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**Marete M. G, Lalah J. O, Mputhia J. & Wekesa V. W**

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# Contamination from Organochlorine Pesticides (OCPs) and other Pesticides in Agricultural Soils of Buuri, Imenti South and Imenti North Sub counties, Meru County Agroecosystem in Kenya

**[*1]Marete M. G, [2]Lalah J. O, [3]Mputhia J. & [4]Wekesa V. W**

**[1]PhD Candidate, the Technical University of Kenya**

**[2]Department of Geoscience and the Environment, the Technical University of Kenya**

**[3]Department of Chemical Science and Technology, the Technical University of Kenya**

**[4]Department of Technical and Applied Biology, the Technical University of Kenya**

***E-mail of the Corresponding Author: gabriel_muthee@yahoo.com**

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# Abstract

The presence and distribution of organochlorine (OCPs) and the residues of other pesticides in Buuri, Imenti South and Imenti North Sub Counties, Meru County agroecosystem in Kenya was examined in the current research. Pesticide residue analysis was done with gas chromatograph mass spectroscopy. Twenty soil samples sampled randomly from each sub county were collected for the multi residue and Quecher's method analysis of OCP sand other pesticides such as organophospahates, carbamates, fungicides, pyrethroids and neonicotinoids respectively. Seventeen OCP residues with notable concentrations were observed from the study area. Alpha-

HCH, beta-HCH, gamma-HCH, delta-HCH, heptachlor, aldrin, heptachlor epoxide, endosulfan I, dieldrin, p,p'-DDT, endosulfan II, p,p'-DDD, Endrin aldehyde, p,p'-DDE, endosulfan sulfate, heptachlor epoxide and methoxychlor were obtained at concentrations ranging from BDL-85.83±1.98, with methoxychlor giving the highest mean concentration. Endrin was not detected. Other pesticides detected included carbendazin (conc range) metalaxyl (range conc) and chlorpyrifos (range of conc). All organochlorine pesticides detected from the three sub counties in Meru County are in the list of priority pollutants of US Environmental Protection Agency (USEPA). Results indicate that persistent organic pollutants (POPs) and other pesticides exist in agricultural soils in Meru and recent illegal use in sampled area for some of the OCPs should and be monitored and regulated. Increased soil monitoring is recommended to detect persistency and changes in target environment. Some of these OCPs and other pesticides had been reported to be in use by farmers in the three sub-counties, although presence of other OCPs which have been banned could be due to environmental cycling and persistence.

**Key Words***: Organochlorine, Pesticides, Contamination, Agricultural soils, Agroecosystem*

### 1.1 Background of the Study

The increase in world population has led to hunger in various parts of the world [1, 2]. This has led to an increase in efforts towards the achievement of the second Goal of the Sustainable Development Goals (SDGs) which is aimed at zero hunger [3]. To achieve this vital goal, various approaches for pest control [4], irrigation, and resistant crop varieties [5] are being advanced. The use of Pesticide in agriculture has mostly contributed to increased crop yield and this has led to economic stability in many Countries [6].

The widespread application of organochlorine pesticides in agriculture during the 19$^{th}$ century raised a serious concern due to their harmful effects on human and environment [7]. Although this resulted in the ban of most of these compounds, their residues are still detectable in

environment due to persistence and bio-accumulative effects. In some cases, illegal application and emissions from obsolete stocks can introduce their residues in environment [8, 9]. Once released into the air pesticides are subjected to different degradation and transport pathways. The rates of degradation and dissipation vary greatly depending on the type of pesticide and the prevailing environmental condition such as temperature, wind, humidity, soil type and biotic factors [30]. Consequently, the concentration, spatial and temporal trends in levels vary from one compound to the other. Organochlorine pesticides have been detected in water, soil and sediment samples around the world including Kenya [8, 9].

Agriculture remains the backbone of the Kenyan economy. It is the most important sector in the economy, contributing approximately 25% of the GDP, and employing 75% of the national labor force [10]. The objectives of agricultural sector strategy have been increasing agricultural growth [11]. It is estimated that more than 80% of Kenyan farmers use pesticides in their operations [12]. Irresponsible use of pesticides may lead to the prevalence of pesticide residues in the environment and agriculture [13]. It is confirmed that pesticide residues exceeding a threshold pose a possible risk for consumers [14]. The toxic effects include acute neurological toxicity, disturbances in the immune and reproductive system, and several other diseases. Of the major classes of pesticides on the market, organochlorines have received a lot of attention from the scientific community due to their low cost, versatility against various pests, persistency, bio-accumulative nature, and potential toxicity to humans and to wildlife [15]. Because of their ability to bio-accumulate and often harm unintended species, organochlorine pesticides (OCPs) have been banned or restricted use in Kenya, with a switch to the less persistent organophosphorus and synthetic pyrethroids being favored in Kenya and other countries [16, 31].

Vegetables constitute a major part of the human diet by contributing to the dietary requirements of nutrients [17]. Lettuce, cabbage, and onions are some of the most consumed vegetables in Kenya. Many farmers in both rural and urban communities in Kenya have taken up vegetable

production on a commercial basis. Meru County which is located in the Mount Kenya region produces horticultural and other agricultural crops for local consumption and export. Many consumers purchase their vegetables directly from the market, whereas few obtain it from the farms [17]. To improve yield, farmers are using a large amounts of various OCPs, organophosphates, carbamates and pyrethroids during the entire period of vegetable cultivation, even at the fruiting stage. Residues of pesticides accumulate in the vegetables, the soil, and in nearby water bodies (from runoffs) [8, 9, 17-20]. The problem of residue accumulation needs more attention in vegetables because they are consumed either raw or without much processing [18]. Pesticide residues in vegetables constitute a possible risk to consumers, and a number of reports have documented this [19, 20]. Pesticides including some OCPs, Ops, carbamates, pyrethroids and neonicotinoids are used in this region. The sources and fate of pesticide residues in agricultural soils therefore need to be investigated.

The soil serves as a reservoir for organic pollutant and plays an important role in their global distribution [21]. Soil not only has a large retention capacity but also re-emits OCPs into the environment as a secondary source [22]. Soil contamination with pesticide residues can also contribute to failure in horticultural produce to meet the Maximum Residue Limits (MRLs) as well as contamination of aquatic ecosystems [4, 9, 12, 15, 22, 26]. A significant proportion of hexachlorocyclohexanes (HCHs) and dichlorodiphenyltrichloroethanes (DDTs), ranging from 20 to 70% of its degradation products, may remain in the soil for a long period of time after having been applied [23]. DDT and the rest of persistent OCPs have been found in soil samples and distributed throughout the environment [23]. The transfer of OCPs from the soil surface layers to lower depths may impact their volatilization rates from surface soil and thus impose potential risks for shallow groundwater [24]. Other pesticides such as OPs and carbamates residues have been found to contaminate agricultural soils in farming systems [29].

Despite global attempts to eliminate OCPs, there is still evidence of their presence in various matrices from both abiotic and biotic components since a considerable amount of pesticides applied ultimately end up in water bodies. In this study, the levels of organochlorine and other pesticide residues in soil were assessed from three sub Counties of Meru County in Kenya, where horticultural and other agricultural productions have been intensified.

## 2.0 Materials and Methods

### 2.1 Study area

The study was conducted in Meru County. Meru County is found in the eastern region of Kenya, approximately 225 kilometers northeast of Nairobi. Meru County has a total area of 6,936 $km^2$ [25] and lies within latitude 0.0515° N and longitude 37.6456° E with an altitude of 5300 feet above the sea level. It has a population of 1,601,629 people [25] and is among the fastest developing towns in Kenya [26]. The growth is associated with rising vegetable and flower farming business in the areas surrounding water resources. Tourism and its related activities in the area together with relocations from rural to urban areas because of decreasing farming incomes from the conventional cash crops have also been contributing factors towards this growth [26]. Meru County has total of nine sub counties namely; Igembe north, Igembe central, Igembe south, Tigania east, Tigania west, Buuri, Imenti central, Imenti south and Imenti north.

### 2.2 Sampling area

Sampling was done in 3 sub counties namely Buuri, Imenti north and ImentiSouth (Figure 1), twenty farms were selected from each sub county for soil sampling as shown in Figure 1.

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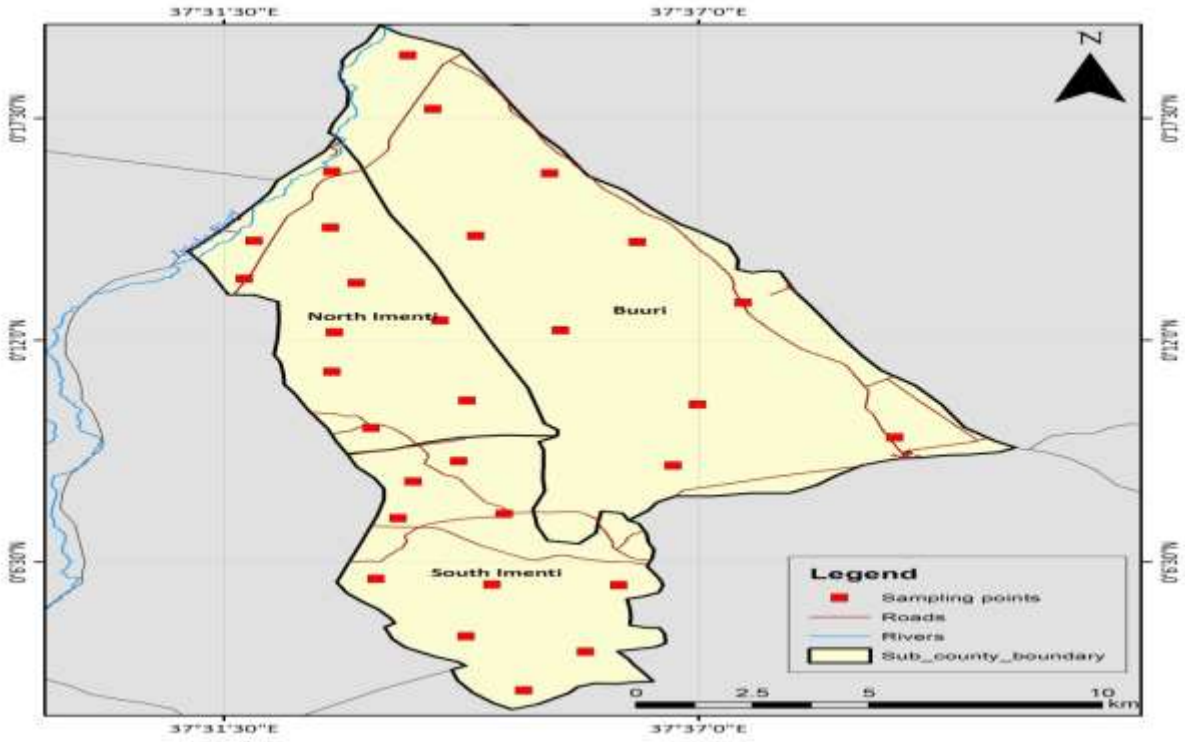


**Figure 1: Map of Meru, Kenya Showing the Sampling Sites**

**2.3 Materials and Chemicals**

Pesticide standard mixture of over 99 % purity was used for identification and quantification of pesticide residues in the samples. Pesticide standards were obtained from Dr. Ehrenstorfer GmbH Company (Germany). Hexane, acetone, dichloromethane and isooctane solvents were obtained from Fisher Scientific (USA). Analytical grade anhydrous sodium sulphate, sodium chloride, aluminum oxide, sodium hydroxide, hydrochloric acid, copper and methanol were also obtained from Fisher Scientific (USA). General purpose solvents were triple distilled before use.

### 2.4 Samples collection

Soil samples were collected from thirty farms, three from each sub county. Sampling sites were randomly selected within each farm. Soil cores were dug using a pre-cleaned hoe and scooped using a stainless steel shovel from a depth of 15- 25cm from five different locations within each farm and market and approximately 200 g of the core scooped. The cores were combined and 500 g of the soil was then placed on clean aluminium foils, wrapped and put inside a black polythene bag labelled' packed in self-sealing bags, put inside cooler boxes and transported to the laboratory. They were then preserved at -20 ºC in the refrigerator prior to extraction [27]

### 2.5 Samples Extraction and Clean-Up

Multiresidue [8, 9] Soxhlet extraction (EPA method 3540) and QuecheRs mehods [12, 18], respectively, were used in soil extraction [28] for OCPs and other pesticide residues. Before extraction, for the OCPs, the soil samples were taken from the freezer and left to thaw for 6 hours. 20 g of anhydrous sodium sulphate was used to dry 20 g of the soil sample; this was done by grinding and mixing thoroughly in a mortar. The mortar containing the dried soil sample was then covered with an aluminium foil and left to stand for about 12 hours. The process was done in triplicates for each of the samples. Extraction was then carried out for sixteen hours in the Soxhlet using a mixture of hexane together with acetone (200 ml) in the ratio of 3:1, respectively. After the sixteen hours, the Soxhlet extractor was turned off and the extracts allowed tocool. This was followed by an addition of 2 ml of isooctane, which acted like a keeper and the extracts concentrated using a rotary evaporator to about 3 ml. The concentrated extracts were thereafter transferred into vials using pasteur pipettes and stored in a refrigerator at 4 ºC pending clean-up.

Cleaning up of the soil samples was done using a chromatographic column filled with 2 g of activated anhydrous $Na_2SO_4$ followed by 15 g of deactivated Aluminium oxide and lastly by 2 g

activated anhydrous sodium sulphate. The column was conditioned with 15 ml of n-hexane and the sample mixture poured into it then the vial rinsed three times with 1 ml hexane. The analytes were then eluted using 175 ml of n-hexane. 2 ml of iso-octane was then added to the cleaned extract which was then concentrated to around 3 ml under vacuum evaporator. The same procedure was applied to all the samples. The last extract was reduced to 0.6 ml using a mild nitrogen stream. At this point the samples were ready for GC analysis.

**2.6 Removal of Sulphur from soil samples**

Approximately 1 g of copper powder that had just been activated was added to the already cleaned soil extracts in order to remove sulphur. All extracts containing Sulphur formed copper sulphide as indicated by the black colouration. A glass funnel filled with glass wool together with 2 g of activated anhydrous $Na_2SO_4$ was used to filter the soil extracts. The anhydrous sodium sulphate was conditioned using 5 ml of HPLC hexane and the samples introduced then 20 ml of HPLC hexane used to elute the analytes into a round bottomed flask. This was followed by an addition of 2 ml Iso-octane before it was concentrated. The reduced extracts were transferred into clean auto vials and further reduced to 0.5 ml under a mild stream of nitrogen ready for GC analysis.

**2.7 Quantification and Analysis of the Samples**

Soil extracts were analysed for selected pesticides using gas chromatography–mass spectrometry (GC–MS) on a 6890N GC instrument (Agilent, USA) equipped with a thermo scientific trace GOLD GC column (TG 5SILMS 30m X 0.25mm X 0.25 µm) coupled to an Agilent 5973 MS (USA). The mass spectrometer (MS) was operated in EI + mode in the resolution of >5000. Injection was splitless with volume of 1µL to 280$^{o}$C, with helium as carrier gas at 1 ml min-1. The injection temperature program applied was as follows: 90˚C (3 min), 90 ºC to 200 ˚C (at 30

˚C/ min and hold time of 15 min), 200 ˚C to 275 ˚C (at 30 ˚C/min and hold time of 5 min). Chemstation software was used in data processing.

**2.8 Identification and quantification**

Organochlorine and other pesticides (obtained from IoIc, Poland), reference standards ranging from 0.01 mg/L to 0.981 mg/L were individually prepared for each standard and quantification was based on calibration curve calculations. Each standard gave a calibration curve with a straight line and the line of best fit drawn from the plot of the response factor (peak area) against standard concentration.

All analyte lines gave a correlation factor ($R^2$) above 0.99 showing high correlation between analyte concentration and instrument response ratio. Standard concentrations were obtained by interpolation from the graphs which applies the equation

$Y= mX +c$

Where Y = Normalised peak area (instrument response)

X = Standard concentration

m = Gradient, and

c = Constant

Concentrations of the sample analyzed were also obtained in the same way and working backwards, concentrations in form of µg/kg dry weight of soil were generated for the pesticide residues.

**2.9 Statistical Data Analysis**

The data obtained was analyzed using Statistical Package for Social Science (SPSS) to establish relationship between pesticide residue levels in the samples from different sampling sites and the

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seasonal variability. Bivariate correlation coefficients were established using Pearson product moment correlation coefficient, "r", which is a dimensionless index, whose value is in the range -1.0≤r≤1.0.

### 3.0 Results and Discussion

The average percentage recoveries of 17 pesticides ranged from 73.29±6.18% of endosulfan sulfate to 102.24±10.04% for dieldrin and the rest of the pesticides had values which have been summarized in Table 1. The pesticide residue levels detected in samples were not corrected since all recovery values (Table 1) were within the acceptable range of 70-120 % [27].

**Table 1: Percent recoveries of Pesticides and Limits of Detection (LOD)**

| Pesticide | Recovery(%±S.D) | LOD (ng/L) | Pesticide | Recovery(%±S.D) | LOD (ng/L) |
|---|---|---|---|---|---|
| **α HCH** | 94.82±1.31 | 1.1 | **Endosulfan sulfate** | 78.25±1.56 | 2.1 |
| **β HCH** | 87.52±4.09 | 1.6 | **Aldrin** | 94.26±5.23 | 3.6 |
| **γ HCH** | 92.06±4.58 | 1.6 | **Dieldrin** | 74.83±3.33 | 3.1 |
| **δ- HCH** | 82.54±6.95 | - | **Endrin** | 80.01±4.21 | 2.2 |
| ***p,p'- DDT*** | 79.89±3.41 | 1.7 | **Endrin aldehyde** | 77.81±3.63 | 2.2 |
| ***p,p'- DDE*** | 78.35± 5.12 | 1.8 | **Heptachlor** | 92.08±4.56 | 1.1 |
| ***p,p'- DDD*** | 79.31±2.84 | 1.6 | **Heptachlor epoxide** | 88.35±2.45 | 1.1 |
| **α- endosulfan** | 72.58±4.95 | 1.1 | **Methoxychlor** | 88.23±6.86 | 1.6 |
| **β- endosulfan** | 93.23±7.13 | 1.5 | | | |

Organochlorine pesticide residues detected in soil from Imenti North Sub County ranged between BDL to79.755±8.45µg/Kg of dry weight. Methoxychlor was the highest detected in soil from sampling site Six. The mean concentration of exachlorocyclohexanes (HCH) isomers ranged between BDL- 60.602±8.18µg/Kg for α-HCH, β-HCH (BDL-50.0911±7.22 µg/Kg), γ-HCH (BDL-40.713±1.86 µg/Kg), δ- HCH (BDL - 0.5801±0.07 µg/Kg).

The Mean concentration of heptachlor ranged from BDL-45.618±4.11 µg/Kg, aldrin (BDL-49.7081±5.26 µg/Kg), heptachlor epoxide (BDL-0.705±0.01 µg/Kg), α-endosulfan (BDL -

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3.085±0.98 µg/Kg), β-endosulfan (BDL-48.046±5.68 µg/Kg), endrin aldehyde (BDL - 24.787±1.95 µg/Kg), endosulphan sulfate (BDL-46.699±1.90 µg/Kg),Dieldrin (BDL-24.788±1.95µg/Kg) and methoxychlor (BDL - 79.755±1.45 µg/Kg), while endrin, was not detected in all the sites.

The mean concentration of *p,p'*-DDT ranged between BDL- 64.8069±3.51 µg/Kg, while the mean concentration ofits*p,p'*-DDEranged between BDL-76.78±6.18 µg/Kg and *p,p'*-DDD was not detected.

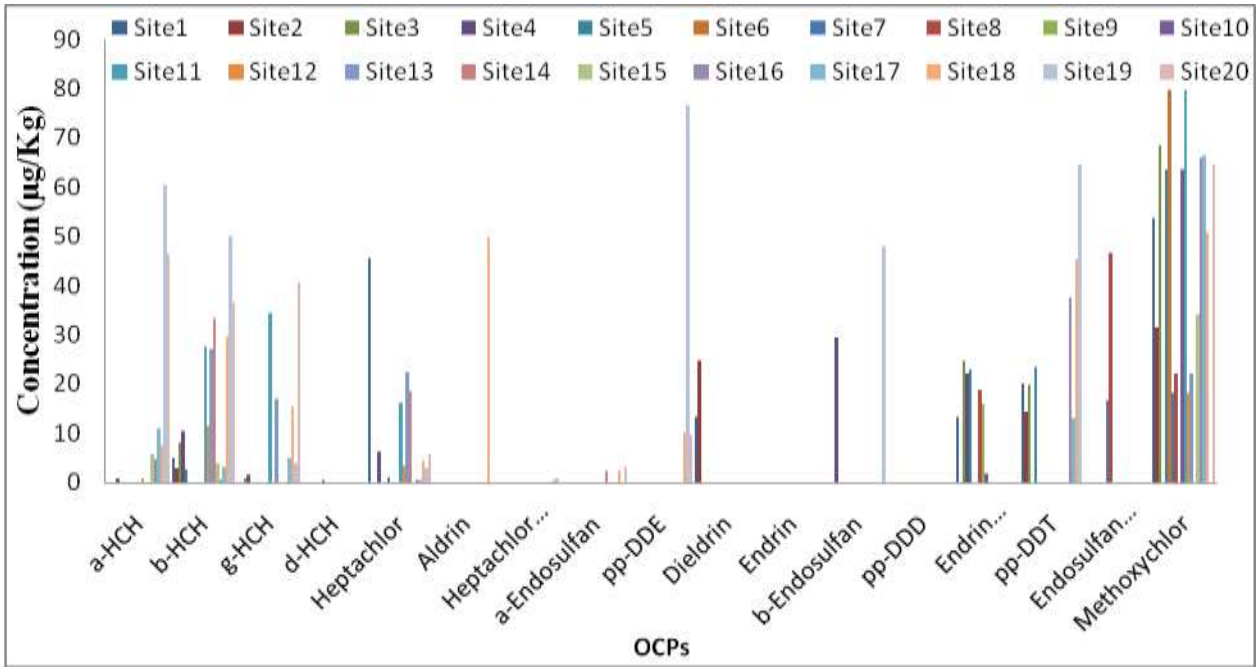


**Figure 2: Organochlorine Pesticides in Soil from Imenti North Sub County**

Organochlorine pesticide residues detected in soil from Imenti South Sub County ranged between BDL to85.825±1.98 µg/Kg. Methoxychlor was the highest detected in soil from sampling site Twelve. The mean concentration of hexachlorocyclohexanes (HCH) isomers

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ranged between BDL- 2.352±0.36 µg/Kg for α-HCH, β-HCH (BDL-14.873±2.17 µg/Kg), γ-HCH (BDL-2.768±0.12 µg/Kg), δ- HCH (BDL - 0.513±0.04 µg/Kg).

The mean concentration of heptachlor ranged from BDL-3.181±0.04 µg/Kg, β -endosulfan (BDL - 12.017±0.68 µg/Kg), endrin aldehyde (BDL - 63.015±6.12 µg/Kg), endosulphan sulfate (BDL-64.638±9.69 µg/Kg), while heptachlor epoxide, a-endosulfan, pp-DDE, dieldrin, endrin were not detected in all the sites.

The mean concentration of *p,p'*-DDT ranged between BDL- 43.627±2.13 µg/Kg, while the mean concentration ofits*p,p'*-DDDranged between BDL-60.514±8.77 µg/Kg and *p,p'*-DDE was not detected.

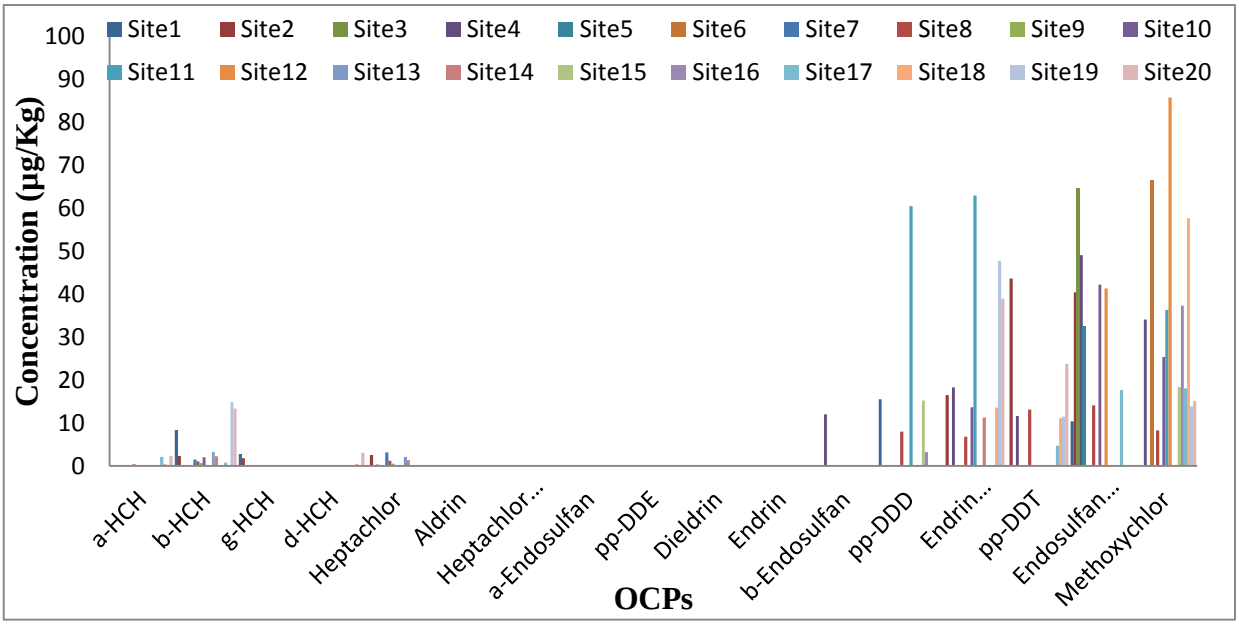


**Figure 3: Organochlorine Pesticides in Soil from Imenti South Sub County**

Organochlorine pesticide residues detected in soil from Buuri Sub County ranged between BDL to72.95±5.06 µg/Kg. Methoxychlor was the highest detected in soil from sampling site Thirteen. The mean concentration of hexachlorocyclohexanes (HCH) isomers ranged between BDL-29.239±4.25 µg/Kg for α-HCH, β-HCH (BDL-3.481±0.54 µg/Kg), γ-HCH (BDL-1.009±0.00 µg/Kg), δ- HCH (BDL - 0.064±0.00 µg/Kg).

The Mean concentration of heptachlor ranged from BDL-0.526±0.056 µg/Kg, β -endosulfan (BDL - 12.017±0.68 µg/Kg), andrin (BDL - 0.841±0.00 µg/Kg), heptachlor epoxide (BDL - 0.805±0.00 µg/Kg), a-endosulfan (BDL-0.803±0.00µg/Kg), pp-DDE (BDL - 0.7336±0.00µg/Kg), dieldrin (BDL-0.803±0.00µg/Kg), endrin aldehyde (BDL - 63.015±6.12 µg/Kg), endosulphan sulfate (31.992±2.04µg/Kg), while endosulfan, endrin were not detected in all the sites.

The mean concentration of *p,p'*-DDT was below detection limit while the mean concentration ofits*p,p'*-DDDranged between BDL-5.408±0.05µg/Kg and *p,p'*-DDEranged between BDL-0.7336±0.00 µg/Kg.

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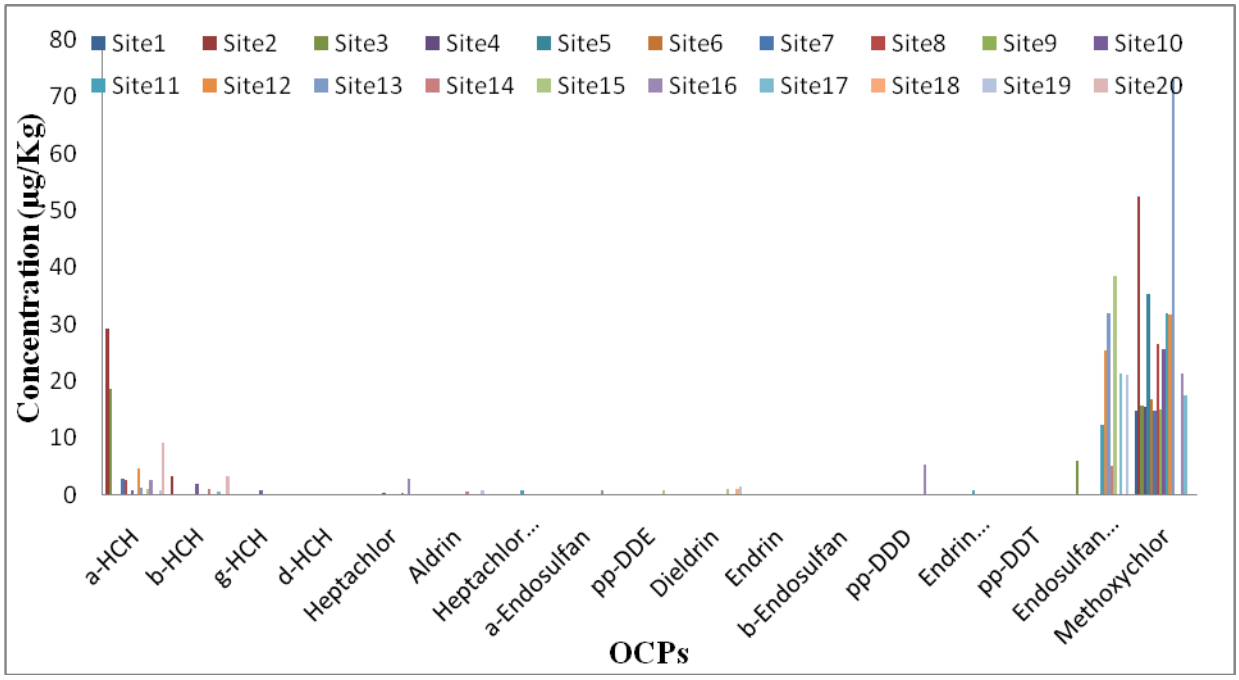


**Figure 4: Organochlorine Pesticides in Soil from Buuri Sub County**

## 4.0 Conclusion

The present study indicates that the banned or restricted organochlorine pesticide compounds are still detected in agricultural soils in Meru County due to their cycling and persistence nature and illegal usage. This poses immediate danger to human and animals due to the pesticides bioaccumulations in food chain. Therefore the Policy makers should put in place a regular environmental monitoring program and mitigation strategies of reducing the pollutants, Awareness campaigns should be conducted to educate the general public on the safe pesticide use and adverse environmental and human health impacts of these pesticides and there should be a follow up on the compounds banned or restricted to ensure that they are not illegally used, and that regulation of OPs, carbamtes, pyrethroids and neonicotinoids should be enhanced to minimize residue levels in agricultural soils.

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