JASEM ISSN 1119-8362
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Full-text Available Online at
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J. Appl. Sci. Environ. Manage. September, 2010
Vol. 14 (3) 37 - 40

Residues of Organochlorinated Pesticides in Soil from Tomato Fields, Ngarenanyuki, Tanzania

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ABSTRACT: This work presents the concentrations of five pesticide residues, lindane, chlorpyrifos, endosulfan, *p, p'*-DDE and *p, p'*-DDD in soil samples collected from tomato fields in Ngarenanyuki, Tanzania. Endosulfan sulphate was detected in 100 % of the sample analysed with mean concentration of 0.2407 mg/kg dw. Chlorpyrifos was detected in 87 % of the samples with mean concentration of 0.1253 mg/kg dw. *p, p'*-DDE and *p, p'*-DDD were detected in 46 and 40 % of the samples analysed with mean concentrations of 0.1482 and 0.154 mg/kg dw, respectively. Lindane was the least detected pesticide. It was detected in 5 (33 %) of soil samples analysed with mean concentration of 0.2126 mg/kg dw. Low concentrations detected indicate the past usage of the pesticides. @ JASEM

The increased use of pesticides in agricultural activities has caused pollution of environmental compartments; soil, water and air. Their chemical properties such as high lipophilicity, bioaccumulation potential, long half-life in the environment, and potential to long-range transport, envisaged the chance to be found in water, soil or food even decades after their applications (Abbassy et al., 1999). Agricultural pesticides most often are applied as liquids, granules or seed sprayed/treated on the crop and/or the soil (Pimentel and Levitan, 1986). Extremely small percentage (less than 0.3%) of the amount applied, goes into direct contact with or consumed by target pests, therefore 99.7% goes somewhere else in the environment (Pimentel, 1995). In Tanzania, most of the pesticides imported are used in agriculture activities (AGENDA, 2006). The aim of this work was to ascertain the levels of pesticide residues in soils from tomato fields at Ngarenanyuki.

MATERIALS AND METHODS

Study Area and Sample Collection: Ngarenanyuki is among the 17 wards in Arumeru District, Arusha City, Tanzania. It comprises of five villages (Uwiro, Ngabobo, Olkung'wado, Kisimiri chini and Kisimiri juu) located at the foot of Mt. Meru, north east of Arusha City (3° 9' 0" South, 36° 51' 0" East). 15 soil samples of 500 g each were randomly collected from 15 different locations at Uwiro village in January and February, 2009 (Figure 1). The soil samples were collected by using stainless spoon at the depth of 0 - 15 cm (the root zone). The sampling procedures were conducted as described by Åkerblom (1995).
Chemicals: Chromatography grade dichloromethane, n-hexane, acetone, cyclohexane and ethyl acetate were used for sample preparation. Samples were quantified using pesticide standard mixture which had over 99 % certified purity. Laboratory glassware were washed with detergents, rinsed with distilled water and acetone, and then dried in an oven at 110 °C overnight prior to use.

Extraction: Two sub-samples of 20 g each were subjected under different experimental treatments. For dry weight determination one sub-sample in a pre-weighed watch glass was heated in the oven at 105 ^{o}C to constant weight. Another sub-sample was put in a teflon stoppered flask (250 ml) for pesticides extraction. 14 ml of 0.2 M ammonium chloride solution was added, the mixture swirled for 2 min and left to stand for 15 min. 100 ml of cyclohexane/acetone mixture (1:1 v/v) was then added, the flask was tightly stoppered and vigorously shaken for 1 min, and then less vigorously after every 10 min for 1 h. The contents were then left to stand overnight. Intermittent shaking was continued the next day for two hours and then left to settle. Distilled water was cautiously added to the neck of the flask. The organic phase was pipetted into an E-flask containing anhydrous sodium sulphate (15 g), swirled and left to stand for 15 min. The contents were then decanted through a plug of glass wool into an evaporation flask. The remaining sodium sulphate was rinsed with 20 ml of acetone/cylohexane mixture and decanted through the same glass wool. The resulting extract was concentrated to less than 2 ml using a rotary evaporator, the solvent was then changed to cyclohexane/ethylacetate (1:1 v/v, 1 ml) ready for clean up.

Clean up: This was done by glass column (60 cm x 22 mm) chromatography packed with florisil (magnesium silicate), glasswool and anhydrous sodium sulphate. Cyclohexane: acetone (9:1 V/V) was added into the glass column and allowed to pass through drop by drop until very little was left on the upper part of the column. The sample concentrate was poured into the column and drained to make 3 ml of the sample. This was then transferred into the Teflon cork vial and stored at 4 °C until analysis.

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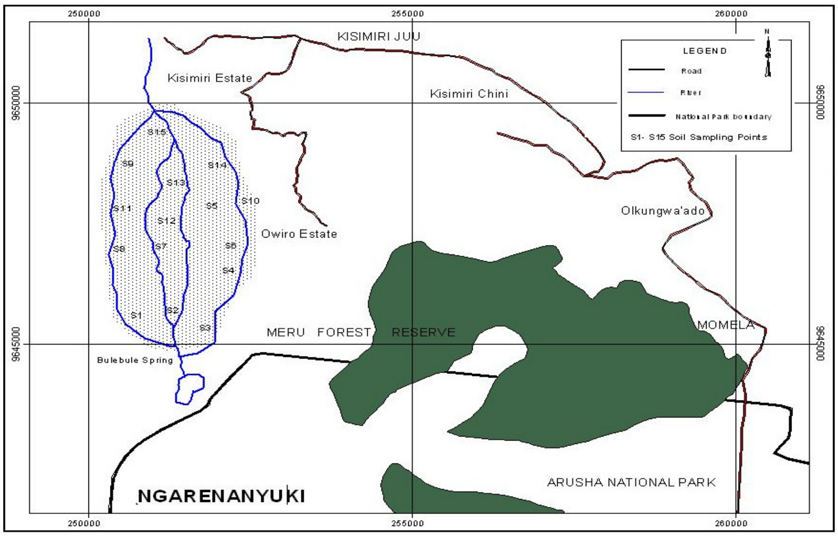

Fig 1.Map of Ngarenanyuki showing sampling sites at Uwiro Estates

Analytical Quality Assurance: A 100 ml aliquot of each n-hexane, dichloromethane, cyclohexane, ethyl acetate and acetone was concentrated to 2 ml and analysed to check the contamination from the solvents used. Two matrix blanks soil were obtained from a virgin land at Ardhi University. The results showed no significant peaks in the chromatograms of the blanks. Recoveries were estimated by spiking the matrix blank with standard pesticide solutions at concentrations ranging from 0.01 to 1.1 µg/ml for each analyte. The average of percentage recoveries ± SD (n= 4) were as follows: Lindane 95.7 ± 0.7 %, Chlorpyrifos 81.2 ± 3.6 %, *p', p-* DDE 94.5 ± 1 %, and *p', p*-DDD 97.7 ± 0.7 %. The results were not corrected for recoveries since they were within the normal acceptable range 70-120 % (Hill, 2000).

Sample Analysis: Analysis of the pesticide residues was done as described by Åkerblom (1995). Gas chromatography Varian CP- 3800 equipped with ^{63}Ni Electron Capture (EC) detector, and capillary column WCOT FUSED SILICA 30 m x 0.32 mm, coated with CP-SIL 8CB DF 1.0 µm was used. Nitrogen was used as both a carrier and make up gas at a flow rate of 30 ± 1 ml/min. The temperature programme was held at 70 °C for 1 min, 15 °C/min to 180 °C, 4 °C/min to 230 °C for 15 min. The injector and detector temperatures were 240 °C and 250 °C, respectively. Identification of residues was effected by running samples and external reference standards in GC and then comparing the chromatograms.

RESULTS AND DISCUSSION

Table 1 shows concentrations of lindane, *p, p'*-DDE, *p, p'*-DDD, chlorpyrifos and endosulfan sulphate detected in the soil samples. The concentrations ranged between 0.036 – 1.25 mg/kg dw. The highest value was found in sample S9 and lowest in sample S4. Endosulfan sulphate, was the most abundant residue detected in all 15 (100 %) samples analysed, with the concentration range 0.075 - 1.25 mg/kg dw and mean of 0.24 mg/kg dw. Although endosulfan sulphate is susceptible to photolysis in the environment, it is expected to have a high occurrence in cultivated areas (Manirakiza *et al*., 2003). Dem *et al*., (2007) also reported high occurrence of 11 (74 %) endosulfan sulphate out of 15 soil samples from Sikasso areas in Mali. Endosulfan sulphate is a major degradation product of endosulfan, and is as toxic as parent compound (Jiang *et al*., 2009). Because of high toxicity, technical endosulfan was restricted in many countries including Tanzania. The high level of endosulfan sulphate detected implies that the pesticide has recently been used. This was also revealed by 83 % of farmers interviewed during field visits that declared to have been using endosulfan under the trade name thionex.

Chlorpyrifos, was detected in 13 soil samples (87 %), with mean concentration of 0.125 mg/kg dw and maximum of 0.172 mg/kg dw. The high detection frequencies of chlorpyrifos can be explained by the fact that it is moderately persistent in soils. The half-life of chlorpyrifos in soil is between 60 and 120 days, but can range from 2 weeks to over 1 year, depending on the soil type, climate, and other conditions (Wauchope *et al.,* 1992).

DDT metabolites *p, p'*-DDD and *p, p'*-DDE were detected in 53 % of the soil samples analysed with Σ of DDT (*p, p'*-DDD + *p, p* –DDE) concentration of 2.27 mg/kg dw. *p, p'*-DDD is sometimes used as a pesticide of its own under the trade name rothane and ISO approved name of TDE (Tomlin, 2000). *p, p'*-DDD was detected in 47 % of samples analysed with concentration ranging from bdl to 0.211 mg/kg dw and mean concentration of 0.15 mg/kg dw. DDE is normally found in the environment as a result of contamination or breakdown of DDT. *p, p'*-DDE was detected in 6 out of 15 samples analysed (40 %) with mean concentration of 0.15 mg/kg dw, and 0.214 mg/kg dw and bdl being the highest and lowest concentrations, respectively. The occurrence of *p, p'*-DDD and *p, p'*-DDE in soil samples correlates with findings from farmers interviewed who claimed to have used DDT till late 1990s. DDD and DDE may last in the soil for a very long time, potentially for hundred of years (ATSDR, 2002) sticking strongly to soils and thus remain in the surface layers of soil. The persistence in the environment of these organochlorines have also been reported in other areas, for instance in China, DDE and DDD have been reported to be widely distributed in soil despite of their discontinued use since 1983 (Wu *et al.*, 2007). Dem et *al.*, (2007) also reported presence of *p, p'*-DDE in soils and water from four cotton growing areas of Mali. In Tanganyika Sugar Cane plantations (TPC), Kilimanjaro, Tanzania, residues of organochlorine pesticides, aldrin, dieldrin, heptachlor epoxide, HCHs, endosulfans and DDTs were detected with mean concentrations ranging from 1.1 to 636.7 µg/l despite of discontinued use (Hellar and Kishimba, 2005).

Table 1 Type and levels of pesticide residue detected in soil

Sampling		Type and Concentrations (mg/kg dw)				
Site	Position	Lindane	*p,p'*-DDE	*p,p'*-DDD	Chlorpyrifos	Endosulfan sulphate
S1	S 03°09.441 E 036° 51.819	0.296	bdl	bdl	0.123	0.115
S2	S 03°09.487 E 036° 51.854	0.22	bdl	bdl	0.096	0.09
S3	S 03°09.760 E 036° 51.902	bdl	bdl	bdl	0.143	0.12
S4	S 03°09.321 E 036° 51.469	0.22	0.036	bdl	0.09	0.9
S5	S 03°09.812 E 036° 51.902	bdl	0.154	0.127	0.129	0.121
S6	S 03°09.811 E 036° 51.633	bdl	0.155	0.13	0.115	0.122
S7	S 03°09.230 E 036° 51.522	bdl	0.162	0.127	0.117	0.123
S8	S 03°09.319 E 036° 51.105	bdl	bdl	bdl	0.136	0.117
S9	S 03°09.934 E 036° 51.522	bdl	0.168	0.132	0.12	1.25
S10	S 03°09.672 E 036° 51.196	bdl	bdl	bdl	bdl	0.075
S11	S 03°09.766 E 036° 51.765	bdl	bdl	0.211	0.153	0.128
S12	S 03°09.188 E 036° 51.833	0.215	bdl	bdl	0.116	0.094
S13	S 03°09.677 E 036° 51.117	bdl	bdl	0.196	0.172	0.125
S14	S 03°09.915 E 036° 51.976	bdl	bdl	bdl	bdl	0.114
S15	S 03°09.433 E 036° 51.745	0.112	0.214	0.155	0.119	0.117
Average	na	0.2126	0.1482	0.154	0.1253	0.2407
% detected	na	33	40	46.67	86.67	100
Σ DDT	na	na	2.27		na	na

bdl=below detection limit, na=not applicable

Soil concentrations of lindane depend on the method of application, soil composition and climatic conditions (Atlas and Giam, 1981). Although it was banned for use in several countries, lindane is still used extensively in many countries for the control of agricultural pests. Lindane like other organochlorine pesticide is highly persistent in most soils, with a field half-life of approximately 15 months (Atlas and Giam, 1981). In this study lindane was detected in 5 (33 %) out of 15 samples analysed with the highest concentration of 0.29 mg/kg dw and mean concentration of 0.21 mg/kg dw. It was the least detected pesticide followed after *p, p'*-DDE. The least detection of lindane can be explained in one hand to be not extensively used pesticides in Ngarenanyuki. It was not mentioned in farmer's interview as one of the pesticides used; therefore detection of lindane indicates the past usage.

Generally, the concentrations detected in this study compares with values reported in other countries in the world. Although the concentrations of pesticide residues reported under this investigation were low, soils pollution can play an important role in the global fate and distribution of pollutants. Soil can act as a sink of pesticides, from which they can be released into water or air, and may be taken up by plant roots, and bioaccumulate in the food chain to an extent higher enough to cause chronic adverse health effects to consumers.

Acknowledgement: *Acknowledgement*: We are gratefully acknowledging the inputs of Mr. Lutufyo Mwamtobe from the Department of Chemistry, University of Dar es Salaam, and Mr. Addo Ndimbo and Ramadhani Mbulume from the School of Environmental Science and Technology, Ardhi University for their invaluable technical assistance during the laboratory extraction and analysis.

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