alleluia ³
 Cenira de A. Baggio ³
 Francisco José Roma Paumgarten ¹

Níveis séricos de pesticidas organoclorados e bifenilos policlorados em habitantes da Área Metropolitana do Rio de Janeiro, Brasil

¹ Laboratório de Toxicologia Ambiental, Escola Nacional de Saúde Pública, Fundação Oswaldo Cruz, Rua Leopoldo Bulhões 1480, Rio de Janeiro, RJ 21041-210, Brasil.
² Instituto Adolfo Lutz, Av. Dr. Arnaldo 355, São Paulo, SP 01246-000, Brasil.
³ Instituto Nacional de Tecnologia, Av. Venezuela 82, Rio de Janeiro, RJ 20081-310, Brasil.

Abstract Levels of persistent organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) were determined in the blood serum of people living and working in the urban area of greater Rio de Janeiro city. Blood samples from 33 volunteers (16 males, 17 females, 19-63 years old) were taken in January 1999. OCP residues (op'DDT, pp'DDT, pp'DDD, pp'DDE, Aldrin, Dieldrin, Endrin, Heptachlor, Heptachlor-epoxide, α -, β - and γ -Hexachlorocyclo-hexane, Hexachlorobenzene) and PCBs (congeners: 28, 52, 101, 138, 153, 180) were extracted with *n*-hexane and analyzed by gas chromatography with electron capture detection. Except for pp'DDE (detection limit = 1.4 μ g/L) no other OCP residue was found in the samples. No PCB congener (detection limit = 2.0 μ g/L) was detected either. pp'DDE was found in 17 out of 33 samples in concentrations that ranged from 1.4 to 8.4 μ g/L of serum or, on a fat basis, from 0.200 to 3.452 μ g/g of serum lipids. Percentage of positive samples (%) and levels of pp'DDE (range of positive samples) increased from the youngest to the oldest group (\leq 29 yrs: 10%, 0.278 μ g/g; 30-39 yrs: 60%, 0.200-0.765 μ g/g; \geq 40 yrs: 77%, 0.257-3.452 μ g/g).

Key words Organic Chemicals; DDT; Polychlorinated Biphenyls; Endocrine Disruptors

Resumo Os níveis de pesticidas organoclorados persistentes (POPs) e de bifenilos policlorados (PCBs) foram determinados no soro sanguíneo de pessoas que vivem e trabalham na área urbana do grande Rio de Janeiro. Amostras de sangue foram coletadas de 33 voluntários (16 homens, 17 mulheres, idade: 19-63 anos) em Janeiro de 1999. Resíduos de POPs (op'DDT, pp'DDT, pp'DDD, pp'DDE, Aldrin, Dieldrin, Endrin, Heptacloro, Heptacloro-epóxido, α -, β - and γ -Hexaclorociclo-hexano, Hexaclorobenzeno) e PCBs (congeneres: 28, 52, 101, 138, 153, 180) foram extraídos com *n*-hexano e analisados em cromatógrafo gasoso com detector de captura de elétrons. Com exceção do pp'DDE (limite de detecção = 1,4 μ g/L), não foram encontrados outros resíduos de POPs nas amostras. Não foram encontrados também, congêneres de PCBs (limite de detecção = 2,0 μ g/L). O pp'DDE foi encontrado em 17 das 33 amostras em concentrações que variavam de 1,4 a 8,4 μ g/L de soro ou, em termos da gordura, de 0,200 a 3,452 μ g/g de lipídios séricos. A percentagem de amostras positivas (%) e os níveis de pp'DDE (intervalo de concentrações das amostras positivas) aumentaram do grupo etário mais jovem para o mais idoso (\leq 29 anos: 10%, 0,278 μ g/g; 30-39 anos: 60%, 0,200-0,765 μ g/g; \geq 40 anos: 77%, 0,257-3,452 μ g/g).

Palavras-chave Compostos Orgânicos; DDT; Bifenilos Policlorados; Desreguladores Endócrinos

Introduction

Organochlorine pesticides (OCPs), such as dichloro-diphenyl-trichloroethane (DDT), aldrin, dieldrin, hexachlorocyclohexane and others, were widely used in agriculture as well as in the control of arthropod-borne diseases from the end of the World War II until the early 1970s (Ecobichon, 1995). In the 1970s, the use of OCPs was phased out in most developed countries because of their environmental persistence and adverse effects on species which are on the top of food chains (Ecobichon, 1995). In Brazil, DDT and other persistent OCPs were banned for use in agriculture in 1985, but they remained in public health use until 1998 (Brazil, 1985, 1998).

Polychlorinated biphenyls (PCBs) have been produced commercially since the 1930s. They have been used as dielectrics in transformers and large capacitors, as ink solvent/carriers in carbonless copy papers, in hydraulic fluids and lubricants, in adhesives, in flame retardants, in plastics, and for several purposes in a variety of other products (WHO, 1993). Because most PCBs are quite resistant to biotic and abiotic degradation and present rather pronounced bioaccumulative properties, there have been increasing concerns on their deleterious effects on human health and wildlife. Owing to the foregoing concerns, the production and industrial uses of PCBs have been more and more reduced all over the world since the mid-1970s (WHO, 1993).

More recently, concerns on the possible health hazards posed by the presence of endocrine disrupting chemicals (EDCs) in the environment have led to a renewed interest in assessing today's exposure of the general population to OCPs and PCBs (Golden et al., 1998). Several OCPs including DDT and its most persistent metabolite DDE (or diphenyldichloroethylene) as well as many PCB congeners have been shown to possess estrogenic/anti-estrogenic and/or anti-androgenic activities in different biological assays (Golden et al., 1998; Kelce et al., 1995). Nevertheless, whether or not current levels of exposure to EDCs have a significant impact on human health is still a matter of controversy. Epidemiological studies performed during the last decade, for instance, have given conflicting results concerning a possible relation between exposure to OCPs and increased risk of breast cancer, a known estrogen-responsive neoplasia (Høyer et al., 1998; Laden et al., 2001; Romieu et al., 2000).

The objective of the present study was to provide data on the contamination of people

living and working in the urban area of greater Rio de Janeiro city by persistent OCPs and PCBs. It is part of a more comprehensive evaluation of the human exposure to organochlorine compounds in Brazil.

Methods

Thirty-three volunteers (16 males and 17 females) were recruited among people that worked at Oswaldo Cruz Foundation (FIOCRUZ) campus and had been living in the urban area of greater Rio de Janeiro city for at least five years in January 1999. All volunteers were asked to sign an Informed Consent form after having received a full explanation of the objectives and procedures to be followed in the study. Following a brief interview, blood samples (10ml) were drawn from an arm vein and collected in silicone-coated Vacutainer® tubes without anticoagulants. Immediately after clotting, serum was separated by centrifugation and serum samples were frozen (-18°C) and sent to the Adolfo Lutz Institute, São Paulo, where they were further analyzed for organochlorine pesticides and PCBs residues. Organochlorine residues were extracted with *n*-hexane (residue grade) and analyzed by gas chromatography with electron capture detection (Ni63) as previously described (Dale et al., 1970). Nitrogen (high purity grade) was the carrier gas at 30ml/min. Chromatographic parameters were as follows: column temperature: 60 to 190°C, 25°C/min, 190 to 280°C, 5°C/min, 280°C (2min), injector temperature: 250°C and detector temperature: 320°C. A capillary column (30m x 0.32mm x 0.25µm) with 5% phenyl methyl siloxane was used. The gas chromatograph (HP6890) was equipped with an automatic sampler. The limits for residue quantification which were validated for the present method were as follows: *op'*DDT, *pp'*DDT and *pp'*DDD = 2.8µg/L; *pp'*DDE, Aldrin, Dieldrin, Endrin, Heptachlor, Heptachlor-epoxide and β-Hexachlorocyclohexane (HCH) = 1.4µg/L; Hexachlorobenzene (HCB), α- and γ-HCH = 0.7µg/L; Polychlorinated biphenyls (PCB 28, PCB 52, PCB 101, PCB 138, PCB 153, PCB 180) = 2.0µg/L. All blood samples were taken and analyzed in January-February 1999.

Results and discussion

Polychlorinated biphenyls (PCBs)

PCBs residues (congeners 28, 52, 101, 138, 153 and 180) were not detected (detection limit = 2.0µg/L) in any of the serum samples analyzed in this study. As far as the authors are aware (MEDLINE and TOXLINE data bases) there is only one previous study on the levels of PCBs in people living in Rio de Janeiro city. In 1992, PCBs (congeners: 28, 52, 101, 138, 153, 180, 118, 156 and 157) were determined in a pooled sample of breast milk from 40 mothers (first or second lactation) and concentrations (total PCB values) as high as 0.15µg/g of milk fat were found (Paumgarten et al., 2000). Data from the present survey showing that serum levels of PCBs 28, 52, 101, 138, 153 180 are below 2.0µg/L, and results from the previous determination in the human milk (Paumgarten et al., 2000), both seem to indicate that people living in Rio de Janeiro have a body burden of PCBs lower than inhabitants of highly industrialized countries. Data from a large number of countries (in the 1970s and 1980s) summarized by World Health Organization (WHO, 1993) suggested that PCBs in human milk from industrialized countries ranged between 0.5 and 3.0µg/g milk fat, *i.e.* levels 3 to 20 times higher than that found in the sample of breast milk from Rio de Janeiro.

The general population is primarily exposed to PCBs by consuming contaminated fish and, to a lesser extent, via other food products from animal origin such as meat, milk, cheese, poultry and eggs (WHO, 1993). PCBs were detected in eight species of fish caught at Guanabara bay, Rio de Janeiro, in January-February 1999 (Ferreira-da-Silva, 2000). Contamination of fish muscle tissue by total-PCBs was rather low (from "not detected" up to 13.2ng/g wet weight) in sardines (*Sardinella brasiliensis*) and mild (up to 112 and 115ng/g wet weight) in "corvina" (*Micropogonias furnieri*) and "robalo" (*Centropomus undecimalis* and *C. parallelus*) (Ferreira-da-Silva, 2000). Except for Good Friday meals, however, fish and sea food are items that are only occasionally included in the diet in Rio de Janeiro. Besides this fact, according to data provided by Ferreira-da-Silva (2000), levels of PCBs were rather low in sardines, the cheapest and most consumed fish species.

Organochlorine pesticides (OCPs)

Except for *pp'*DDE, no other OCP residue was found in the blood serum samples taken from

this group of inhabitants of greater Rio de Janeiro city. As shown in Tables 1 and 2, *pp'*DDE was detected in the serum of 51.5 % of the individuals, *i.e.* in 56.2 % of the men and in 47% of the women who took part in the study. Concentrations of *pp'*DDE in the positive samples (detection limit = 1.4µg/L) ranged from 1.4 to 8.4µg/L or, on a fat basis, from 0.200 to 3.452µg/g of serum lipids (Table 1). The highest concentration of *pp'*DDE was found in the blood serum of volunteer No. 26, a female laboratory technician aged 41 (Table 1), who was reported to have had a six-month exposure (in mosquito control activities) to technical DDT in 1986. No other volunteer who took part in the present study informed us of being occupationally exposed to DDT or any other pesticide. As can be seen in Table 1, there is no apparent relation between the volunteer's present occupation and serum levels of *pp'*DDE. Neither the percentage of positive samples nor the serum levels of *pp'*DDE seemed to have differed between male and female participants (Table 2). It should be pointed out that our results clearly showed that serum concentrations of *pp'*DDE are age-dependent. The proportion of positive samples, as well as the median levels of *pp'*DDE were much higher in the oldest, (76.9%; 0.401µg/g lipids) than in the youngest (10%; not detected) age group. This trend was noted both in men (youngest group: 33.3%, not detected; oldest group: 71.4%, 0.407µg/g lipids) and in women (youngest group: 0 %, not detected; oldest group: 83.3%, 0.378µg/g lipids). Age dependency of *pp'*DDE blood serum concentrations has also been found in other studies with non-occupationally exposed populations (Paumgarten et al., 1998).

There are very few studies on the levels of exposure of inhabitants of Rio de Janeiro state, Brazil, to DDT and OCPs. In a previous survey of the serum concentrations of OCPs in agricultural workers from Paty-do-Alferes, Rio de Janeiro, the only residues found were β-HCH (in 6 out of 26 samples ranging from 1.4 to 5.3µg/L), dieldrin (in one out of 26 samples, 3.7µg/L) and *pp'*DDE (in 16 out of 26 samples, in 3 of them concentrations did not exceed 1.4µg/L, and in the remaining levels were as high as 1.8, 2.4 and 4.4µg/L) (Paumgarten et al., 1998). β-HCH and dieldrin were not detected in any of the samples of the present study. Levels of *pp'*DDE in people living and working in the urban area of greater Rio de Janeiro city, however, were somewhat higher than those observed in inhabitants of rural districts. At any rate, concentrations found in the present study as well as those determined in agricultural

Table 1

Levels of *pp'*DDE (per volume of serum – µg/L –, and on a fat basis – µg/g of serum lipids) in the blood of people living and working in greater Rio de Janeiro city, Rio de Janeiro, Brazil, January 1999.

Volunteer (No.)	Sex	Age (years)	City of residence	Occupation	<i>pp'</i> DDE in blood serum	
					µg/L	µg/g lipids
1	F	19	Rio de Janeiro	Service worker (cleaning)	ND	ND
2	F	21	Rio de Janeiro	Laboratory technician	ND	ND
3	F	22	Rio de Janeiro	Service worker (cleaning)	ND	ND
4	M	22	Niterói	Laboratory technician	1.5	0.278
5	F	23	Rio de Janeiro	Laboratory technician	ND	ND
6	M	23	Rio de Janeiro	Laboratory technician	ND	ND
7	F	23	Rio de Janeiro	Laboratory technician	ND	ND
8	F	24	Rio de Janeiro	Service worker (cleaning)	ND	ND
9	F	25	São Gonçalo	Telephone operator	ND	ND
10	M	25	Paracambi	Watchman	ND	ND
11	M	30	Quiemados	Watchman	ND	ND
12	F	30	Rio de Janeiro	Laboratory technician	1.4	0.330
13	F	30	Nilópolis	Office worker	1.8	0.476
14	M	31	Rio de Janeiro	Electrician	1.9	0.366
15	M	32	Rio de Janeiro	Service worker (cleaning)	ND	ND
16	F	34	Rio de Janeiro	Laboratory technician	3.2	0.765
17	F	34	Rio de Janeiro	Service worker (cleaning)	ND	ND
18	M	37	Nova Iguaçu	Laboratory technician	ND	ND
19	M	39	Rio de Janeiro	Laboratory technician	2.8	0.740
20	M	39	Rio de Janeiro	Building painter	1.4	0.200
21	F	40	Rio de Janeiro	Service worker (cleaning)	ND	ND
22	M	40	Rio de Janeiro	Building painter	1.6	0.257
23	F	41	Rio de Janeiro	Service worker (cleaning)	1.9	0.401
24	M	41	Rio de Janeiro	Watchman	ND	ND
25	F	41	Rio de Janeiro	Service worker (cleaning)	2.4	0.354
26	F	41	Rio de Janeiro	Laboratory technician	6.0	3.452
27	M	42	Rio de Janeiro	Watchman	8.4	1.459
28	M	42	Duque de Caxias	Laboratory technician	ND	ND
29	M	48	Rio de Janeiro	Motor-vehicle driver	4.0	0.787
30	M	52	Rio de Janeiro	Electrician	1.8	0.419
31	F	55	Rio de Janeiro	Service worker (cleaning)	2.1	0.309
32	F	62	Rio de Janeiro	Service worker (cleaning)	3.5	0.828
33	M	63	Rio de Janeiro	Office worker	2.5	0.407

M = male; F = female; ND = not detected, below the limit for quantification (1,4µg/L). Recovery for *pp'*DDE was as high as 82.2%.

workers from Rio de Janeiro state (Paumgarten et al., 1998), were much lower than serum levels of *pp'*DDE and total DDT in samples of the general population from São Paulo, Goiás and Bahia states in the 1980s and early 1990s (Almeida, 1972; Carvalho, 1991; Fernícola & Azevedo, 1982; Lara et al., 1987; Minelli & Ribeiro, 1996).

The presence of *pp'*DDE in human tissues results from the dechlorination of *pp'*DDT (a major constituent of technical DDT) in the body and/or from a direct intake of *pp'*DDE residues in food from animal origin such as

meat, dairy products, eggs, fish and others (Ecobichon, 1995; Tordoir & Van Sitter, 1994; WHO, 1979). Since *pp'*DDE is more persistent in animal tissues than the parent compound itself, the percentage of *pp'*DDE in total DDT related material increases with the time elapsed after contamination by technical DDT, as well as with the level of the species in the food chain (Ecobichon, 1995; WHO, 1979). Thus, data from this survey, where *pp'*DDE was the only DDT derivative found in the blood serum, suggest that current low levels in the population of Rio de Janeiro are due to *pp'*DDE residues still

Table 2

Age-dependence of *pp'*DDE serum levels (lipids adjusted; $\mu\text{g/g}$) in people living and working in the urban area of Rio de Janeiro city. Blood serum samples were taken from 33 volunteers in January 1999.

Samples	Age (years)			
	≤ 29	30 to 39	≥ 40	17 to 60
Men				
Total (Nr)	3	6	7	16
Positive samples (%)	33.3	50.0	71.4	56.2
<i>pp'</i> DDE ($\mu\text{g/g}$ lipids) ^a	ND (ND-0.278)	0.100 (ND-0.740)	0.407 (ND-1.454)	0.229 (ND-1.454)
Women				
Total (Nr)	7	4	6	17
Positive samples (%)	0	75.0 ^b	83.3 ^b	47.0
<i>pp'</i> DDE ($\mu\text{g/g}$ lipids) ^a	ND (ND-ND)	0.403 (ND-0.765)	0.378 (ND-3.452)	ND (ND-3.452)
Men + Women				
Total (Nr)	10	10	13	33
Positive samples (%)	10	60 ^b	76.9 ^b	51.5
<i>pp'</i> DDE ($\mu\text{g/g}$ lipids) ^a	ND (ND-0.278)	0.265 ^c (ND-0.765)	0.401 ^c (ND-3.452)	0.200 (ND-3.452)

ND: Not detected.

^a Median (range).

Chi-square test: ^b significantly different ($p \leq 0.05$) from ≤ 29 years group.

Kruskal-Wallis test followed by the Mann-Whitney *U* test: ^c significantly different ($p \leq 0.05$) from ≤ 29 years group.

circulating in food products. A similar conclusion can also be drawn from data on the levels of DDT-related material in the breast milk from 40 mothers who had been living in the urban area of Rio de Janeiro for at least 5 years in 1992 (Paumgartten et al., 2000). The concentration of *pp'*DDE in the foregoing pooled sample was as high as $1.52\mu\text{g/g}$ of milk fat, a level which corresponds to 89.4 % of total DDT ($1.7\mu\text{g/g}$ milk fat) (Paumgartten et al., 2000).

Differences in the diet (*e.g.* less consumption of dairy products and fatty food) or ingestion of less contaminated food products could both be an explanation for the lower levels of *pp'*DDE in inhabitants in rural districts (Paty do Alferes) from Rio de Janeiro state (Paumgartten et al., 1998) in comparison with those of people of the same age group living in urban areas (present study data).

DDT was employed by the National Health Foundation (FUNASA) for malaria control in the Amazon region until mid 1990s (Oliveira Filho, 1997). Nevertheless, as far as the authors are aware, DDT and other OCPs have not been used for insect-borne diseases control in Rio de Janeiro state during the last two decades. Moreover, DDT was forbidden by the Brazilian

Ministry of Agriculture as early as 1985. Thus, the current low levels of *pp'*DDE in the general population from Rio de Janeiro state seems to confirm that technical DDT has not been sprayed in the region for a number of years.

Conclusions

Serum levels of *pp'*DDE, the only OCP residue found in the present study, were low, but somewhat higher than those previously determined in people living in rural districts of Rio de Janeiro state. It was additionally shown that serum concentrations of *pp'*DDE increased from the youngest to the oldest group, a finding that seems to confirm an age-trend previously noted in agricultural workers from the county of Paty do Alferes. PCB congeners were not detected (detection limit = $2.0\mu\text{g/L}$) in any of the blood serum samples analyzed in the present survey.

In conclusion, data from this study suggest that inhabitants of the urban area of greater Rio de Janeiro city have a relatively low body burden of persistent OCPs and PCBs.

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