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DDTs in Soils Affected by Mosquito Fumigation in Belize

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DDTs were sprayed extensively in Belize to combat malaria but widespread use ceased after 1997. To determine if DDTs still persist in Belize's soils, 23 composite soil samples were collected from each of the two towns of Dangriga and Punta Gorda. Dichlorodiphenyltricholoroethane (DDT) and its breakdown products dichlorodiphenyldichloroethylene (DDE) and dichlorodiphenyldichloroethane (DDD) were the only organochlorine compounds detected in the soils. Results show that DDTs are transferred to the soil environment as a result of spraying houses and that the pesticides still persist in soils in southern Belize after more than 10 years of non-use. Sprayed lots had DDT concentrations up to 240 μg/kg in Dangriga and up to 410 μg/kg in Punta Gorda. All unsprayed lots had below detection limit concentrations, with the exception of two sites in Punta Gorda. Hot spot analysis in GIS indicates that significant spatial variability exists in detected concentrations of DDTs, which has implications for extrapolation of local data. Belize currently has no guidelines for determining risk of DDTs to human health and soil cleanup, but soil cleanup guidelines employed by foreign governmental entities such as the Netherlands and California show that all detected pesticide concentrations in this study are below mandated concentrations of concern for the residential areas of these regions. However, since exposure scenarios may be different in Belize, it is recommended that the Belize Ministry of Health conducts a risk analysis to ascertain if the concentrations of DDTs in sprayed areas pose a risk to the inhabitants of the two towns.

Keywords DDT, soil quality, Belize, GIS

Introduction

Background

Dichlorodiphenyltrichloroethane (DDT) has been considered one of the most effective means worldwide to combat pests in agricultural crops and in the fight against dreaded insect-borne diseases such as malaria and dengue hemorrhagic fever, especially in the tropics (Kannan et al., 1995). The heavy use of DDTs globally has led to widespread environmental contamination and accumulation in humans and wildlife (Saxena and Siddiqui, 1980). DDT is very persistent in the environment (Nash and Woolson, 1967; Heberer and Dunnbier, 1999) and is almost insoluble in water (ATSDR, 2002). Residues have been detected in places as remote as the Arctic (Heberer and Dunnbier, 1999). As much as 50%

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can remain in the soil up to 20 years after application (Andrade et al., 2005; ATSDR, 2002). DDT's persistence and high solubility in fats causes it to readily bio-accumulate and bio-concentrate in organisms (Falandysz et al., 2001; USEPA, 2007). The World Health Organization (WHO) classifies DDT as "moderately hazardous," based on the rat oral LD50 (dose required to kill half the members of a tested population). The U.S. Department of Health and Human Services, the International Agency for Research on Cancer, and the USEPA have all determined that DDT may cause cancer in humans (ATSDR, 2002). Adverse impacts of DDTs on animals have also been documented for a long time, including effects in fish, birds, land and marine mammals (George and Stickel, 1949; Adams et al., 1949; Hrdina et al., 1975; Ware, 1975; Tanabe et al., 1994).

In Belize, DDTs have been severely restricted for use as insecticides only in case of public health emergencies. However, DDTs were used extensively in Belize between 1957 and 1997 but the overall quantity used in the country remains unknown (Belize National Malaria Eradication Service, 2003). It is estimated that worldwide approximately 1,000 tons of DDT mixture is used on an annual basis in countries where malaria poses a serious health problem (Callaway, 2008). It is also believed to be utilized illegally in many nations, particularly in the tropics (Racke et al., 1997) and because restrictions are often evaded in these countries the resistance of the disease-carrier mosquito to the pesticide may be growing (Chapin and Wasserstrom, 1981).

Pesticides such as DDTs that enter the soil environment are subject to a variety of degradation and transport processes (Cheng, 1990). The overall dissipation of a pesticide from soil results from a combination of mechanisms such as microbial degradation, chemical hydrolysis, photolysis, volatility, leaching, and surface runoff. The degree to which each mechanism will contribute to the overall loss of the pesticide is in turn dependent on the physicochemical properties of the pesticide (e.g., water solubility, sorptive affinity) (Altschuh et al., 1999; Morrison and Newel, 1999), characteristics of the soil (e.g., pH, texture, organic matter content, biomass, redox status) (Chassin and Calvet, 1984; Spark and Swift, 2002; Walker et al., 1999), environmental conditions (e.g., temperature, moisture, biological activity) (Baczynski et al., 2010), and management practices (e.g., application rate, formulation type) (WHO, 2006). Complex interactions and interdependencies that are difficult to quantify in-situ exist between these variables (Donzel and Dorn, 1992). While there is no real difference in the type of soil-forming processes operating in the tropics and temperate areas, there are significant differences in degree (Sotomayor-Rios and Pitman, 2000). In most of the tropics, the combination of continually high temperatures and large amounts of rainfall produce extreme leaching conditions. The types and properties of clay minerals are much more varied in the tropics than in temperate areas and many tropical soils exhibit significant anion exchange capacities (Sanchez, 1976). While organic matter contents in the tropics can be similar to those of temperate regions, the rate of organic decomposition can be five times greater in the tropics (Sanchez, 1976).

Use of DDTs in Belize

The total amount of DDTs used historically in Belize is not known accurately; however, between 1985 and 1994 an average of about 25 tons was used on an annual basis (Figure 1) (Belize National Malaria Eradication Service, 2003). While other persistent organic pollutants (POPs) were used as agricultural chemicals, DDTs were restricted in Belize for use only to combat malarial outbreaks. Approximately 794 grams (28 ounces) of a 75% wettable powder formulation was applied twice annually to the interior and exterior walls and ceiling of each house, and also to adjacent structures such as outdoor kitchens and

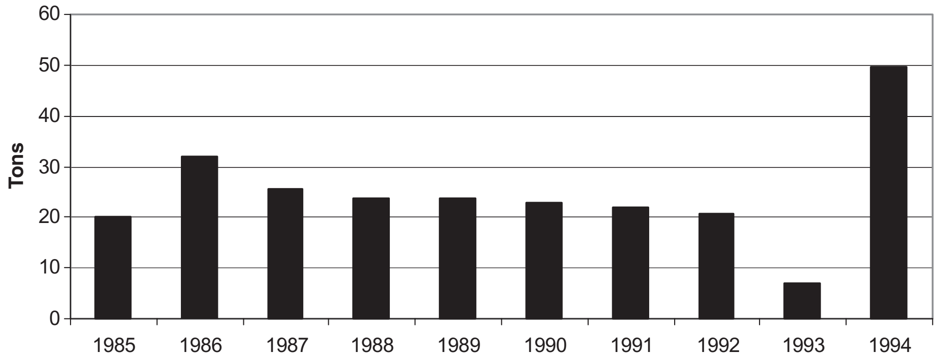

Figure 1. Annual quantity of DDT used in Belize for the period 1985–1994.

outhouses. Consequently, a higher level of soil DDTs can be expected in older sections of Belizean towns that existed before the 1997 restriction than in newer sections. In the newer populated areas, DDTs were never officially applied so any DDTs found would be due to natural transport processes or undocumented anthropogenic causes (Wilderness Group Consulting, 2006).

There are growing concerns about the extent of exposure of Belizeans to DDTs that persist in the Belizean soil environment. Although DDTs have not been officially used in Belize since 1997, a study on airborne concentrations of POPs revealed that some of the highest levels were for DDTs (Alegria et al., 2000). The study suggests that DDTs may have been recently used in the country. Another study conducted in Belize (Fernandez, 2001) showed DDT residue in one soil sample at 260 μg/kg, and in sediments ranging from 5380 μg/kg for samples taken at the mouth of the Belize River to 3350 μg/kg for samples taken from a location on the upper Macal River (Figure 2). These findings indicate that higher DDT concentrations may be present in coastal areas of the country.

The effect that fumigation with DDT mixture has on DDTs in soil has not been assessed in Belize but the limited data available from elsewhere indicate that the potential for contamination with DDTs exists. Samples collected from the bare dirt floor of a house in Oaxaca, Mexico, that was fumigated with DDT mixture had concentrations of 83,000 μg/kg of DDT, 41,000 μg/kg of DDD, and 14,000 μg/kg of DDE two years after final spraying (WHO, 2006). Outside the same house, soil samples had 49,000 μg/kg of DDT, 13,000 μg/kg of DDD, and 5,700 μg/kg of DDE. In Brazil, soil samples from close to fumigated outside walls of a house had up to 209 μg/kg DDT and up to 135 μg/kg DDE seven years after spraying (Vieira et al., 2001).

Objectives

The limited data that exist for Belize on levels of DDTs in environmental samples do not allow for a meaningful assessment of human health and environmental impacts. Moreover, no substantive studies have been carried out to investigate the levels of DDTs in the soil environment of southern Belize, where thousands of local people rely heavily upon the soil environment for their livelihood. Most of what is currently known about levels of DDTs in soils comes from research and studies conducted in temperate areas, and few studies have actually been carried out to investigate the levels of DDTs in tropical soils (Racke et al., 1997).

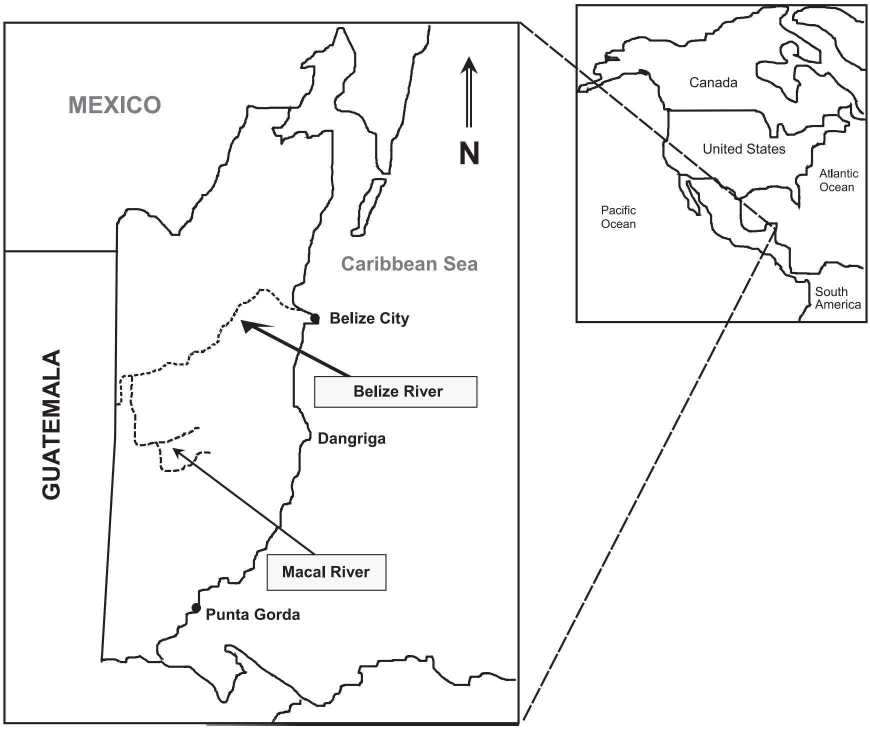

Figure 2. Map of Belize showing location of Dangriga and Punta Gorda towns.

The objective of this study is to contribute to the knowledge about the persistence of DDTs in urban soils in the tropics at sites that were fumigated with DDT mixture. We aim to achieve this objective by determining the existing levels of DDTs in two urban areas in southern Belize (Dangriga and Punta Gorda towns) (Figure 2) that have sprayed and unsprayed sections. Specific objectives include: 1) to assess differences in concentrations of DDTs in soils near sprayed and unsprayed dwellings; 2) to compare the observed levels of DDTs to soil pollution guidelines for human and ecosystem health; and 3) to assess spatial variability in concentrations of DDTs in the two towns.

This study examines multiple sites in two cities and compares areas that were sprayed and unsprayed to assess the impact of spraying, an approach that to our knowledge has not been used before. It is useful to authorities in Belize and elsewhere that are concerned with the potential adverse impacts of fumigation with DDTs for disease vector control. This is especially important for other tropical areas where data like ours are usually not available. Results also are of broader scientific relevance to those working on pesticide sources and pathways in tropical soils.

Materials and Methods

Study Area

Dangriga and Punta Gorda towns are two of the largest coastal communities in Belize and were among the more heavily DDT-sprayed areas of the country during the Ministry of Health (MOH) Anti-Malaria Campaign from 1985 to 1994. DDTs are also believed to have been used in these towns up to 1997 (Wilderness Group Consulting, 2006). Dwellings constructed after 1997 remained unsprayed and soils near these buildings are expected to be uncontaminated with DDTs.

With a population of approximately 12,000, Dangriga is the largest town in southern Belize (Statistical Institute of Belize, 2010). It was settled before 1832, and is situated at about mean sea level. The town of Punta Gorda has a population of approximately 6,000 and is the southernmost "sizeable" community in Belize (Statistical Institute of Belize, 2010). It was a small fishing camp before being settled in 1823. The town is situated about 15 feet above sea level. The entire coastal area of Belize and all the islands are flat and low-lying. Parts of the coastline have an elevation of less than 1m to a distance of several miles inland.

The climate of coastal south Belize is sub-tropical, with temperatures ranging from 21°C from October to February to 32°C during May to September (Belize National Meteorological Service, 2010). The annual mean relative humidity is 82%; the total annual rainfall is about 406 cm. There are two distinct seasons: the rainy season, which normally begins in late May and lasts until November, and the dry season, which begins in December and ends in early May.

Selecting Sprayed and Unsprayed Areas

Because spraying of DDTs officially ended in Belize in 1997 (Wilderness Group Consulting, 2006) it was assumed that all houses that were in existence up to this date were sprayed, and all houses that were built after 1997 were not sprayed. No aerial photos or detailed street maps of Dangriga and Punta Gorda are available for 1997 to verify this. Since the towns grew primarily block by block, areas that were and were not sprayed were easily identified on Google Earth aerial photos based on personal communication with the towns' citizens and Belizean health department personnel. The city blocks in each town were numbered on Google Earth aerial photos and a random number generator was used to select 10 blocks each from sprayed and unsprayed areas from both towns. Houses that fell within selected blocks were also numbered and chosen using the same random selection process.

The estimated age-class of the houses (pre- or post-1997) was used as the primary determinant when selecting sample sites that were and were not sprayed. The true age for each house of interest was verified with the local resident prior to sampling. Pre-selected houses from within selected blocks were not sampled if their true age did not fall within the estimated age-class; another house from within the same block would then be randomly chosen in the field.

Soil Sampling

Twenty-three soil samples were collected from each of the two towns: 11 each from the sprayed areas (including one duplicate), 10 each from the unsprayed areas, and two background samples from within apparently undisturbed wooded areas away from any development. DDTs are mostly insoluble in water and are almost immobile in most soils, and tend to accumulate in the top layer of soils. However, residues at the soil's surface have a greater chance of being broken down by sunlight or dissipated than those just below the surface (USEPA, 2007). Therefore, samples were collected from 5–15 cm depth to ensure capturing any DDTs present.

Samples were taken from near the four sides of each house to eliminate factors such as sun/shade and unequal spraying, and completely mixed in a pre-cleaned container to form a composite sample. All samples were collected by hand using a stainless steel scoop and placed in cushioned pre-cleaned glass sampling jars with teflon-lined lids after mixing. Samples were kept preserved on a combination of ice and gel packs and transported in a sealed cooler to the analytical laboratories.

Soil samples were extracted ultrasonically according to USEPA method 3550B (USEPA, 2008a) by mixing the sample with anhydrous sodium sulfate and a hexane-acetone solvent, prior to vacuum filtration. Extracts were analyzed on a gas chromatograph equipped with an electron capture detector and a wide-bore analytical capillary column. Helium was used as the carrier gas and argon/methane as the makeup gas (EPA method 8081A) (USEPA, 2008b). Decachlorobiphenyl and tetrachloro-m-xylene were used as surrogates to monitor the performance of the method. Average recoveries were 88.9% and 96.8% for decachlorobiphenyl and tetrachloro-m-xylene, respectively. All recoveries were between the formal quality control acceptance limits of 65–123% for decachlorobiphenyl and 63–125% for tetrachloro-m-xylene. For the statistical analysis, half the values of the analytical lab's reported minimum detection limit (MDL) were used for those concentrations that were reported as below detection limit (BDL). A summary of the detection levels is provided in Table 1.

Global Positioning System (GPS) coordinates were obtained for all sample locations using a handheld GPS receiver with a positional accuracy of three meters as per the manufacturer. Other site attributes (i.e. age of house, house type, ground cover type, amount of ground cover, and yard condition) were also recorded for each sample site.

Soil Attributes Analysis

Some studies suggest that the residual and persistent nature of DDTs is affected by soil pH, organic matter content, moisture content, and particle size (Gong et al., 2004; Schnitzer and Khan, 1978). These factors were therefore determined to assess their possible effects on the observed concentrations of the DDTs. Soil organic matter was analyzed with the

Table 1
Summary of minimum detection limits (MDL) [μg/kg]

	DDT	DDE	DDD
		Punta Gorda, sprayed	
Minimum MDL	0.052	0.081	0.072
Maximum MDL	0.86	1.3	1.2
		Punta Gorda, unsprayed	
Minimum MDL	0.045	0.059	0.052
Maximum MDL	0.19	0.1	0.092
		Dangriga, sprayed	
Minimum MDL	0.037	0.059	0.052
Maximum MDL	0.39	0.31	0.27
		Dangriga, unsprayed	
Minimum MDL	0.039	0.061	0.053
Maximum MDL	0.19	0.3	0.27

dry combustion method (measured sample heated in a muffle furnace to 600°C for six hours). Soil pH was determined using a bench top pH meter in a 1:1 deionized water-soil slurry mixture (Jackson, 1964) with an equilibrium period of 60 minutes, as specified in EPA Method 9045D (USEPA, 2008c). Soil moisture content was determined by overnight drying at 104°C. Particle size distribution was determined using the pipette method (USDA Method 3A1) (Burt, 2004).

Data Analysis

A multifactor Analysis of Variance (ANOVA) was conducted to examine the significance of the difference in concentrations of DDTs for the sprayed and unsprayed areas within and between the two towns. A Pearson correlation analysis was conducted to determine the relationships between concentrations of DDTs and the soil characteristics. Data for unsprayed areas were omitted from the correlation analysis because of the predominance of BDL results. Because spraying of DDTs officially ended in 1997, the building of new and different types of homes during recent expansion of the towns may have affected the concentrations of DDTs in the soils. A Pearson Chi Square test was thus performed to determine if house type (i.e. construction material) was correlated with the concentrations of DDTs in the soils.

A Gettis-Ord GI* Hot Spot Analysis was carried out using the Spatial Analyst tools for ArcGIS 9.3 (Environmental Systems Research Institute, 2009) to evaluate the spatial pattern of the DDT concentrations and to identify possible hot spot areas where higher concentrations might cluster together. Spatial relationships were conceptualized using the inverse distance band and the Euclidean distance method. The Gettis-Ord GI* Hot Spot Analysis calculates the Gettis-Ord GI* statistic for hot spot analysis and the output Z-score shows areas where higher (or lower) than average DDT concentrations tend to be found near each other. A similar hot spot analysis was used by the USEPA to evaluate arsenic concentrations in soils in residential areas in Minneapolis, MN, that were believed to have been affected by a facility that produced arsenic-containing pesticides (USEPA, 2007).

Results and Discussion

Concentrations of DDT, DDE, and DDD

The geomean DDT concentrations for the sprayed areas in Dangriga and Punta Gorda are 5.10 μg/kg and 22.79 μg/kg, respectively (Table 2). These concentrations are comparable to those in soils in larger cities such as Beijing and Taiyuan, China, where no direct fumigation took place (Yang et al., 2010; Fu et al., 2009). Of the sprayed samples from Dangriga, three (30%) had no detectable levels of DDTs. The lack of detection of any of the DDTs at these sites suggests that these houses were skipped during the spraying campaign. None of the unsprayed samples from Dangriga were positive for any of the compounds. In Punta Gorda, 90% of the sprayed samples had detectable concentrations of DDT and DDE, and 70% had detectable concentrations of DDD (Table 2). At all sites in the sprayed area at least one of the three DDTs was detected. Two of the unsprayed samples were positive for all three compounds. One of these sites had low detected concentrations (4.1 μg/kg total DDT) but concentrations at the second site were relatively high (100 μg/kg DDT, 36 μg/kg DDE, and 5.1 μg/kg DDD, 141 μg/kg total DDT). This latter site is located just across the street from the sprayed zone in Punta Gorda, unlike any other unsprayed site that was sampled, and may have experienced drift of DDTs that has been observed elsewhere during

Table 2

Concentrations of DDTs [μg/kg], soil characteristics [%], and site characteristics for sprayed and unsprayed areas in Dangriga and Punta Gorda

	DDT	DDE	DDD	Total DDT	OM[3]	Sand	Clay	Silt	pH	Age (years)	House Type[4]
Punta Gorda, sprayed											
Minimum[1]	7.1	6.7	8.7	7.420	2.16	70.14	1.73	7.87	6.6	21	
Maximum	410	300	67.0	741	35.04	89.57	12.06	20.55	7.31	34	
Geomean[2]	22.79	16.54	6.39	68.13	7.91	80.66	4.13	13.14	6.94	26.24	8c, 2w
Stand. dev.	125.12	90.09	21.72	223.55	9.49	7.15	4.20	3.65	0.25	4.70	
% BDL	10	10	30	0[5]							
Punta Gorda, unsprayed											
Minimum[1]	1.1	1.3	1.7	4.1	6.27	29.18	12.37	17.57	6.28	1	
Maximum	100.2	36.0	5.1	141.1	23.96	70.06	42.76	46.95	7.94	10	
Geomean[2]	0.09	0.17	0.20	0.78	9.53	37.60	26.15	32.24	6.85	2.19	10c
Stand. dev.	31.58	11.28	1.73	44.23	5.14	12.28	9.68	8.31	0.51	3.03	
% BDL	80	70	60	50							
Dangriga, sprayed											
Minimum[1]	13.75	1.2	3	25.3	0.35	86.93	0.67	2.19	6.21	15	
Maximum	240	34.5	23.0	282	4.79	96.97	3.1	10.57	8.3	37	
Geomean[2]	5.10	2.10	0.95	9.92	1.31	92.02	1.42	5.79	7.04	22.31	4c, 6w
Stand. dev.	72.73	12.30	8.03	87.16	1.29	3.21	0.83	2.50	0.66	6.54	
% BDL	30	30	40	30							
Dangriga, unsprayed											
Minimum[1]	BDL[6]	BDL	BDL	BDL	1.07	44.7	1.17	2.28	5.14	1	
Maximum	BDL	BDL	BDL	BDL	6.86	96.54	17.05	38.89	7.52	10	
Geomean[2]	na[7]	na	na	na	2.72	75.01	4.71	12.34	6.53	4.39	8c, 2w
Stand. dev.	na	na	na	na	1.91	17.91	5.40	12.61	0.77	3.17	
% BDL	100	100	100	100							

1. Minimum values are the lowest detected value.
2. For the geomean and standard deviation half the minimum detection limit (MDL) was used for those concentrations that were reported as below detection limit (BDL).
3. OM = organic matter.
4. C = concrete, W = wood.
5.% BDL for total DDT refers to percentage of sites without detection of any of the three compounds.
6. Below detection limit.
7. Not available, because both the minimum and maximum were BDL.

application (UNEP, 2003; Burkhard and Guth, 1981). However, it is impossible to establish with certainty the reason(s) for the high concentrations at this site. In any case, the detection of at least one of the DDTs at half of the sites in an area of Punta Gorda where no mosquito fumigation has taken place (Table 2) indicates that some type of transport mechanism is bringing DDTs to this unsprayed area. With one exception the concentrations in this area are very low and do not seem to pose any threat to human or ecosystem health. The highest observed DDT concentrations are 410 μg/kg and 240 μg/kg for Punta Gorda and Dangriga, respectively, and are from the sprayed areas (Table 2). ANOVA revealed no significant differences between the DDT concentrations in Dangriga and Punta Gorda ($p =$ 0.510) in general. The ANOVA showed that significant differences in concentrations of DDTs exist between the sprayed and unsprayed areas for the two towns combined (ANOVA p-values are 0.023, 0.057, 0.002, and 0.017 for DDT, DDE, DDD, total DDT respectively), indicating that spraying is the likely source of DDTs in these two towns.

The range of DDT concentrations for the two towns is from BDL to 410 μg/kg, and total DDT (DDT + DDE + DDD) ranges from BDL to 741 μg/kg. The concentrations reported here for the Dangriga and Punta Gorda sites are much lower than those found in soils outside of a house in Oaxaca, Mexico (49,000 μg/kg of DDT, 13,000 μg/kg of DDD and 5,700 μg/kg of DDE), where the house interior was sprayed with DDT mixture two years prior (WHO, 2006). The concentrations in the soils for this house in Mexico were obtained more recently after spraying (two years) than for the Belize sites (>15 years), which at least in part explains the much higher values. In Brazil, maximum DDT and DDE concentrations seven years after application (209 μg/kg and 135 μg/kg, respectively) (Vieira et al., 2001) were more comparable to ours. Concentrations of DDTs comparable to those observed in the present study have been found in surface soils impacted directly or indirectly by spraying of crops with DDT mixtures, such as, for instance, in Chile after widespread spraying ended about 20 years after initial use (Instituto de Investigaciones Agropecuarias, 1990). In surface soils in Tianjin, China, average concentrations for DDT and DDE (27.5 and 18.8 μg/kg, respectively) that are comparable to those from the present study have been found (Gong et al., 2004). In that study, the spatial distribution of the DDT and DDE in the soils generally matched pesticide application patterns, similar to the link we find between presence or absence of fumigation and concentrations of DDTs.

Concentrations of DDT are higher than those of its breakdown products DDE and DDD (Table 2). This seems consistent with the relatively low levels of contamination, even in the sprayed areas, as it has been argued that DDE and DDD exceed DDT concentrations mostly at highly contaminated sites (Brown, 1997). The ratio of the concentration of DDT to that of DDE and DDD combined has been used to evaluate the age of last major pesticide input (Xu et al., 2005; Yang et al., 2010) and Alegria et al. (2000) used the DDT:DDE ratio to describe the "freshness" of atmospheric sources of DDTs. A ratio >1 has been interpreted as evidence for "recent" pesticide use while ratios <1 were thought to represent "older" and more degraded input (Jaga et al., 2003). In Dangriga 7 of the 10 sprayed sites had a ratio >1, suggesting "recent" use although recent can not be quantified with this approach and thus does not necessarily point to illegal recent use. In Punta Gorda 4 of the 10 sprayed sites had a DDT/DDE+DDD ratio >1 but with one exception the ratio was <2. This indicates that recent and potentially illegal use in Punta Gorda has been very limited. One Punta Gorda sample, however, had a ratio of 22, which strongly suggests that DDTs may have been used recently and illegally at that location. Recent unauthorized use of DDTs was also suspected in parts of Tianjin, China, even though a ban on usage of DDTs had been in place for 20 years prior (Gong et al., 2004). A high correlation of DDT with its metabolites DDE and DDD (Table 3) shows that the pesticide's dissipation pathways might be similar

Table 3
Pearson correlation coefficients for DDT compounds and soil characteristics and dwelling age

	DDT	DDE	DDD	Total DDT
DDT	1.000			
DDE	0.866[3]	1.000		
DDD	0.499	0.428	1.000	
Total DDT	0.977[3]	0.943[3]	0.564	1.000
OM[1]	−0.204	−0.097	−0.006	−0.159
Sand	0.228	0.096	−0.105	0.160
Clay	−0.204	−0.106	−0.121	−0.174
Silt	−0.221	−0.078	0.261	−0.133
pH	−0.03	0.078	−0.156	−0.004
Age[2]	0.603	0.442	0.529	0.582

1. Organic matter content.
2. Age of the dwelling at sampled location.
3. Significant at $p < 0.05$.

to those of its breakdown products in the soil environment of the two towns (Racke et al., 1997).

Influence of Soil and Site Attributes

Soil characteristics. A Pearson correlation analysis revealed weak and non-significant correlations between concentrations of the DDTs and soil characteristics (Table 3). Other studies have found stronger correlations between DDTs and organic matter content (Chassin and Calvet, 1984), soil reaction (Barrick and Prahl, 1987; Gong et al., 2004), and particle size (Wenzel et al., 2002). Some studies have failed to find a significant correlation between DDTs and particle size as we do (Yang et al., 2010). Providing an explanation for the weak correlations is impossible based on our data due to uncertainties in the actual application rate of the DDTs, differences in time of last application between sites, potential soil movement since the last application, and given the complex interactions of the various factors that affect the retention of DDTs in soils (Donzel and Dorn, 1992). Some have interpreted a lack of correlation between soil characteristics and DDTs as indicative of a dominance of differences in application rate over the effects of soil characteristics (Gong et al., 2004), but it is hard to firmly establish if that applies to our study area given the uncertainties.

Clay content is lower in the sprayed areas of the two towns than in the respective unsprayed areas (Table 2). This difference in clay content is due to the sandy fill material used to elevate the areas near the coastline where the early portions of the towns are located. This lower clay content in the sprayed areas indicates that even if the theoretical positive relationship between clay content and concentrations of DDTs would exist in the two towns, it could not explain the higher concentrations of DDTs in the sprayed older areas. This lends credence to the above contention that the higher concentrations of DDTs in the sprayed areas are the result of the fumigation process.

Site characteristics. Because the majority of the spraying of DDTs ended after 1997, the building of new homes with different construction materials during the expansion of the

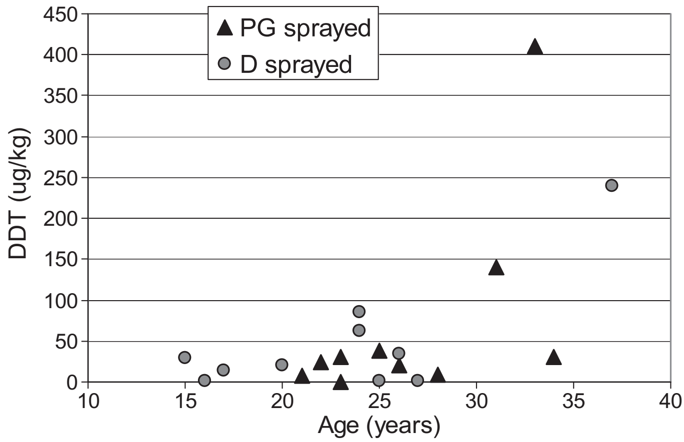

Figure 3. Scatter plot of DDT versus age of house for the sprayed areas of Dangriga (D) and Punta Gorda (PG).

towns may in principle have affected the concentrations of the DDTs in the soils. However, a Pearson Chi-Square test reveals no differences between the concentrations of DDTs near dwellings constructed with different materials (wood or concrete) (p-values are 0.461, 0.333, 0.395, and 0.395 for DDT, DDE, DDD, and total DDT, respectively). This indicates that the building material is unimportant in determining concentrations of DDTs in the soils. Correlation analysis for concentrations of DDTs and house age shows moderate-to-strong correlations ($r = 0.603$ for DDT) (Table 3, Figure 3). This indicates that older houses have DDT present in their soils in greater proportions than younger houses, because older houses received more applications than younger ones, and corroborates the observation that fumigation is the main factor explaining differences in soil DDT content.

Soil Quality Guidelines

Because no environmental standard for DDTs in soils currently exists in Belize, we compared the concentrations from Belize with intervention values for total DDT from the Netherlands (Ministry of Housing, 2000) and with soil Health Screening Levels (CHHSLs) for DDT, DDE, and DDD separately from the California Environmental Protection Agency (EPA) (California EPA, 2005) (Table 4). The Dutch soil remediation intervention values indicate when a soil is seriously impaired or threatened for humans, plant and animal life. They represent a level of contamination above which there is significant soil contamination and maximum permissible risk for humans may be exceeded. California EPA CHHSLs are direct-exposure screening levels that address human exposure to chemicals in soil via incidental ingestion, dermal absorption, and inhalation of vapors and particulates emitted to outdoor air.

No samples from Punta Gorda and Dangriga exceeded the intervention value from the Netherlands for total DDTs nor the California EPA CHHSLs for DDT, DDE or DDD (Table 4). Consequently, concentrations of DDTs in these towns do not seem to pose a threat to ecosystem or human health. Since official widespread spraying of DDTs was discontinued in Belize after 1997, degradation mechanisms such as hydrolysis and oxidation/reduction (Yoshida, 1975), photo-degradation (Samuel and Pillai, 1989), microbial degradation (Racke and Coats, 1990), volatility (Burkhard and Guth, 1981), and leaching

Table 4
Comparison of maximum concentrations of DDTs [μg/kg] in soils from Dangriga and Punta Gorda with soil quality guidelines

	Belize[1]			California EPA CHHSLs	
Substance	D	PG	Netherlands Intervention value	Residential	Commercial/Industrial
DDT	240	410	na[2]	1600	6300
DDE	37	300	na	1600	6300
DDD	23	67	na	2300	9000
Total DDT	282	741	4000	na	na

1. Maximum concentrations.
2. Not available.

(Gustafson, 1989) may be diminishing the overall concentrations of the DDTs in the soils in these two towns. Thus, future threats from soil DDTs to human health must be minimal in Dangriga and Punta Gorda in the absence of renewed use of DDTs.

DDT Hot Spots in Dangriga and Punta Gorda

Gettis-Ord GI* hotspot analysis was carried out for DDT, its two breakdown products, and total DDT. The hotspot analysis revealed the presence of at least one zone in each town where higher DDT concentrations clustered. For Dangriga, DDT and total DDT (Figure 4) concentrations both revealed one hot spot (DDT: highest $z = 3.17$, $p = 0.0006$; total DDT:

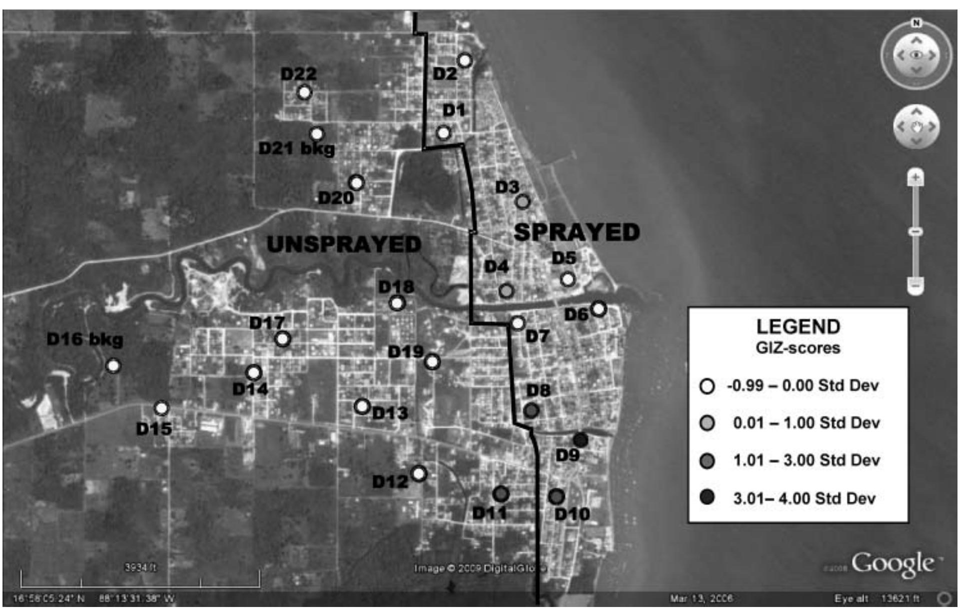

Figure 4. Hot spot in Dangriga using total DDT concentrations. Background image source: Google Earth © 2008 Google, image © 2009 DigitalGlobe.

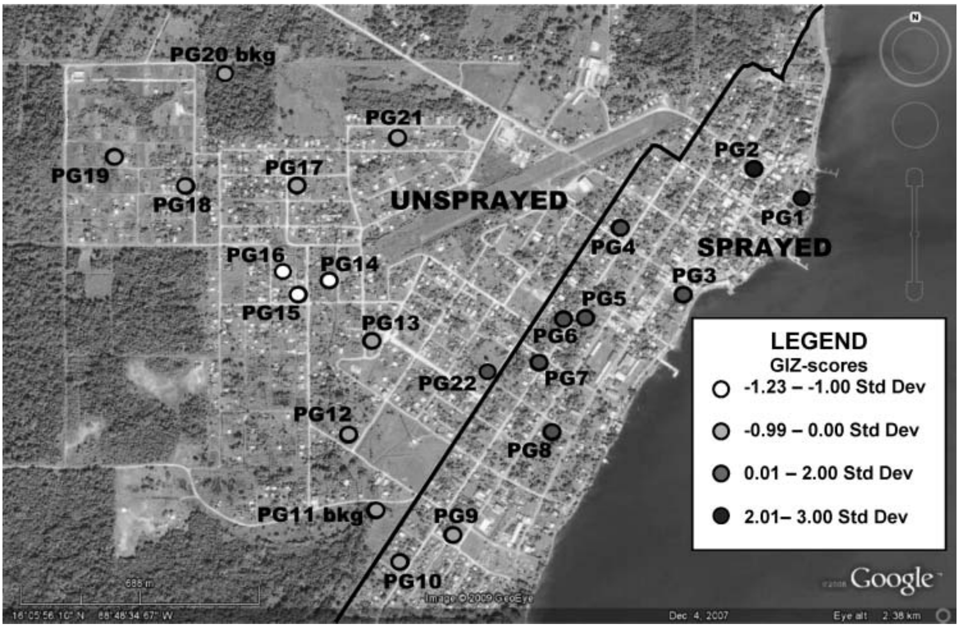

Figure 5. Hot spot in Punta Gorda using total DDT concentrations. Background image source: Google Earth © 2008 Google, image © 2009 GeoEye.

highest $z = 3.43$, $p = 0.0006$) in the southern part of the sprayed area of the town. In contrast, DDE and DDD concentrations revealed no hot spots, which is consistent with the observation made based on compound ratios that DDT may have been used relatively recently in this town. The DDT and total DDT hotspot suggests that the south part of Dangriga was sprayed most intensely.

For Punta Gorda, total DDT concentration values revealed one hot spot in the north of the sprayed region of this town (highest $z = 2.74$, $p = 0.006$) (Figure 5). Hot spots for DDT and DDE are in the same general area but are larger than the one for total DDT (DDT: highest $z = 2.92$, $p = 0.003$; DDE: highest $z = 3.47$, $p = 0.0005$). A weak hot spot for the DDD concentration (highest $z = 4.52$, $p = 0.000006$) exists just to the south of the total DDT hotspot. The general co-occurrence of the hotspots indicates that the northern part of Punta Gorda may have been sprayed most intensely and that the DDT metabolites DDE and DDD locally accumulated in the soil. The houses in the hot spots in both towns seem to be older than elsewhere in the sprayed areas but no firm statistical conclusion can be drawn about house age being a co-determinant in the location of the hot spots. Although these hot spots represent locally high values the concentrations of the pesticides show that the hot spots are not of immediate concern for human health.

Conclusions

Widespread use of DDTs ceased in Belize after 1997 but results indicate that DDTs still persist in soils in southern Belize after more than 10 years without official use. Elevated concentrations of DDTs were found locally in the soil environment of the two towns of Dangriga and Punta Gorda. Concentrations in sprayed areas of the towns were significantly higher than in the unsprayed areas, suggesting that DDTs are primarily transferred to the soil environment by spraying houses. Greater abundance of DDT than DDE and DDD at some sites might be a result of post-1997 usage of this pesticide. Half of the sites in the unsprayed area of Punta Gorda had low but detectable concentrations of some of the DDTs, indicating that atmospheric or surface transport of DDTs affects parts of this unsprayed area.

This study found little evidence for influence of soil characteristics (such as particle size, organic matter, and pH) on concentrations of DDTs. Others have explained similar observations by differences in spraying intensity that dominate the effects of soil characteristics on short timescales. Low correlations between concentrations of DDTs and soil characteristics in the present study area are hard to interpret because of potential variability in the fumigation process. However, lower clay content in the sprayed than the unsprayed areas shows that this soil characteristic does not explain the higher concentrations of DDTs in the sprayed areas. This supports the contention that spraying is the cause of the differences in concentrations of DDTs between sprayed and unsprayed areas. The influence of spraying is also corroborated by the moderate-to-high positive correlations between concentrations of DDTs and house age because older houses have had more applications of DDT mixture.

Previous lack of data for DDTs in Belize's soils diminishes the understanding of this pesticide's possible effects on the health of Belizeans and their environment. Belize currently has no guidelines for determining risk of DDTs to human health and soil cleanup. Soil quality guidelines employed by foreign governmental entities such as the Netherlands and the California EPA show that all detected pesticide concentrations in this study are below mandated concentrations for the residential areas of these regions and hence do not appear to present any significant risk to human health. However, since exposure scenarios may be different in Belize, it is recommended that the Belize Ministry of Health conduct a risk analysis to ascertain if these pesticide concentrations pose significant risk to the inhabitants of the studied areas.

GIS analysis identified local zones with statistically higher concentrations in the sprayed areas of both towns. The existence of these hot spots shows that local observations of DDTs can not necessarily be extrapolated spatially, even within relatively small areas. It implies that other studies of DDTs in fumigated areas, and management decisions in such areas, should take into account the possibility of spatial variability.

In Belize, further studies are necessary to investigate whether or not DDTs are indeed being sprayed or applied unofficially in the Belizean environment. In addition, a larger-scale investigation of the spatial and temporal distribution of DDTs in soils in Belize might contribute to the understanding of this pesticide's current worldwide concentrations and trends. Interpreting concentrations of DDTs in Belizean soils and its wider environment must consider contributions from long-range and regional transport because of extensive use of this chemical in neighboring Central American and South American countries.

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References

Adams, L., Hanavan, M. G., Hosley, N. W., and Johnston, D. W. 1949. The effects on fish, birds and mammals of DDT used in the control of forest insects in Idaho and Wyoming. *J. Wildlife Man* **13**(3), 245–254.

Alegria, H. A., Bidleman, T. F., and Shaw, J. S. 2000. Organochlorine pesticides in ambient air of Belize, Central America. *Environ. Sci. Technol.* **34**(10), 1953–1958.

Altschuh, J., Bruggemann, R., Santl, H., Eichinger, G., and Piringer, O. G. 1999. Henry's law constant for diverse set of organic chemical: Experimental determination and comparison of estimation methods. *Chemosphere* **39**, 1871–1887.

Andrade, M. L., Reyzabal, M. L., Covelo, E. F., and Vega, F. A. 2005. Organochlorine pesticides in soils of the horticultural belt of Bahia Blanca (Argentina). *Can. J. Soil Sci*. **85**(2), 273–282.

ATSDR (Agency for Toxic Substances and Disease Registry). 2002. *Toxicological Profile for DDT, DDE, and DDD*. US Department of Health and Human Services, Public Health Service, Atlanta, GA.

Baczynski, T. P., Pleisnner, D., and Grotenhuis, T. 2010. Anaerobic biodegradation of organochlorine pesticides in contaminated soil—Significance of temperature and availability. *Chemosphere* **78**, 22–28.

Barrick, R. C., and Prahl, P. G. 1987. Hydrocarbon geochemistry of the Puget Sound regions-III. Polycyclic aromatic hydrocarbons in sediments. *Est. Coast. Shelf Sci.* **25**, 175–191.

Belize National Malaria Eradication Service. 2003. *Annual Quantity of DDT used in Belize for the Period 1985–1994*. Ministry of Health, Belmopan, Belize.

Belize National Meteorological Service. 2010. http://www.hydromet.gov.bz/ [Accessed 1 March 2010].

Brown, L. R. 1997. Concentrations of chlorinated organic compounds in biota and bed sediment in streams of the San Joaquin Valley, California. *Arch. Enviro. Cont. Toxic.* **33**, 357–368.

Burkhard, N., and Guth, J. A. 1981. Chemical hydrolysis of 2-chloro-4,6-bis-(alkylamino)-1,3,5-triazine herbicides and their breakdown in soil under the influence of adsorption. *Pest. Sci.* **12**(1), 45–52.

Burt R. (ed.). 2004. *Soil Survey Laboratory Methods Manual*. Soil Survey Investigations Report No. 42. USDA NRCS, Washington, DC.

California EPA. 2005. *Use of California Human Health Screening Levels (CHHSLs) in Evaluation of Contaminated Properties*. California Department of Environmental Protection, Sacramento, CA.

Callaway, E. 2008. Melting glaciers release toxic chemical cocktail. *New Scientist*. http://environment.newscientist.com/article/dn13848 [Accessed 5 July 2008].

Chapin, G., and Wasserstrom, R. 1981. Agricultural production and malaria resurgence in Central America and India. *Nature* **293**(5829), 181–185.

Chassin, P., and Calvet, R. 1984. Retention of herbicide molecules in soils: Reflections on the role of organic matter. In: *Proceedings of the Congress: Comportement et Effets-secondaires des Pesticides dans le Sol*, pp. 25–133. INRA, Versailles, France.

Cheng, H. H. 1990. *Pesticides in the Soil Environment: Processes, Impacts, and Modeling*. Soil Science Society of America, Madison, WI.

Donzel, B., and Dorn, E. 1992. Fate of agrochemicals in plants and soil: A cost-effective integrated approach. *Pure Applied Chem*. **64**(12), 1965–1976.

ESRI (Environmental Systems Research Institute). 2009. *Hot Spot Analysis* (Getis-Ord Gi* Spatial Statistics). http://webhelp.esri.com/arcgisdesktop/9.3/index.cfm?TopicName=Hot_Spot_Analysis_(Getis-Ord_Gi*)_(Spatial_Statistics) [Accessed 22 January 2009].

Falandysz, J., Strandberg, L., Puzyn, T., and Gucia, M. 2001. Chlorinated cyclodiene pesticide residues in blue mussel, crab, and fish in the Gulf of Gdansk, Baltic Sea. *Environ. Sci. Technol.* **35**, 4163–4169.

Fernandez, M. 2001. *Diagnostic Situation on the Use of DDT and the Control and Prevention of Malaria in Belize*. http://www.bvsde.paho.org/bvsdepl/fulltext/ddt/BLZ.pdf [Accessed 14 April 2008].

Fu, S., Cheng, H. X., Liu, Y. H., and Xu, X. B. 2009. Levels and distribution of organochlorine pesticides in various media in a mega-city, China. *Chemosphere* **75**, 588–594.

George J. L., and Stickel, W. H. 1949. Wildlife effects of DDT dust used for tick control on a Texas prairie. *Am. Midland Natur.* **42**(1), 228–237.

Gong, Z. M., Tao, S., Xu, F. L., Dawson, R., Liu, W. X., Cui, Y. H., Cao, J., Wang, X. J., Shen, W. R., Zhang, W. J., Qing, B. P., and Sun, R. 2004. Level and distribution of DDT in surface soils from Tianjin, Chin. *Chemosphere* **54**(8), 1247–1253.

Gustafson, D. I. 1989. Groundwater ubiquity score: A simple method for assessing pesticide leachability. *Enviro. Toxic. Chem.* **8**, 339–357.

Heberer, T., and Dunnbier, U. 1999. DDT metabolite bis(chlorophenyl)acetic acid: The neglected environmental contaminant. *Environ. Sci. Technol.* **33**, 2346–2351.

Hrdina, P. D., Singhal, R. L., Ling, G. M. 1975. DDT and related chlorinated hydrocarbon insecticides: pharmacological basis of their toxicity in mammals. *Adv. Pharmacol. Chemother.* **12**(0), 31–88.

Instituto de Investigaciones Agropecuarias. 1990. *Fuentes de Contaminacion con Residuos de Plaguicidas Organoclorados y Metales Pesados en Sectores Agrıcolas*. Regiones IV a XI. Ministerio de Agricultura, Chile.

Jackson, M. L. 1964. *Soil Chemical Analysis*. Prentice-Hall, Englewood Cliffs, NJ.

Jaga, K., and Dharmani, C. 2003. Global surveillance of DDT and DDE levels in human tissues. *Int. J. Occup. Med. Enviro. Health.* **16**, 7–20.

Kannan, K., Tanabe, S., and Tatsukawa, R. 1995. Geographical distribution and accumulation features of organochlorine residues in fish of tropical Asia and Oceania. *Environ. Sci. Technol.* **29**(10), 2673–2683.

Ministry of Housing. 2000. *Circular on Target Values and Intervention Values for Soil Remediation*. Ministry of Housing, Spatial Planning and Environment, Directorate-General for Environmental Protection. http://www.vrom.nl/37604.

Morrison, R. D., and Newel, A. E., 1999. The cosolvation transport of DDT and toxaphene in xylene at a pesticide formulation facility. *J. Soil Contam.* **8**, 63–80.

Nash, R. G., and Woolson, E. A. 1967. Persistence of chlorinated hydrocarbon insecticides in soils. *Science* **157**(3791), 924–927.

Racke, K. D., and Coats, J. R. 1990. *Enhanced Biodegradation of Pesticides in the Environment* (Symposium Series No. 426). American Chemical Society, Washington, DC.

Racke, K. D., Skidmore, M. W., Hamilton, D. J., Unsworth, J. B., Miyamoto, J., and Cohen, S. Z. 1997. Pesticide fate in tropical soils. *Pure and Applied Chem*. **69**(6), 1349–1371.

Samuel, T., and Pillai, M. K. K. 1989. The effect of temperature and solar radiation on volatilisation, mineralisation, and degradation of 14C-DDT in soil. *Enviro. Poll.* **57**, 63–77.

Sanchez, P. A. 1976. *Properties and Management of Soils in the Tropics*. John Wiley and Sons, New York.

Saxena, S., and Siddiqui, M. 1980. Role of chlorinated hydrocarbon pesticides in abortions and premature labour. *Toxicology* **17**, 323–331.

Schnitzer, M., and Khan, S. 1978. *Soil Organic Matter*. Elsevier, New York.

Sotomayor-Rios, A., and Pitman, W. D. 2000. *Tropical Forage Plants: Development and Use*. CRC, Boca Raton, FL.

Spark, K. M., and Swift, R. S. 2002. Effect of soil composition and dissolved organic matter on pesticide sorption. *Sci. Tot. Environ.* **298**, 147–161.

Statistical Institute of Belize. 2010. Mid-year population by district and sex, 2009. http://www.statisticsbelize.org.bz/[Accessed 1 March 2010].

Tanabe, S., Iwata H., and Tatsukawa R. 1994. Global contamination by persistent organochlorines and their ecotoxicological impact on marine mammals. *Sci. Total Enviro.* **154**(2-3), 163–177.

UNEP (United Nations Environment Programme). 2003. *Regionally Based Assessment of Persistent Toxic Chemicals, Global Report*. United Nations Environment Programme, Chemicals Division. www.chem.unep.ch/pts [Accessed 3 July 2008].

USEPA (United States Environmental Protection Agency), Region V. 2007. *FIELDS Statistical Evaluation Report: South Minneapolis Soil Contamination Superfund Site*. http://www.health.state.mn.us/divs/eh/hazardous/sites/cass/stregis/stregishc0408.pdf [Accessed 14 October 2008].

USEPA (United States Environmental Protection Agency). 2008a. *3000 Series Methods*. http://www.epa.gov/wastes/hazard/testmethods/sw846/online/3_series.htm [Accessed 2 May 2008].

USEPA (United States Environmental Protection Agency). 2008b. *8000 Series Methods*. http://www.epa.gov/wastes/hazard/testmethods/sw846/online/8_series.htm [Accessed 2 May 2008].

USEPA (United States Environmental Protection Agency). 2008c. *9000 Series Methods*. http://www.epa.gov/wastes/hazard/testmethods/sw846/online/9_series.htm [Accessed 2 May 2008].

Vieira, E. D. R., Torres, J. P. M., and Malm, O. 2001. DDT environmental persistence from its use in a vector control program: A case study. *Environ. Res. Sect. A.* **86**, 174–182.

Walker, A., Turner, I. J., Cullington, J. E., and Welch, S. J. 1999. Aspects of the adsorption and degradation of isoproturon in a heavy clay soil. *Soil Use Man*. **15**(1), 9–13.

Ware, G. W. 1975. Effects of DDT on reproduction in higher animals. *Residue Rev.* **59**, 119–140.

Wenzel, K. D., Manz, M., Hubert, A., and Schüürmann, G. 2002. Fate of POPs (DDX, HCHs, PCBs) in upper soil layers of pine forests. *Sci. Total Enviro.* **286**, 143–154.

WHO (World Health Organization). 2006. *Indoor Residual Spraying: Use of Indoor Residual Spraying for Scaling Up Global Malaria Control and Elimination*. http://whqlibdoc.who.int/hq/2006/WHO_HTM_MAL_2006.1112_eng.pdf [Accessed 23 August 2007].

Wilderness Group Consulting. 2006. *Environment and Human Health Concerns Related to the Use of Persistent Organic Pollutants in Belize* (BELIZE POPS-003). Department of the Environment, Belmopan, Belize.

Xu, D., Dan, M., Song, Y., Chai, Z., and Zhuang, G. 2005. Concentration characteristics of extractable organohalagens in PM2.5 and PM10 in Beijing, China. *Atmos. Enviro*. **39**, 4119–4128.

Yang, L. Y., Xia, X. H., Liu, S. D., and Bu, Q. W. 2010. Distribution and sources of DDTs in urban soils with six types of land use in Beijing, China. *J. Haz. Mat.* **174**, 100–107.

Yoshida, T. 1975. Microbial metabolism of flooded soils. In: *Soil Biochemistry*, pp. 83–122 (Paul, A., and McLaren, A. D., eds.), Marcel Dekker, New York.