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Assessment of the Levels of DDT and its Metabolites in Soil and Dust Samples from Chiapas, Mexico

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Abstract The aim of this study was to assess the levels of DDT and its metabolites in two environmental matrices (soil and dust) in five communities in Chiapas, Mexico. DDT and its metabolites were analyzed by gas chromatography/mass spectrometry. The soil levels of total DDT ranged from non detectable (<LOD) to 26.98 mg/kg, while in dust samples the levels ranged from non detectable (<LOD) to 2,119.8 mg/kg. Several studies have identified soil and dust as important pathways of toxicant exposure. Whether the people living in our study area are at risk is an issue that deserves further analysis.

Keywords DDE · DDT · Dust · Mexico · POPs · Soil

The extraordinary stability and persistence of Persistent Organic Pollutants (POPs), together with their lipophilic properties, leads to their accumulation in the food chain and causes considerable health and ecotoxicological effects (Colborn et al. 1993; Olsson et al. 1994; Ratcliffe 1967). Therefore, production prohibition or stronger restrictions in the application or emission of POPs have been necessary; this was the aim of the Stockholm Convention on POPs (UNEP 2004). This convention sought to determine baseline levels in environmental and biological samples; however, in developing countries, the levels of these chemicals in hot spots may be an issue of public health due to their magnitude. Furthermore, taking into account the scarcity of data in any matrix, there is an urgent need to assess the concentrations of POPs in environmental and biological samples.

The Stockholm Convention on POPs, which came into force on the 17th May, 2004, outlawed the use of 12 industrial chemicals, including 1,1,1-trichloro-2,2-bis(p-chlorophenyl)ethane (DDT; UNEP 2004). However, one exemption clause allows malaria-endemic nations to use DDT strictly for indoor residual wall spraying. The United Nations Environment Programme estimates that about 25 countries will use DDT under exemptions from the DDT pesticide ban (UNEP 2004). In Mexico, DDT was used until the year 2000, and DDT and its metabolites have been found in the environment (Yanez et al. 2002) and in human tissues (Perez-Maldonado et al. 2006; Yanez et al. 2002) in Mexico.

Therefore, the aim of this study was to assess the levels of DDT and its metabolites in two environmental matrices (soil and dust) in five communities in Chiapas, Mexico. All communities studied are malaria-endemic communities.

Materials and Methods

The communities were selected from those previously identified in the study zone as villages where DDT was used for malaria control (1957–2000). Inclusion criteria were: the age of the house (to ensure that DDT spraying was carried out in selected houses not more than 15 years

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old), and the location (which should be in rural areas). The geographical location and the names of each community are depicted in Fig. 1 and Table 1.

In order to confirm the presence of DDT and its metabolites, surface soil samples (1–5 cm in depth) were collected. Surface soil was collected outdoors in children's recreational areas located next to the dwellings. Indoor dust samples were collected from the corners and center of the main room (one compound sample was obtained from each dwelling). Outdoor surface soil samples and indoor dust samples were collected in all communities.

Soil and dust samples (1 g) were microwave-extracted in acetone–hexane (1:1) as described by Yanez et al. (2002). After extraction, samples were evaporated to 0.2 mL under nitrogen, and the extract was resuspended to 2.0 mL with hexane. Finally, cleanup was carried out on a Florisil column packed in a 6 mL solid-phase extraction cartridge, where the extraction was performed with 6% ethyl ether in hexane, and the Florisil eluate was concentrated to 1 mL under nitrogen. We performed an analytical determination of the target analyte using gas chromatography/mass spectrometry. PCB-141 or PCB-29 was used as an internal standard.

Quantification was performed using a HP 6890 gas chromatograph coupled with a HP 5973 mass spectrometer. A HP5-MS column, 60 m × 0.25 mm ID, 0.25-μm film thickness was used (J&W Scientific, Bellefonte, PA, USA). Column temperatures were: initial, 100°C (2 min), final,

Fig. 1 Location of communities studied

Table 1 Characteristics of the studied sites

Community	Characteristics
Nuevo Nicapa (A)	Rural and indigenous community with agriculture activity.
San Miguel 2da. Sección (B)	Semi-urban community with industrial activity.
Nuevo Francisco León (C)	Rural and indigenous community with agriculture activity.
San Martín Chamizal (D)	Rural and indigenous community with agriculture activity.
Frontera Corozal (F)	Rural and indigenous community with agriculture activity.

310°C (rates: 20°C/min up to 200°C, 10.0°C/min up to 245°C, 4.0°C/min up to 280°C and 30°C/min up to 310°C). Injector temperature was 270°C operated in pulsed splitless mode. Helium was used as the carrier gas at a linear velocity of 1.0 mL/min. Under these conditions and using data from nine replicates near the lowest concentration attainable on the calibration curve, the method detection limits for DDT and its metabolites were approximately 0.30 µg/L. For DDT and its metabolites in soil and dust, Analytical Reference Material EC-2 (Environmental Canada, National Water Research Institute) was used. Our extraction efficiency was 90–110% for all tested analytes.

Mean levels of all contaminants were compared between communities, using the Kruskal–Wallis test, followed by the Mann–Whitney U test to compare between communities. For all statistical analyses we used JMP IN Start Statistics Software 5.0 (SAS Institute).

Results and Discussion

Tables 2 and 3 show the levels of DDT and its metabolites in outdoor soil and indoor dust, respectively. The levels of total DDT in outdoor soil ranged from non detectable (<LOD) to 26.98 mg/kg, with the highest mean levels in community C (1.11 mg/kg). The levels of total DDT in indoor dust ranged from non detectable (<LOD) to 2,119.8 mg/kg, but in this case, the mean levels for indoor dust were higher in community E (4.08 mg/kg). Taking into account the guideline for total DDT in residential soil of 0.7 mg/kg in Canada (Environment Canada 2007), in our study different scenarios were observed in the studied communities. Regarding outdoor soil levels, community A (10%), C (77%), D (71%) and E (25%) had samples with levels above the Canadian guideline (Table 2), with the highest mean levels found in communities C and D. For indoor dust samples, similar results were obtained to those in outdoor samples, where community A (45%), C (100%), D (100%) and E (66%) had samples with levels above the Canadian guideline. In community C, DDT and its metabolites were found at higher concentrations than all the other communities (Tables 2, 3). With the exception of community B, all communities had samples above the Canadian guideline. It is possible that the DDT levels in San Miguel (community B) were lower because in this community DDT was used only for a short period in the malaria control program. Moreover, when the levels found in our study were compared with levels found in Mesoamerican countries (Perez-Maldonado et al. 2010), it was noted that the levels of total DDT in outdoor soil were higher in communities C, D and E than the levels assessed in outdoor soils in seven countries in Mesoamerica (Perez-Maldonado et al. 2010). For indoor dust samples, the levels in communities C, D and E were higher than in six countries in Mesoamerica (Honduras, Nicaragua, El Salvador, Guatemala, Panama and Mexico), only Costa Rica (14.0 mg/kg) had levels higher than the communities in our study.

Table 2 Levels of DDT and its metabolites (mg/kg) in outdoor surface soil

Community	n	Compound	[a]G.M ± SD	Min–max	% > 0.7
A	20	DDE	0.045 ± 2.160^{+}	0.004–9.240	10
		DDD	0.011 ± 0.181	<LOD–0.710	
		DDT	0.016 ± 2.080^{+}	<LOD–9.320	
		Total DDT	0.072 ± 4.33^{+}	0.008–19.280	
B	20	DDE	0.003 ± 0.002*	<LOD–0.007	0
		DDD	0.002 ± 0.0*	<LOD–0.002	
		DDT	0.005 ± 0.0*	<LOD–0.005	
		Total DDT	0.010 ± 0.002*	0.002–0.010	
C	22	DDE	0.366 ± 0.619	0.016–2.090	77
		DDD	0.088 ± 0.429$^{\&}$	0.005–1.870	
		DDT	0.493 ± 0.523	0.010–1.990	
		Total DDT	0.947 ± 1.140$^{\&}$	0.040–3.740	
D	14	DDE	0.225 ± 0.427	0.002–1.370	71
		DDD	0.022 ± 0.063	0.005–0.248	
		DDT	0.379 ± 0.465	0.012–1.760	
		Total DDT	0.626 ± 0.880	0.019–3.220	
E	16	DDE	0.153 ± 1.620	0.005–6.070	25
		DDD	0.031 ± 1.180	0.005–4.130	
		DDT	0.252 ± 4.740	0.012–16.70	
		Total DDT	0.436 ± 7.500	0.022–26.980	

[a] Geométric mean ± Standard deviation (SD)

* $p < 0.01$ when compared to A,C, D and E communities; $^{+}$ $p < 0.05$ when compared to C, D and E communities; $^{\&}$ $p < 0.05$ when compared to D and E communities. <LOD below detection limit (LOD = 0.0003 mg/kg)

Table 3 Levels of DDT and its metabolites (mg/kg) in indoor dust samples

Community	n	Compound	[a]G.M ± SD	Min–max	% > 0.7
A	20	DDE	0.112 ± 0.406^{+}	0.004–1.080	45
		DDD	0.059 ± 0.245$^{\varepsilon}$	0.003–0.680	
		DDT	0.129 ± 1.120^{+}	<LOD–3.470	
		Total DDT	0.367 ± 1.630^{+}	0.009–4.850	
B	20	DDE	0.003 ± 0.006*	<LOD–0.015	0
		DDD	<LOD	<LOD	
		DDT	0.014 ± 0.0*	<LOD–0.014	
		Total DDT	0.018 ± 0.011*	0.002–0.030	
C	22	DDE	3.400 ± 18.10$^{\&}$	0.240–60.40	100
		DDD	1.510 ± 8.110$^{\&}$	0.009–31.30	
		DDT	1.920 ± 9.030	0.010–36.90	
		Total DDT	6.830 ± 26.90$^{\&}$	0.663–95.90	
D	14	DDE	1.360 ± 3.350	0.381–10.60	100
		DDD	0.043 ± 0.104$^{\varepsilon}$	0.016–0.286	
		DDT	1.770 ± 1.700	0.527–6.210	
		Total DDT	3.170 ± 4.110	1.30–12.20	
E	16	DDE	0.791 ± 22.60	0.100–78.40	66
		DDD	0.414–140.90	0.016–489.90	
		DDT	1.80 ± 444.90	0.010–1551.40	
		Total DDT	3.00 ± 608.00	0.409–2119.00	

[a] Geométric mean ± Standard deviation (SD)

* $p < 0.01$ when compared to A, C, D and E communities; $^{+}$ $p < 0.05$ when compared to C, D and E communities; $^{\varepsilon}$ $p < 0.5$ when compared to C and E communities; $^{\&}$ $p < 0.05$ when compared to D and E communities. <LOD below detection limit (LOD = 0.0003 mg/kg)

Table 4 DDT/DDE quotient in outdoor surface soil and indoor dust samples

Community	n	DDT/DDE soil	DDT/DDE dust
A	20	0.3	1.1
B	20	–	2.0
C	22	1.3	0.5
D	14	1.6	1.3
E	16	1.6	2.2

When analyzing the results for DDT and DDE, the quotient for DDT/DDE can be obtained, and the results for outdoor soil and indoor dust samples are shown in Table 4. For soil samples in community C (1.3), D (1.6), and E (1.6), the DDT/DDE ratio was above the unit. For dust samples, only community C (0.5) had a DDT/DDE ratio below the unit. We also performed correlation analyses between outdoor soil and indoor dust levels of DDT, DDE and the DDT/DDE quotient, but no associations were found (data not show). It is important to mention that the ratio of DDT/DDE can be used as a rough indication of the period of its application, a ratio of DDT/DDE near to 1.0 or higher than 1.0 indicates use in the last 5 years (Tavares et al. 1999). In general, a ratio higher than 1.0 was found in all the communities, suggesting recent use of the insecticide. There are different explanations for this finding; but it appears that recent use of this insecticide is the most reasonable explanation, as also suggested by Alegría et al. (2006). In this regard, it is important to note that in different countries, the product used for the control of malaria is still available in some warehouses (for general pest control). Therefore, it is probable that the communities studied have deposits of DDT (used for malaria control several years ago), and it is possible that the people living in those communities are still using this store of DDT. Special attention should be paid to the results in community C, where the quotient was above the unit in outdoor soil samples, but was below the unit in indoor dust samples. It is possible that DDT was recently used in outdoor areas, however, DDT is not applied indoors.

Several studies have identified indoor house dust as an important pathway of toxicant exposure. Often levels of pollutants found in house dust, including compounds banned long ago such as DDT, are significant sources of exposure for the general population, especially children (Butte and Heinzow 2002; Hwang et al. 2008; Rudel et al. 2003). Moreover, analyses of compounds in house dust are a measure of indoor contamination, but may also provide valuable information on the assessment of human indoor exposure (Butte and Heinzow 2002). Also, outdoor soil is considered an important exposure pathway for the general population and children to compounds banned long ago (Herrera-Portugal et al. 2005). However, it is important to note that longer residence times and elevated contaminant concentrations in the indoor environment may increase the chance of exposure to these contaminants by 1,000-fold compared to outdoor exposure (Hwang et al. 2008).

It should be noted that additional pathways of exposure and uptake require to be considered. In this respect, we must point out the limitations of our assessment, as we did not study pathways such as breast milk; other food items; and other areas (soil/dust samples around warehouses or in areas where DDT was utilized for agriculture; etc.). Nevertheless, in order to analyze the possibility of other sources of DDT, a comprehensive study of the environmental fate and distribution of the insecticide in the studied area must be conducted. Whether the people living in our study area are at risk is an issue that deserves further analysis; however, applying the precautionary principle, it is important to begin a risk reduction program to decrease exposure of the people in the studied area to DDT and its metabolites.

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