academicJournals

Vol. 7(12), pp. 382-393, December, 2013
DOI: 10.5897/AJPAC2013.0531
ISSN 1996 - 0840 ©2013 Academic Journals
http://www.academicjournals.org/AJPAC

African Journal of Pure and Applied Chemistry

Full Length Research Paper

Status and distributions of pesticides buried at five sites in Arusha and Mbeya regions, Tanzania

John A. M. Mahugija

Chemistry Department, University of Dar es Salaam, P. O. Box 35061 Dar es Salaam, Tanzania.

Accepted 21 November, 2013

This study investigated the compositions and distributions of pesticides buried at five sites in Tanzania. Samples were collected from various depths up to 220 cm. The analytes in samples were extracted by pressurized fluid extraction. Labelled internal standards were used for identification and quantification. Clean-up of extracts was conducted by using column chromatographic techniques. Determination of the analytes was performed by using a high resolution GC–MS. The dominant compounds detected were 4,4'-DDT (dichlorodiphenyltrichloroethane), 2,4'-DDT, 4,4'-DDE, 2,4'-DDE, 4,4'-DDD, 2,4'-DDD, hexachlorocyclohexane (HCH) isomers (α-, β-, γ-, δ- and ε-HCH), aldrin, dieldrin, endrin, pentachlorobenzene, hexachlorobenzene, pentachloroanisole, endosulfans, chlordanes, heptachlor and heptachloroepoxides. The highest concentrations of total DDT and total HCH ranged 5.2 to 5410 mg/kg dry weight (dw) and 1.4 to 42200 mg/kg dw, respectively, while the highest concentrations for other compounds varied from 1.1 to 7200 mg/kg dw. The results indicated aged contaminants but the rate of degradation was generally very slow. The concentrations of the compounds decreased with increase in distance from the burial points. There were variations in the distribution of the compounds among the sampling sites and depths; some showed even distribution or accumulation downwards indicating some risks for groundwater contamination.

Key words: Buried pesticides, soil, Tanzania.

INTRODUCTION

The banning of pesticide introductions into certain countries, poor storage and lack of store management, unawareness, overstocking, inadequate assessment of needs, difficulties in forecasting outbreaks of pests, inappropriate formulations and containers supplied by pesticide distributors, excessive or uncoordinated donations are among the factors which contribute to the accumulation of obsolete pesticides in developing countries (FAO, 1998, 2001). Common types of obsolete pesticides include organochlorine pesticides such as dichlorodiphenyltrichloroethane(DDT),hexachlorocylohexanes (HCHs), aldrin, dieldrin, endrin, hexachlorobenzene, chlordane and heptachlor, which have been withdrawn or banned for public health and environmental reasons (FAO, 1996, 2001). Organochlorine pesticides are described as persistent organic pollutants (POPs) which are organic compounds that, to a varying degree, resist photolytic, biological and chemical degradation in natural environments. POPs are often characterized by low water solubility and high lipid solubility, leading to their bioaccumulation in fatty tissues and toxicity. They are also semi-volatile, enabling them to move long distances in the atmosphere before deposition occurs (Ritter et al., 1995).

There are no easy disposal methods that are safe, cheap and generally applicable under circumstances

E-mail: johnmahugija@yahoo.com. Tel: + 255-222410038.

prevailing in developing countries. As a result, the pesticides are improperly disposed, which often leads to serious environmental contamination. The common disposal methods used include open burning, discharge to sewer, burying or landfill disposal, which are likely to cause severe damage to public health and the environment (FAO, 1996).

The problem of pesticide burial sites exists in most African, East European and Caucasian countries. For example, significant contamination has been found in soil around sites where tonnes of obsolete pesticides were buried in Belarus (PAN UK, 2005), Armenia (Manvelyan et al., 2006), Poland (Gałuszka et al., 2011) and other countries. Typically, the content of buried pesticides is dominated by organochlorine pesticides. Other pesticide types, such as organophosphates, carbamate pesticides, dinitrophenols, phenoxyacids, and inorganic compounds are stored in smaller quantities (Gałuszka et al., 2011).

Soil is an important component of the terrestrial ecosystem. It generally acts as a primary sink and emission source for contaminants (Rauf et al., 2002). The concentrations and distribution features of pesticides in soil profiles are subject to physical, chemical and biological processes. The fate processes of pesticides in the environment are divided into three major types: (i) Adsorption, which is the binding of pesticides to mineral or organic matter; (ii) Transfer processes that move pesticides in the environment; and (iii) Degradation processes that break down pesticides (Harrison, 1990; Sun et al., 1997).

The tendency of different pesticides to adsorb to soil varies with their chemical and physical characteristics. Some pesticides, bind very tightly while others bind only weakly and are readily desorbed, that is, released back into the soil solution. Adsorption influences whether a pesticide is free to enter into other biological and chemical pathways. Since a strongly adsorbed pesticide is less likely to be dissolved in water, it is also less likely to be absorbed into plant roots and soil organisms or to leach beyond the root zone (Harrison, 1990; EXTOXNET, 1993; Waldron, 1992; Mansour, 1993). Many soil characteristics influence pesticide adsorption, including structure, texture, organic matter content, pH, and moisture content. Soil texture affects movement of water through soil and, therefore, affects the movement of dissolved pesticides. The coarser the soil, the faster the movement of the percolating water, and the less opportunity for adsorption of dissolved chemicals. Soils with more clay tend to hold water and dissolved chemicals longer. These soils also have far more surface area on which pesticides can be adsorbed (McEwen and Stephenson, 1979; Harrison, 1990). Soil organic matter influences how much water a soil can hold and how well it will adsorb pesticides. Increasing fractions of organic matter increases the adsorptive capacity of soils. This characteristic is due in part to particles of organic matter providing soils with a large number of sites, onto which pesticides can bind. The chemical nature of a pesticide determines sorption equilibria by influencing its direct affinity for the clay or organic matter or by influencing its solubility or affinity for the soil solution (McEwen and Stephenson, 1979; Harrison, 1990; Mansour, 1993). Soils with highly developed structure may encourage the movement of pesticide-laden water between the structural units, known as peds, without contacting the individual soil particles within the units. This has the effect of reducing the potential for adsorption (Harrison, 1990). The fate of pesticides in soils varies with pH differences, or the acid/base balance of the soil solution, primarily because of the influence of pH on sorption phenomena (Mansour, 1993). Soil pH also affects chemical degradation. Soil pH impacts the chemical's reactivity and certain soil functions such as microbial metabolism (Buyuksonmez et al., 1999). Wet soils tend to adsorb fewer pesticides than dry soils, either because water molecules compete for the binding sites or because some of the pesticide molecules remain dissolved in the soil water (Harrison, 1990). Soil temperature also affects pesticide adsorption because when the temperature increases, some bonds break and cause the desorption of some pesticide molecules. At high temperatures the greater solubility of pesticides also results into more pesticides being available in the soil solution (McEwen and Stephenson, 1979; Waldron, 1992).

Pesticide transfer refers to the movement of pesticides from their sites of origin. Too much movement can lead to injury of nontarget species (including humans), and surface and groundwater contamination. Five processes that can move pesticides are volatilization, runoff, leaching, absorption, and physical removal. All these are influenced by management practices such as the methods used to store, apply or dispose the pesticides (Harrison, 1990; Waldron, 1992).

Pesticide degradation processes break down pesticide molecules into simpler and, generally, less toxic compounds, although in some cases may be more toxic. The three major types of pesticide degradation are microbial degradation, chemical degradation, and photodegradation (Harrison, 1990; Buyuksonmez et al., 1999).

The aim of this study was to investigate the compositions and distributions of pesticides buried at the five sites located in Arusha and Mbeya regions in Tanzania. There were no clear records as to when the pesticides were buried at the studied sites, but it was generally indicated that the pesticides were buried more than ten years before this study.

MATERIALS AND METHODS

Study areas

The study areas were located in Arusha and Mbeya regions (Figure 1). At the Plant Health Services Office at Tengeru in Arumeru

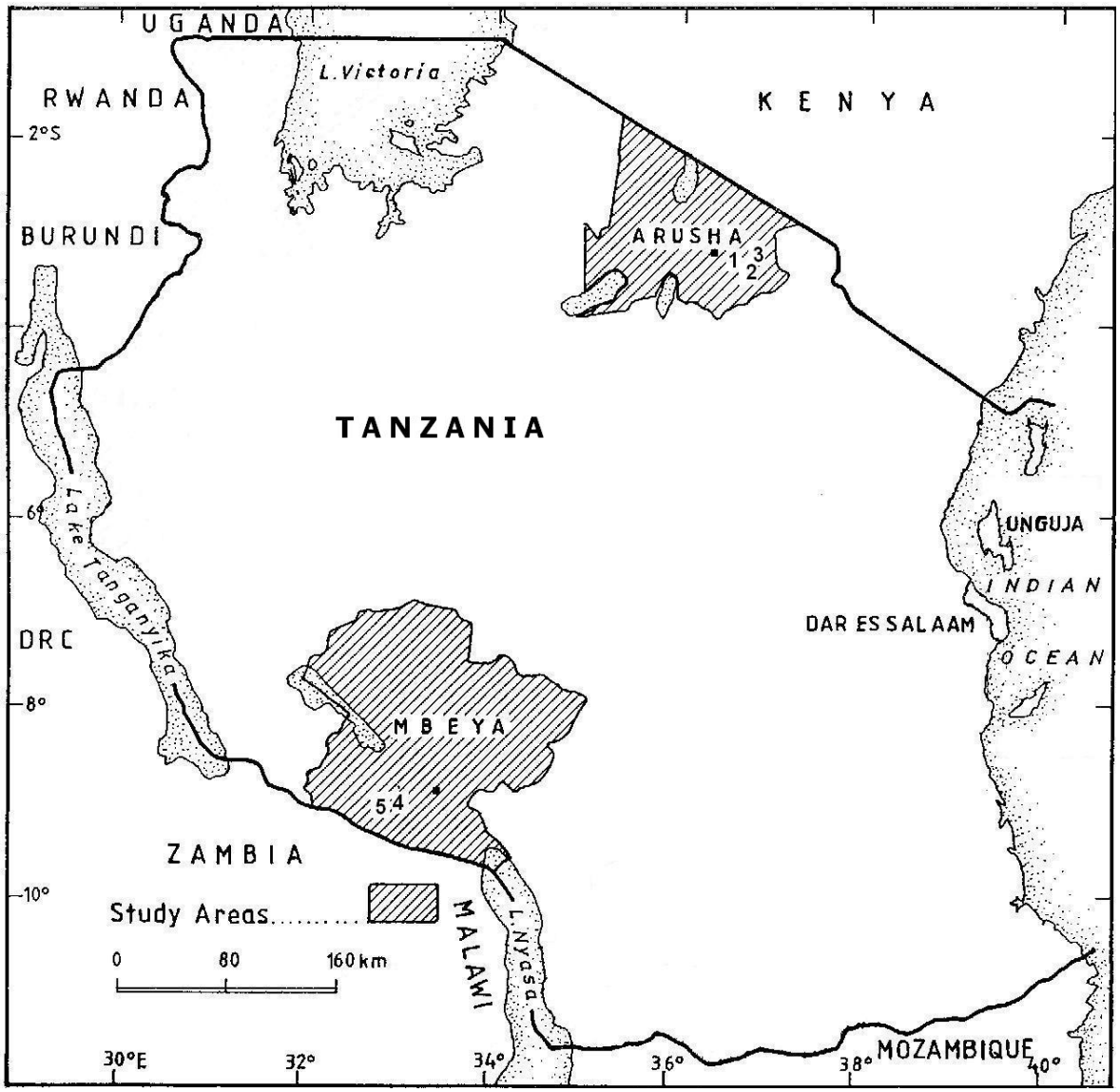


Figure 1. Map showing the sampling sites: *1 = Tengeru, 2 = ACU maize farm (Usa river), 3 = Akheri vegetable garden, 4 = TaCRI-Mbimba, 5 = MBOCU.*

district, Arusha region (locust control centre for Tanzania), pesticides contained in drums were kept outside the office at the backyard area. The drums rusted and the contents leaked into the soil. The drums were removed to another unknown location while the contaminants were buried in soil, covered with a polyethylene sheeting and fresh soil added on top to avoid the odour, but that did not prevent the smell from permeating the area. The location is beside the Plant Health Services Offices and the workers there often complained of the strong smell invading the offices, especially during dry season. The location is at the top of a sloping bank of approximately 3 m. Cultivation activities were being carried out very close to the contaminated area.

Pesticides collected from some stores by the Arusha Co-operative Union (ACU) were buried at another site located in a maize farm owned by ACU at Usa River Chemchem maize farm in Arumeru district, covering an area approximately 160 m^2. No vegetation was found to grow at the area where the pesticides were buried.

At Akheri village in Arumeru district in Arusha region, pesticides collected from a nearby former storage building owned by Meru Co-operative Union, were buried at a small area in a vegetable garden. This was a working garden with continuous planting and harvesting of vegetables such as spinach, onions and cabbages. The pesticides were buried directly in the vegetable garden right in the irrigation path, which implied the possibility of being dispersed and introduced into the food chain.

Other sites where pesticides were buried are Tanzania Coffee Research Institute (TaCRI)-Mbimba and Mbozi Cooperative Union (MBOCU). The TaCRI-Mbimba site is located at Mbimba village in Mbozi district, Mbeya region within the coffee research farm at the Tanzania Coffee Research Institute. At this site, pesticides were dumped and buried in a pit of an area approximately 25 m^2. At MBOCU headquarters in Vwawa town, Mbozi district in Mbeya region, pesticides and empty containers were dumped in a pit of about 42 m^2. The site is located very close to residential areas and

office buildings. Cultivation was being carried out in the vicinity of this site.

Sampling

Soil samples were collected from five sites located in Arusha and Mbeya regions (Figure 1). Soil samples from Tengeru, ACU maize farm and Akheri were collected in January 2009. Samples from Mbimba and MBOCU were collected in April 2009. Samples were collected at points within the burial areas and approximately 5 to 60 m from those points down and up slope. Soil samples were collected from 19 points at Tengeru and the sampling depths were 30, 75, 150 and 220 cm. At ACU maize farm, samples were collected at 10, 30 and 75 cm depths and were from 13 points. The sampling depths at Akheri were 10, 30 and 50 cm and the samples were collected from 8 points. The samples were collected from 50 and 100 cm at Mbimba, while 10, 50 and 100 cm were the sampling depths for MBOCU.

Sampling was conducted applying the standard guidelines (Åkerblom, 1995). The tools used for sampling of soil included clean spades, hoes, clean aluminium foil, clean buckets, measuring tapes, folding rulers, insulating boxes, solvents for cleaning, bottles for waste solvents, kitchen roll papers, polyethylene bags, waste plastic bags and stainless steel spoons. Using a hoe and spade, a test pit was prepared a little deeper than the desired depth, then using a clean spade a slice about 5 cm thick was made along the vertical wall of the pit at the desired depth and the soil was thrown away. Another clean spade or spoon was used to take the sample. Samples at deeper points and presumably less contaminated points were collected first before the surface or presumably contaminated depths or points. A sample was obtained by collecting at least five subsamples from different points at the same depth within the pit. The stones, sticks, plant roots and other unwanted materials were removed by using a clean spoon. The sample was ground and mixed very thoroughly on aluminium foil. The sample was immediately wrapped in an aluminium foil and placed in a polyethylene bag then put in an insulated box. The samples were transported to the laboratory and stored in a freezer at -28°C until extraction.

Extraction, clean up and gas chromatographic analysis

Extraction, clean up and analysis of the soil samples were conducted at the Institute of Ecological Chemistry, German Research Centre for Environmental Health in May – October 2009. The procedures by Schramm et al. (2008) were adopted with modifications. Soil samples were extracted by pressurized fluid extraction using an Accelerated Solvent Extractor (ASE 200 Dionex). A cellulose filter was inserted into the inner bottom of the extraction cell, then sea sand dried at 550°C (ca. 1 g) was added. The sample (0.5-5 g) mixed with hydromatrix for drying and dispersing was added into the cell and a filter placed on top. Prior to extraction, the sample was spiked with ^{13}C–labelled and deuterated internal standards (10 µl of a mixture containing 333-1000 pg/µl of organochlorine compounds in nonane). All the internal standards for the compounds determined were ^{13}C–labelled except for 4,4′-DDD, which was a deuterated standard. Highly contaminated samples were not spiked with internal standards prior to extraction, but the diluted extracts were spiked during clean-up. The samples were quantitatively extracted by an accelerated solvent extractor at a temperature of 120°C and pressure of 120 bar and with *n*-hexane:acetone (75:25) as the extraction solvent mixture. Two static cycles of 10 min were applied for a complete extraction. Another sub-sample of each sample was dried for 24 h at 105°C and then weighed for moisture and dry weight determination. The extracts were passed over anhydrous sodium sulfate to remove water. The extracts were concentrated using vacuum rotary evaporation and the solvent was reconstituted to hexane:dichloromethane (1:1).

Clean-up of the concentrated extracts (0.5-1 ml for undiluted extracts or 50-100 µl of diluted extracts for highly contaminated samples) to remove interferences was conducted by using 10 g silica gel (grade 60), 5 g alumina with 3% H_2O and 5 g anhydrous sodium sulfate, packed in that order in glass column (30 cm long with an internal diameter of 2.5 cm). The extracts were eluted with 100 ml of a mixture of hexane and dichloromethane (1:1) at a flow rate of *ca*. 2 drops per second (about 0.1 ml/s) and concentrated to 1 ml using a rotary evaporator, then using a very gentle stream of nitrogen to ca. 0.2 ml. The solvent was changed to acetonitrile and concentrated using nitrogen to ca. 0.2 ml. Further clean-up was performed through a C18 SPE cartridge using 1 g C_{18}-modified silica gel and the eluting solvent used was acetonitrile (5 ml). The extracts were concentrated by blowing a very gentle stream of nitrogen to ca. 0.2 ml. The concentrated extracts were transferred into clean vials containing a recovery standard (20 µl of a 1 ng/µl solution of ^{13}C-pentachlorotoluene and ^{13}C-1,2,3,7,8,9-hexachlorodibenzo-*p*-dioxin in nonane) and extracts were concentrated with a gentle flow of nitrogen to 20 µl ready for analytical determination.

Instrumental analysis of the organochlorine pesticides and metabolites was performed by using a high resolution gas chromatograph coupled to a high resolution mass spectrometer (HRGC–MS). An Agilent 6890 GC equipped with a capillary column (Rtx-Dioxin2, 40 m, 0.18 mm ID, 0.18 µm film thickness, Restek) was used. The temperature program was 60°C (1.5 min), 25°C/min to 140°C (0 min), 8°C/min to 300°C (20 min). 0.5 µl was injected using an autosampler (A200S, CTC) in pulsed splitless mode by a cold injection system CIS 4 (Gerstel). The temperature programme for the injector was: 120°C, 12°C/s, 280°C, 5 min. The carrier gas was helium in a constant flow of 1.3 ml/min. The temperature at the transferline was 300°C. The measurement was conducted with a Finnigan MAT 95S mass spectrometer (Thermo) with a resolution of 10 000. The ionisation mode was EI at 50 eV and 260°C and the detection was by using the selected ion monitoring (SIM) mode. The two most intense ions of the molecular ion cluster were monitored for the analytes and labelled standards. The identification and quantification criteria included confirmation of retention times, relative retention times and isotope ratios for the labelled standards and respective analytes. The mass fragment with the highest intensity of the molecular ion was used for quantification while the other was used as a ratio mass (Schramm et al., 2008).

Analytical quality assurance and control

Separate tools were used to collect different samples from different depths and points. Tools to be reused were thoroughly cleaned with water and soap and rinsed with dichloromethane and acetone. The labelled pesticide standards were of over 99% certified purity (obtained from Dr. Ehrenstorfer, Augsburg, Germany). The standard pesticide solutions and samples were stored in glass-stoppered flasks or vials in deep freezers at –28°C. All organic solvents were of picograde quality (obtained from LGC Promochem, Wesel, Germany). After use, all glassware and tools were rinsed with a technical mixture of toluene, acetone and hexane, and washed with water and detergent in a washing machine. Thereafter, the glassware were dried in an oven overnight at programmed temperatures up to a maximum temperature of 450°C. The silica gel was heated overnight at 550°C to reduce background levels. Analysis of blanks, certified reference materials and recovery tests were used to check contamination and performance of the method. No significant peaks appeared in the chromatograms of the blanks. Recoveries of labelled internal standards varied between 51 and 120% with average recoveries ranging from 76 to 112% and the

Table 1a. Minimum, maximum and mean concentrations and detection frequencies of pesticides and degradation products in soil at Tengeru -Plant Health Services Office.

Compound	Concentration (mg/kg dw), n = 54		Detection frequency (%)
	Min–Max	Mean	
4,4'-DDT	0.0014–4800	110	100
2,4'-DDT	0.0004–435	11	100
4,4'-DDD	0.0002–88	3.3	100
2,4'-DDD	nd–9.0	0.3	94.4
4,4'-DDE	0.00014–110	3.3	100
2,4'-DDE	nd–10	0.24	98.1
Total DDT	0.0024–5410	124	100
α-HCH	0.0001–5000	92.4	100
β-HCH	0.0001–18000	324	100
γ-HCH	nd–13000	232.2	98
δ-HCH	0.0001–3200	59.1	100
ε-HCH	nd–3011	56	98
Total HCH	0.0004–42200	764	100
Pentachlorobenzene	nd–52	1.0	92.6
Hexachlorobenzene	nd–2.0	0.041	98
Pentachloroanisole	nd–0.082	0.002	74.1
trans-Chlordane	nd–0.061	0.002	50
cis-Chlordane	nd–0.019	0.0007	32
oxy-Chlordane	nd	nd	0
Heptachlor	nd–0.094	0.0018	42.6
cis-Heptachloroepoxide	nd–0.00004	0.00001	59.3
trans-Heptachloroepoxide	nd–0.0002	0.00002	27.8
Aldrin	nd–2.2	0.04	90.7
Dieldrin	0.00004–20.2	0.4	100
Endrin	nd–0.09	0.002	61.1
Endosulfan-I	nd–0.31	0.01	70.4
Endosulfan-II	nd–0.21	0.008	74.1
Methoxychlor	nd–0.0003	0.00001	64.8
Mirex	nd	nd	0

nd = not detected, dw = dry weight.

coefficients of variation were in the range of 4 to 19%. The detection limit was defined as three times the average noise value measured. The limits of detection ranged from 0.001 to 0.005 ng/g. The limit of quantification was three times the limit of detection and every signal below this limit was treated as not detectable.

RESULTS AND DISCUSSION

Composition of pesticides and degradation products

The concentrations of the compounds detected in soil samples are presented in Tables 1 to 4. The major contaminants detected in soil samples included DDT isomers and their major degradation products (dichlorodiphenyldichloroethane (DDD) and dichlorodiphenyldichloroethylene (DDE), HCH isomers (α-HCH, β-HCH, γ-HCH, δ-HCH and ε-HCH), aldrin, dieldrin, endrin, endosulfans, pentachlorobenzene, hexachlorobenzene, chlordanes, and heptachlors.

DDT, DDE and DDD

The highest concentrations of total DDT (4,4'-DDT + 2,4'-DDT + 4,4'-DDD + 2,4'-DDD + 4,4'-DDE + 2,4'-DDE) in samples varied from 5.2 mg/kg dw (ACU maize farm) to 5410 mg/kg dw (Tengeru). The highest concentrations of DDT isomers and degradation products ranged as follows: Tengeru 9.0 mg/kg dw (2,4'-DDD) to 4800 mg/kg dw (4,4'-DDT), ACU maize farm 0.035 mg/kg dw (2,4'-DDE) to 3.0 mg/kg dw (4,4'-DDT), Akheri vegetable garden 1.1 mg/kg dw (2,4'-DDE) to 70 mg/kg dw (4,4'-DDT), Mbimba 4.0 mg/kg dw (2,4'-DDE) to 45 mg/kg dw (4,4'-DDT) and MBOCU 6.1 mg/kg dw (2,4'-DDD) to

Table 1b. Distribution of the concentrations of DDT, DDD and DDE in soil samples at Tengeru.

Compounds	Concentrations (mg/kg dw)						
	Burial point (2 sampling points)						
	30 cm, n = 2		75 cm, n = 2		150 cm, n = 2		220 cm
	Min–Max	Mean	Min–Max	Mean	Min–Max	Mean	n = 1
4,4'-DDT	0.015–47	24	900.4–4800	2850	0.41–0.83	0.62	3.022
2,4'-DDT	0.001–4.4	2.201	150–435	293	0.018–0.075	0.047	0.4
4,4'-DDD	0.0002–2.520	1.3	87–88	87.5	0.018–0.0553	0.037	0.046
2,4'-DDD	0.00003–0.634	0.32	4.3–9.0	6.7	0.0004–0.0231	0.012	0.005
4,4'-DDE	0.005–3.51	1.8	59–110	85	0.0683–0.0934	0.081	0.078
2,4'-DDE	0.0001–0.12	0.06	2.81–10	6.4	0.0022–0.003	0.003	0.0053
Total DDT	0.021–58	29	1210–5410	3310	0.542–1.10	0.821	4.0
(DDE+DDD)/DDT	0.1–0.3	0.2	0.04–0.2	0.12	0.2–0.3	0.3	0.04
	Down slope (10–60 m from burial point, 8 sampling points)						
	30 cm, n = 8		75 cm, n = 8		150 cm, n = 8		220 cm
	Min–Max	Mean	Min–Max	Mean	Min–Max	Mean	
4,4'-DDT	0.026–1.3	0.240	0.008–0.464	0.13	0.006–0.900	0.18	na
2,4'-DDT	0.0023–0.12	0.026	0.002–0.056	0.019	0.0013–0.11	0.0252	na
4,4'-DDD	0.0007–0.083	0.016	0.0011–0.19	0.031	0.0006–0.02	0.0042	na
2,4'-DDD	0.0001–0.0053	0.0011	0.0003–0.032	0.0052	0.0003–0.0023	0.0007	na
4,4'-DDE	0.0057–2.0	0.533	0.0011–0.0272	0.0132	0.002–0.014	0.0058	na
2,4'-DDE	0.0001–0.053	0.0092	0.0001–0.0012	0.0005	0.0001–0.0006	0.0003	na
Total DDT	0.041–3.41	0.80	0.013–0.56	0.20	0.011–1.0	0.21	na
(DDE+DDD)/DDT	0.1–6.9	1.8	0.1–0.9	0.4	0.03–0.5	0.2	na
	Up slope (5–20 m from burial point, 9 sampling points)						
	30 cm, n = 9		75 cm, n = 9		150 cm, n = 9		220 cm
	Min–Max	Mean	Min–Max	Mean	Min–Max	Mean	
4,4'-DDT	0.0014–0.083	0.023	0.0052–0.0454	0.0223	0.003–0.063	0.0174	na
2,4'-DDT	0.0004–0.011	0.003	0.0007–0.0084	0.0041	0.0005–0.0133	0.0035	na
4,4'-DDD	0.0002–0.0023	0.0011	0.0005–0.0021	0.0013	0.0008–0.002	0.0011	na
2,4'-DDD	nd–0.0004	0.0002	0.0001–0.0006	0.0003	0.0002–0.00042	0.0003	na
4,4'-DDE	0.0002–0.09	0.023	0.0011–0.046	0.0077	0.0008–0.0033	0.002	na
2,4'-DDE	nd–0.0011	0.0002	0.00003–0.001	0.0002	0.00004–0.0002	0.0001	na
Total DDT	0.003–0.18	0.05	0.0081–0.0742	0.036	0.0052–0.08	0.025	na
(DDE+DDD)/DDT	0.1–2.0	0.8	0.1–1.9	0.4	0.04–0.7	0.3	na

nd = not detected; na = not analyzed.

324 mg/kg dw (4,4'-DDT). The average concentrations were in the following orders:

Tengeru: 4,4'-DDT > 2,4'-DDT > 4,4'-DDD ≈ 4,4'-DDE > 2,4'-DDD ≈ 2,4'-DDE
ACU maize farm: 4,4'-DDT > 2,4'-DDT > 4,4'-DDD ≈ 4,4'-DDE > 2,4'-DDD > 2,4'-DDE
Akheri garden: 4,4'-DDT > 2,4'-DDT > 4,4'-DDD > 2,4'-DDD ≈ 4,4'-DDE > 2,4'-DDE
Mbimba: 4,4'-DDT > 2,4'-DDT > 4,4'-DDE > 4,4'-DDD >2,4'-DDD > 2,4'-DDE
MBOCU: 4,4'-DDT >4,4'-DDE > 2,4'-DDT > 4,4'-DDD >2,4'-DDD ≈ 2,4'-DDE

The compositions of total DDT, that is, the relative distribution of DDT isomers and their degradation products in total DDT in soil at different sampling sites, showed that 4,4'-DDT was the dominant contaminant in the soil samples. The concentrations of 4,4'-DDT were greater than those of 2,4'-DDT in all samples. This indicated that the main source of contamination in the samples from the studied sites was technical DDT. The ratio of 2,4'-DDT/4,4'-DDT can be used to distinguish DDT pollution caused by technical DDT from that due to dicofol. Dicofol would produce a higher 2,4'-DDT/4,4'-DDT ratio than technical DDT. Dicofol is known to contain approximately 3 to 7% DDTs as impurities (Qiu et al.,2005).

Table 1c. Distribution of the concentrations of HCH isomers in soil samples at Tengeru.

Compounds	Concentrations (mg/kg dw)						
	Burial point (2 sampling points)						
	30 cm, n = 2		75 cm, n = 2		150 cm, n = 2		220 cm
	Min–Max	Mean	Min–Max	Mean	Min–Max	Mean	n = 1
α-HCH	0.0041–5000	2500	0.113–2.44	1.3	0.0027–0.0113	0.007	0.034
β-HCH	0.0052–18000	9000	0.0743–74	37	0.0087–0.147	0.078	0.274
γ-HCH	0.003–13000	6500	nd–37.2	19	0.0021–0.007	0.0046	0.041
δ-HCH	0.0121–3200	1600	0.036–9.0	4.52	0.0034–0.0631	0.0333	0.076
ε-HCH	0.003–3011	1510	nd–1.24	0.622	0.0014–0.033	0.0172	0.20
Total HCH	0.0273–42200	21100	0.223–123.3	62	0.0183–0.261	0.14	0.623
α-/γ-HCH	0.4–1.4	0.9	0.1–15	8	1.3–1.6	1.5	0.8
	Down slope (10–60 m from burial point, 8 sampling points)						
	30 cm, n = 8		75 cm, n = 8		150 cm, n = 8		220 cm
	Min–Max	Mean	Min–Max	Mean	Min–Max	Mean	
α-HCH	0.0002–2.4	0.302	0.00021–0.08	0.014	0.0003–1.0	0.15	na
β-HCH	0.0001–2.42	0.472	0.0004–0.0752	0.015	0.0005–0.173	0.0443	na
γ-HCH	0.0001–0.204	0.0263	0.0001–0.0085	0.0017	0.0001–0.009	0.0015	na
δ-HCH	0.0002–0.5	0.0673	0.00032–0.10	0.015	0.0003–0.0019	0.0007	na
ε-HCH	0.00003–0.17	0.0333	nd–0.027	0.0041	0.0001–0.014	0.0047	na
Total HCH	0.001–4.5	0.9	0.0017–0.29	0.049	0.0016–1.20	0.20	na
α-/γ-HCH	1.2–12	4.0	0.7–33	8.4	1.7–110	23	na
	Up slope (5–20 m from burial point, 9 sampling points)						
	30 cm, n = 9		75 cm, n = 9		150 cm, n = 9		220 cm
	Min–Max	Mean	Min–Max	Mean	Min–Max	Mean	
α-HCH	0.0003–0.013	0.0031	0.0001–0.0077	0.0012	0.0001–0.0083	0.0015	na
β-HCH	0.0004–0.0453	0.0082	0.0003–0.0054	0.0019	0.00011–0.0049	0.001	na
γ-HCH	0.0001–0.016	0.0026	0.00005–0.0006	0.0003	0.00004–0.0003	0.0001	na
δ-HCH	0.0001–0.201	0.027	0.0002–0.0076	0.0013	0.0001–0.0007	0.0002	na
ε-HCH	0.00004–0.047	0.0066	0.00004–0.001	0.0002	0.00002–0.0004	0.0001	na
Total HCH	0.0014–0.322	0.048	0.001–0.014	0.005	0.0005–0.015	0.003	na
α-/γ-HCH	0.8–3.8	1.9	0.5–29	5.1	1.1–28	8.1	na

nd = not detected; na = not analyzed.

DDE and DDD are the two main products of DDT dechlorination. DDT is dechlorinated to DDE under aerobic conditions and reductively dechlorinated to DDD under anaerobic conditions. The ratios of (DDE + DDD)/DDT were employed to assess the significance of degradation (transformation) of DDT in the soil. A high value of (DDE + DDD)/DDT ratio (> 1) indicates aged and microbiologically significantly degraded DDT, while a small value (< 1) indicates non-significantly degraded DDT or recent input (Zhang et al., 2006).

The (DDE+DDD)/DDT ratios were generally low (<1) in most soil samples from Tengeru, ACU maize farm, and all samples from Akheri vegetable garden and Mbimba, suggesting that no significant degradation had occurred to the DDT residues at these sites. The high (DDE+DDD)/DDT ratios (>1) in some soil samples from Tengeru, MBOCU and ACU maize farm indicated that the DDT residues had been significantly transformed into their degradation products at those points.

Hexachlorocyclohexanes

The highest concentrations of total HCH (α-HCH + β-HCH + γ-HCH + δ-HCH + ε-HCH) varied from 1.4 mg/kg dw (ACU maize farm) to 42200 mg/kg dw (Tengeru). The concentrations of the HCH isomers were up to 18000 mg/kg dw (β-HCH, Tengeru).

The composition of total HCH and the average concentrations of HCH isomers revealed a heterogeneous nature in soil samples from Tengeru, ACU maize farm and Akheri vegetable garden. This may be related to the isomerization of HCH isomers during transformation process in soil as well as the differences

Table 2. Concentrations of pesticides in soil at ACU maize farm.

Compounds	Concentration (mg/kg dw), burial point and 5 –10 m away					
	10 cm, n = 13		30 cm, n = 9		75 cm, n = 5	
	Min-Max	Mean	Min-Max	Mean	Min-Max	Mean
4,4'-DDT	0.048–3.0	0.59	0.008–1.403	0.40	0.016–0.59	0.313
2,4'-DDT	0.011–0.95	0.20	0.002–1.10	0.34	0.004–0.30	0.122
4,4'-DDD	0.0069–0.72	0.15	0.0006–0.33	0.065	0.0014–0.11	0.038
2,4'-DDD	0.0025–0.212	0.044	0.0002–0.122	0.028	0.0005–0.059	0.02
4,4'-DDE	0.0124–0.301	0.10	0.0041–0.25	0.061	0.0023–0.068	0.035
2,4'-DDE	0.0004–0.031	0.01	0.0002–0.035	0.012	0.0001–0.0131	0.0072
Total DDT	0.101–5.20	1.10	0.022–3.20	0.90	0.0244–1.0	0.534
(DDE+DDD)/DDT	0.2–0.9	0.5	0.14–1.3	0.4	0.12–0.4	0.22
α-HCH	0.001–0.36	0.083	0.00031–0.033	0.01	0.0015–1.20	0.241
β-HCH	0.0003–0.083	0.027	0.0002–0.054	0.016	0.001–0.213	0.047
γ-HCH	0.0011–0.052	0.011	0.0001–0.014	0.0051	0.0007–0.011	0.0045
δ-HCH	0.0002–0.23	0.045	0.0001–0.054	0.0077	0.0003–0.0054	0.0032
ε-HCH	nd–0.0063	0.0005	nd–0.0003	0.00003	nd	nd
Total HCH	0.0034–0.64	0.17	0.001–0.13	0.039	0.0034–1.40	0.295
α-HCH/γ-HCH	0.04–91	11.1	0.3–15	4.3	1.1–110	24
Pentachlorobenzene	0.0004–0.0281	0.007	0.00012–0.003	0.001	0.00011–0.0014	0.0005
Hexachlorobenzene	0.0091–2.40	0.51	0.0017–0.3	0.054	0.0031–0.019	0.0092
Pentachloroanisole	0.0003–0.0141	0.004	nd–0.004	0.0014	nd–0.004	0.001
trans-Chlordane	nd–0.0021	0.0004	nd–0.00041	0.0001	nd–0.00024	0.0001
cis-Chlordane	nd–0.0018	0.00021	nd–0.0001	0.00002	nd–0.0001	0.00002
oxy-Chlordane	nd	nd	nd	nd	nd	nd
Heptachlor	nd–0.043	0.005	nd–0.0006	0.0001	nd–0.00012	0.00004
cis-Heptachloroepoxide	nd–0.0001	0.00001	nd–0.0103	0.002	nd–0.00002	0.00001
trans-Heptachloroepoxide	nd–0.00051	0.0001	nd–0.061	0.012	nd	nd
Aldrin	0.0022–0.70	0.063	0.00013–0.016	0.0034	0.0002–0.0032	0.0012
Dieldrin	0.111–39.3	5.60	0.0009–2.0	0.431	0.0271–0.182	0.0763
Endrin	nd–0.830	0.086	nd–0.023	0.006	nd–0.0045	0.0015
Endosulfan-I	0.0057–17	1.62	nd–0.204	0.0392	nd–0.03	0.0086
Endosulfan-II	0.01–20	2.52	0.0003–0.45	0.12	0.0038–0.075	0.024
Methoxychlor	nd–0.00072	0.0001	nd–0.0007	0.0001	nd–0.0006	0.00012
Mirex	nd–0.0012	0.0002	nd–0.0001	0.00001	nd	nd

in physico-chemical properties and degradation rates (ATSDR, 2005). β-HCH was the dominant HCH isomer in soil samples from Tengeru. This can be explained by the fact that β-HCH is the most stable and persistent in soil. The equatorial configuration of the chlorine atoms in the molecular structure of β-HCH confers the greatest metabolic stability to this isomer. The persistence of β-HCH in soils is also due to higher K_{ow} and lower vapour pressure than other isomers, which enhance its adsorption to the soil and reduce loss by evaporation from the soils (Willett et al., 1998). β-HCH was also found to be the dominant HCH isomer in some studies (Concha-Grana et al., 2006; Wang et al., 2006; Zhang et al., 2006). The α-isomer was the predominant HCH isomer in all samples from Mbimba, indicating technical HCH as the source of contamination. The γ-isomer was the predominant HCH isomer in all samples from MBOCU, indicating lindane as the main source of contamination. The α-HCH/γ-HCH ratio can be used to identify the source of HCHs contamination. The ratios of α-HCH/γ-HCH are in the range of 4 to 15 for technical mixtures, and almost zero for lindane, or range from 0.2 to 1 due to transformation of γ-HCH into α-HCH. Technical HCHs mixture contains the isomers in the following percentages: 60-70% α-HCH, 5-12% β-HCH, 10-15% γ-HCH, 6-10% δ-HCH and 3-4% ε-HCH, while lindane contains >99% γ-HCH (ATSDR, 2005; Willett et al., 1998; Walker et al., 1999; Gong et al., 2004). The α-HCH/γ-HCH ratios were generally low in most samples from Tengeru, Akheri vegetable garden and all samples from MBOCU, indicating lindane as the main source of HCH contamination. High ratios may be related to

Table 3. Concentrations of pesticides and degradation products in soil at Akheri vegetable garden.

Compounds	Concentrations (mg/kg dw)						
	Burial point	Down slope, 5 m		Down slope, 20 m		Up slope, 5–20 m, n = 5	
Depth (cm)	50	10	50	10	50	10	30
4,4'-DDT	70	44	0.045	0.10	0.015	0.012–0.038	0.006–0.047
2,4'-DDT	54	40	0.014	0.025	0.0031	0.0025–0.012	0.0022–0.0102
4,4'-DDD	18	15	0.015	0.013	0.0014	0.001–0.0021	0.0007–0.0014
2,4'-DDD	7.2	6.3	0.0044	0.0044	0.0006	0.0003–0.0013	0.0004
4,4'-DDE	5.7	4.75	0.0052	0.0204	0.0016	0.0015–0.0112	0.0005–0.0023
2,4'-DDE	1.10	0.78	0.0005	0.002	0.0003	0.0001–0.0003	0.0001–0.0003
Total DDT	160	110	0.084	0.165	0.022	0.019–0.058	0.010–0.062
(DDE+DDD/DDT)	0.3	0.4	0.4	0.3	0.2	0.2–0.5	0.1–0.3
α-HCH	nd	0.924	0.0005	0.0022	0.00024	0.0001–0.0016	0.0004–0.0055
β-HCH	nd	nd	0.0007	0.0023	0.00024	0.00013–0.0012	0.00022–0.0034
γ-HCH	4.10	1.51	0.02	0.015	0.0016	0.0002–0.0011	0.0001–0.0015
δ-HCH	1.70	20	0.0012	0.0021	0.0001	0.0001–0.00013	0.0001–0.0006
ε-HCH	nd	nd	nd	nd	nd	nd	nd–0.0002
Total HCH	5.80	23	0.023	0.022	0.0026	0.0007–0.0025	0.0008–0.01
α-HCH/γ-HCH	0	0.6	0.03	0.1	0.2	0.2–10	0.5–11.3
Pentachlorobenzene	56.2	5.50	0.0083	0.08	0.0008	0.0005–0.0014	0.0002–0.0003
Hexachlorobenzene	2300	310	0.83	6.30	0.0333	0.021–0.12	0.0024–0.014
Pentachloroanisole	1.0	0.61	0.0023	0.0734	0.0011	0.0004–0.0022	0.0003–0.001
trans-Chlordane	nd	nd	0.0001	0.0003	nd	nd–0.0001	nd–0.0001
cis-Chlordane	1.5	nd	0.00011	0.0001	nd	nd–0.00003	nd–0.00004
oxy-Chlordane	nd	nd	nd	nd	nd	nd	nd
Heptachlor	nd	1.6	nd	0.0004	0.0001	nd–0.00012	nd–0.0002
cis-Heptachloroepoxide	0.36	0.35	0.00003	0.00002	nd	0.00001–0.00003	0.00001–0.00003
trans-Heptachloroepoxide	1.90	2.0	nd	nd	nd	nd–0.0002	nd–0.00014
Aldrin	6.20	0.42	0.0045	0.01	0.0003	0.0013–0.011	0.0004–0.001
Dieldrin	920	170	3.0	4.314	0.12	0.143–3.51	0.0048–0.096
Endrin	35	3.23	0.049	0.093	0.0013	0.0005–0.029	0.00002–0.0004
Endosulfan-I	4340	1100	3.0	2.50	0.042	0.0065–0.252	0.0002–0.01
Endosulfan-II	2820	800	2.50	3.10	0.049	0.022–0.25	0.0007–0.011
Methoxychlor	0.067	0.039	nd	nd	nd	nd	nd
Mirex	0.162	nd	0.0003	0.0005	nd	nd–0.00011	nd

transformation of γ-HCH into α-HCH. Both low and high α-HCH/γ-HCH ratios were found in samples from ACU maize farm and Mbimba, suggesting both lindane and technical HCH as the sources of HCH contamination. The high α-HCH/γ-HCH ratios found in this study are comparable to the findings from other studies in which the contamination was related to technical HCH. For example, α-HCH/γ-HCH ratios of up to 52 have been reported (Concha-Grana et al., 2006; Zhang et al., 2006).

Endosulfans

The highest concentrations of endosulfan-I and endosulfan-II ranged 0.31–4340 mg/kg dw and 0.21–2820 mg/kg dw, respectively and the total concentrations of endosulfans were up to 7200 mg/kg dw (Akheri vegetable garden). Endosulfan was mainly represented as endosulfan-I in most soil samples from Tengeru, MBOCU (10 cm) and some samples from Akheri vegetable garden (burial point and nearby points), which was similar to the technical formulation, suggesting lack of significant degradation or recent inputs. The concentrations of endosulfan-II were greater than those of endosulfan-I in all samples from ACU maize farm, Mbimba, most samples from Akheri vegetable garden, and some samples from MBOCU (50 cm, 100 cm and 10 m from burial point), which differs from the technical formulation and can be related to old input and the faster degradation of endosulfan-I than endosulfan-II, in soils. The half-lives for endosulfan-I and endosulfan-II are about 60 and 800 days, respectively. Another possible

Table 4. Concentrations of pesticides and degradation products in soil at TaCRI-Mbimba and MBOCU (mg/kg dw).

Sample site	TaCRI-Mbimba			MBOCU			
Sample point	Burial point		10 m from burial point	Burial point			10 m from burial point
Depth (cm)	50	100	50	10	50	100	10
4,4'-DDT	17	45	0.023	324	2.20	12	1.30
2,4'-DDT	7.10	24	0.01	76	0.41	2.30	0.30
4,4'-DDD	10.3	10	0.001	19	0.22	2.70	0.20
2,4'-DDD	4.40	5.30	0.001	6.10	0.1	0.1	0.1
4,4'-DDE	6.0	21	0.004	123	3.10	2.90	2.60
2,4'-DDE	0.80	4.0	0.001	6.20	0.10	0.10	0.03
Total DDT	45.6	110	0.04	554	6.10	20	4.50
(DDE+DDD)/DDT	0.9	0.6	0.2	0.4	1.3	0.4	1.9
α-HCH	0.90	9.0	0.001	nd	0.08	0.01	0.004
β-HCH	0.40	0.80	0.0001	nd	0.01	0.01	0.004
γ-HCH	0.60	1.0	0.00012	11	0.05	0.11	0.012
δ-HCH	0.50	0.70	0.0001	nd	0.01	0.01	0.01
ε-HCH	nd	nd	nd	nd	nd	nd	nd
Total HCH	2.40	12	0.0013	11	0.15	0.14	0.03
α-HCH/γ-HCH	1.5	8.9	8.3	0	1.6	0.1	0.3
Pentachlorobenzene	11	1.30	0.0001	6.0	0.05	0.10	0.01
Hexachlorobenzene	16	4.20	0.001	1003	0.50	0.50	0.10
Pentachloroanisole	1.0	0.70	0.00004	1.10	0.05	0.01	0.02
trans-Chlordane	nd	29	0.0004	nd	nd	nd	nd
cis-Chlordane	nd	25	0.001	nd	nd	nd	nd
oxy-Chlordane	nd	nd	nd	nd	nd	nd	nd
Heptachlor	nd	0.50	nd	nd	nd	nd	nd
cis-Heptachloroepoxide	nd	1.10	0.0001	nd	0.003	nd	nd
trans-Heptachloroepoxide	nd	nd	nd	nd	nd	nd	nd
Aldrin	1.40	2.0	0.0002	1.50	0.10	0.01	0.04
Dieldrin	18	480	0.02	602	9.50	3.0	7.10
Endrin	0.70	8.0	0.0003	18	0.03	0.02	0.01
Endosulfan-I	1.20	3.50	0.0002	2100	0.62	0.04	0.04
Endosulfan-II	1.30	6.0	0.0003	1400	1.0	0.30	0.20
Methoxychlor	nd	0.10	nd	nd	nd	nd	nd
Mirex	nd	nd	nd	nd	nd	nd	nd

reason is that endosulfan-I undergoes isomerization to endosulfan-II especially at high temperature or when exposed to light (ATSDR, 2000).

Aldrin, dieldrin and endrin

The highest concentrations of aldrin, dieldrin and endrin were as follows, respectively: Tengeru 2.2, 20.2 and 0.09 mg/kg dw; Akheri 6.2, 920 and 35 mg/kg dw; ACU maize farm 0.7, 39.3 and 0.83 mg/kg dw; Mbimba 2.0, 480 and 8 mg/kg dw, and MBOCU 1.5, 602 and 18 mg/kg dw. The concentrations of dieldrin were greater than those of aldrin and endrin in all samples from all the studied sites. This can partly be explained by the fact that, apart from being a pesticide, dieldrin is also formed as a metabolite of aldrin (FAO, 2000; ATSDR, 2002).

Hexachlorobenzene, pentachlorobenzene and pentachloroanisole

Hexachlorobenzene, pentachlorobenzene and pentachloroanisole were detected in 74 to 100% of the soil samples from the studied sites. The highest concentrations for these compounds varied from 2 to 2300, 0.028 to 56.2 and 0.014 to 1.1 mg/kg dw, respectively. The highest concentrations were generally found in Akheri samples and the lowest were found in Tengeru and ACU samples. Pentachloroanisole is the main degradation product of pentachlorophenol and pentachloronitrobenzene (UNECE, 2009); therefore, its

detection indicated contamination due to these compounds.

Chlordanes and heptachlors

Chlordane isomers, heptachlor and its degradation products were detected in soil samples mainly from Akheri and Mbimba. The highest concentrations of the compounds in Akheri samples were: *cis*-chlordane 1.5 mg/kg dw, *trans*-chlordane 0.0003 mg/kg dw, heptachlor 1.6 mg/kg dw, *cis*-heptachloroepoxide 0.36 mg/kg dw and *trans*-heptachloroepoxide 2 mg/kg dw. The highest concentrations of *trans*-chlordane, *cis*-chlordane, heptachlor and *cis*-heptachloroepoxide in samples from Mbimba were 29, 25, 0.5 and 1.1 mg/kg dw, respectively. The proportions of the pesticides and their degradation products suggested lack of effective degradation.

Methoxychlor and mirex

Methoxychlor and mirex were detected in some of the samples. Their highest concentrations varied from 0.0003 mg/kg dw (Tengeru) to 0.162 mg/kg dw (Akheri). Their concentrations were generally low suggesting that the contamination status of these compounds was attributed to environmental sources, that is, there were no significant sources at the studied sites.

Distribution of the pesticides and degradation products

There are strong positive correlations in the concentrations of the compounds at each studied site, indicating common sources. There were significant differences in the concentrations of the main pesticides and degradation products among the sampling depths in soil at Tengeru burial points, whereby the distribution for DDT, DDE and DDD was 75 cm > 30 cm > 150 cm ≈ 220 cm, while for HCH isomers, aldrin, dieldrin, pentachlorobenzene and hexachlorobenzene was 30 cm > 75 cm > 150 cm ≈ 220 cm ($p < 0.05$). The distribution pattern for DDT and DDE was slightly different from that observed in a study which compared the concentrations of DDT and DDE in soil samples collected at 30-40, 60-70 and 90-100 cm depths from an undisturbed contaminated site in Mato Grosso, Brazil, in which the highest concentrations at the most contaminated point were found at the 30 to 40 cm depth (Villa et al., 2006). This fact is explained because some years ago the affected area was filled up with foreign soil and turned over and then the original deposit was at different depth depending on the sampling point. The concentrations of pesticides and metabolites showed significant differences for some of the points and depths at Akheri (10 cm > 50 cm), ACU maize farm (10 cm > 30 cm ≈ 75 cm-except for DDTs and HCHs) and MBOCU (10 cm > 50 cm). These findings suggested that the vertical movement of the compounds was very slow maybe due to their low water solubility and their strong interactions with soil components (FAO, 2000). There were no significant differences in the concentrations of the main contaminants among the sampling depths at the following sites/points: Tengeru up and down slope (30 cm ≈ 75 cm ≈ 150 cm), ACU maize farm (10 cm ≈ 30 cm ≈ 75 cm-DDTs and HCHs) and Akheri (10 cm ≈ 30 cm) ($p > 0.05$) indicating even distribution of the compounds among the sampling depths. The concentrations of the pesticides and degradation products increased with depth in soil samples from Mbimba and MBOCU (100 cm > 50 cm) and some sampling points at Tengeru (150 cm > 75 cm and 30 cm), suggesting accumulation at lower depths despite that the differences were not quite significant.

The highest concentrations of the pesticides and degradation products in soil were found at the sampling points which were located within the burial areas and the concentrations showed a tendency of decreasing with increase in distance from those points. The concentrations in samples collected from points that were located down slope were significantly greater than those from up slope points, indicating that the distribution of the compounds was influenced by run-off, although this was only limited to short distances.

The distribution of the pesticides and degradation products in soil at the studied sites had been influenced by the management operations (e.g. turning over of soil, mixing with foreign soil and burying), agricultural operations (e.g. cultivation and irrigation activities) carried out especially at Tengeru, ACU maize farm and Akheri vegetable garden, as well as the environmental conditions (e.g. soil properties) and the physico-chemical properties of the compounds.

The results of the present study are comparable to the data from different locations, although the sampling depths are slightly different. The findings from other studies have generally shown that the concentrations of most of the organochlorine compounds decrease with depth, while fluctuations or lack of direct correlation between concentrations and depths are observed in disturbed soils such as cultivated soils. Wang et al. (2006) reported that the concentrations of most organochlorine compounds (HCHs and DDTs) decreased with depth, while fluctuations were observed in the plow layers of some cultivated soils due to some cultivation activities and batch irrigation. The sampling depths were 0-20, 20-25, 25-30, 30-40, 40-50, 50-60, 60-80 and 80-100 cm. Another study reported that at several points the highest concentration of HCH isomers was found in the superficial samples, whereas the more contaminated sample was at 10 cm depth and in others between 36 and 48 cm, despite that no significant difference was found among the sampling depths (0-20 to 85–95 cm)

(Concha-Grana et al., 2006).

Conclusions

High concentrations of organochlorine pesticides and major degradation products were detected in samples from all the studied sites. The results indicated aged contaminants but which had not undergone significant degradation. The concentrations of the compounds generally decreased with increase in depth and distance from the burial points. Therefore, the mobility of these compounds in soil was generally low certainly due to their low water solubility and their strong interactions with soil components, mainly clay and organic matter. However, there were some variations in the distribution of the compounds among the sampling depths as some showed even distribution or accumulation downwards indicating some risks for groundwater contamination. It is recommended that remediation should be carried out at the burial points.

ACKNOWLEDGEMENTS

The German Academic Exchange Service (DAAD) and the University of Dar es Salaam are gratefully acknowledged for funding this work. The late Prof. Michael A. Kishimba is greatly acknowledged for guidance during this study. Sincere thanks to Prof. Dr. Dr. Karl-Werner Schramm, Mr. Bernhard Henkelmann, and the German Research Center for Environmental Health, Munich-Germany for facilitation of the laboratory analyses. Thanks are also due to Mr. Norbert Fischer and Mrs. Silke Bernhoft (Germany) for their technical contributions during the laboratory analyses.

REFERENCES

Åkerblom M (1995). Guidelines for environmental monitoring of pesticide residues in the SADC region, SADC/ ELMS monitoring techniques series, Sweden.

ATSDR (2002). Toxicological profile for aldrin and dieldrin, Atlanta, GA: U.S. Department of Health and Human Services, Public Health Service.

ATSDR (2005). Toxicological profile for hexachlorocyclohexane, Atlanta, GA: U.S. Department of Health and Human Services, Public Health Service.

ATSDR (Agency for Toxic Substances and Disease Registry) (2000). Toxicological profile for endosulfan, Atlanta, GA: U.S. Department of Health and Human Services, Public Health Service.

Buyuksonmez F, Rynk R, Hess TF, Bechinski E (1999). Occurrence, Degradation and Fate of Pesticides During Composting, Compost Sci. Util. 7:4.

Concha-Grana E, Turnes-Carou MI, Muniategui-Lorenzo S, Lopez Mahia P, Prada-Rodriguez D, Fernandez-Fernandez E (2006). Evaluation of HCH isomers and metabolites in soils, leachates, river water and sediments of a highly contaminated area. Chemosphere 64:588-595.

Extension Toxicological Network (EXTOXNET) (1993). Movement of Pesticides in the Environment, Oregon State University.

FAO (1998). Prevention and disposal of obsolete and unwanted pesticides stocks in Africa and the Near East, FAO, Rome.

FAO (2000). Assessing soil contamination – A reference manual, FAO, Rome.

FAO (2001). Baseline study on the problem of obsolete pesticide stocks, FAO, Rome.

FAO (Food and Agriculture Organization of the United Nations) (1996). Disposal of bulk quantities of obsolete pesticides in developing countries, FAO, Rome.

Gałuszka A, Migaszewski ZM, Manecki P (2011). Pesticide burial grounds in Poland: a review. Environ Int. 37(7):1265-1272.

Gong ZM, Xu FL, Dawson R, Cao J, Liu WX, Li BG, Shen WR, Zhang WJ, Qin BP, Sun R, Tao S (2004). Residues of hexachlorocyclohexane isomers and their distribution characteristics in soils in the Tianjin area, China, Arch. Environ. Contam. Toxicol. 46: 432-437.

Harrison SA (1990). The Fate of Pesticides in the Environment, Agrichemical Fact Sheet No.8, Pesticide Education Program, Penn State Cooperative Extension.

Mansour M (1993). Fate and Prediction of Environmental Chemicals in Soils, Plants and Aquatic Systems, Boca Raton: Lewis Publishers, P.291.

Manvelyan E, Simonyan L, Anakhasyan E (2006). Hazards of burial site in Armenia, A report, J. Pestici. Action Netw. UK.

McEwen FL, Stephenson GR (1979). The Use and Significance of Pesticides in the Environment, A Wiley Interscience Publication, New York, P.538.

Pesticide Action Network (PAN UK) (2005). Buried pesticide threat in Belarus, Press release. P.29.

Qiu X, Zhu T, Yao B, Hu J, Hu S (2005). Contribution of dicofol to the current DDT pollution in China, Environ. Sci. Technol. 39:4385-4390.

Rauf VG, Bashkin, VN, Galiulina, RA (2002). Behaviour of persistent organic pollutants in the air-plant-soil system: a review. Water Air Soil Pollut. 137:179-191.

Ritter L, Solomon KR, Forget J, Stemeroff M, C.O'Leary D (1995). Persistent Organic Pollutants (POPs): An Assessment Report. United Nations Environ. Program.

Schramm KW, Henkelmann B, Kotalik J (2008). Standard operating procedures for determination of pesticides, PAHs and PCBs, Accreditated Testing Laboratory According to DIN EN ISO/IEC 17025, Helmholtz Research Centre for Environmental Health. Inst. Ecol. Chem. Munich-Germany.

Sun WJ, Cai JM, Huang B (1997). Study on the residues of pesticides in tea garden soil and tea. J. Fujian Agric. Univ. 26:9-43.

UNECE (United Nations Economic Commission for Europe) (2009) Task Force on Persistent Organic Pollutants (POPs), Summary of expert reviews of pentachlorophenol (PCP), UNECE.

Villa RD, Dores EFG, Carbo L, Cunha MLF (2006). Dissipation of DDT in a heavily contaminated soil in Mato Grosso, Brazil, Chemosphere 64:549-554.

Waldron AC (1992). Pesticides and Ground Water Contamination, Bulletin 820, Ohio Cooperative Extension Service, The Ohio State University.

Walker K, Vallero DA, Lewis RG (1999). Factors influencing the distribution of lindane and other hexachlorocyclohexanes in the environment. Environ. Sci. Technol. 33:4373-4378.

Wang X, Piao X, Chen J, Hu J, Xu F, Tao S (2006). Organochlorine pesticides in soil profiles from Tianjin, China. Chemosphere 64:1514-1520.

Willett KL, Ulrich EM, Hites RA (1998). Differential toxicity and environmental fates of hexachlorocyclohexane isomers: Critical review. Environ. Sci. Technol. 32:2197-2207.

Zhang HB, Luo YM, Zhao QG, Wong MH, Zhang GL (2006). Residues of organochlorine pesticides in Hong Kong soils, Chemosphere 63:633-641.