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Fate of Aldrin and Dieldrin in Nanumba-North Municipality, Ghana

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Abstract

The aim of this study was to assess the levels of contamination of aldrin and dieldrin in the Nanumba-North Municipality of the Northern Region of Ghana. Gas Chromatography equipped with electron capture detector (GC-ECD) was used to analyse the samples. In all, 148 samples were analysed, consisting ten (10) soil samples, 108 water samples and 30 food crop items. The study revealed the presence of aldrin and dieldrin at varying mean concentrations, with dieldrin recording significant concentrations in the samples. Aldrin was the least frequently detected residue in the soil samples, and occupied 30% with a mean value of 0.003 ± 0.001 µg/L. On the other hand, dieldrin occupied 70% with a mean value of 0.007 ± 0.004 µg/L. The trends of aldrin and dieldrin residues distribution in water samples from the various depths indicated frequent occurrence and higher residue concentrations in water samples at depths 30 cm and below than depths 15-30 cm and 0-15 cm. The presence of aldrin and dieldrin in the water samples could be traced to wind drift or atmospheric transport of volatilized pesticides, direct overspray, direct spillage, pesticide misuse by farmers, run-off and leaching from application fields and surrounding areas during and after their applications. The concentrations of aldrin and dieldrin residues in the samples analysed were generally low and below the WHO (2017) MRLs. Therefore, food crops analysed in the study area will not pose any significant threat to the food industry as far as shipment to other parts of the world is concerned. Besides, aldrin and dieldrin detected showed significant differences at $p < 0.05$. However, just because there are low levels does not mean that consumers will not be exposed to its health risks. Since continuous availability can lead to bioaccumulation and biomagnification through the food chain. The study, therefore, recommends that in order to reduce environmental contamination and to protect both wildlife and human health appropriate authorities in the area should instil means to enlighten pesticides users on the best way of sustainable pest control methods as well as implementing integrated pest management strategies that can magnify the ambition of worthwhile environmental conditions.

Keywords: *Aldrin; dieldrin; metabolites; Nanumba-North Municipality; GC-ECD*

1.0 Introduction

Aldrin and dieldrin are members of the organochlorine group of insecticides, effective at protecting plants and animals from insect pests, and sharing similar toxicity because of their similar structure and properties. They were widely used in the late 1940s; however, due to their toxicity and accumulation in the food chain, they were gradually banned in developed and developing countries in the late 1970s and 1980s. Cyclodiene pesticides such as aldrin and dieldrin account for more than 60% of reported cases of resistance. Therefore, various countries have launched the monitoring of organochlorine pesticides, Pesticide contamination is more severe in poorer countries, and it has a pernicious effect on ecology and wildlife. The break-down (degradation) of POPs includes physical, chemical and biological factors. Physical, chemical, and biological factors in the natural environment influence the degradation of POPs, of which biological factors play the critical role. Microorganisms are among the most important biological factors, including bacteria, fungi, and algae. Physical and chemical degradation produce by-products that cannot be completely degraded and sometimes are even more toxic than the parent compounds. In the natural environment, aldrin is usually converted to dieldrin by biotic or abiotic mechanisms, and the half-life of dieldrin is significantly longer. Each time an aldrin is introduced into the environment

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as a result of application, disposal, or spillage, several things may happen to the aldrin and can be influenced by several processes and factors. The ultimate fate of the aldrin is determined by processes such as its mobility, persistence, non-target toxicity, and volume of use. The fate processes can have both positive and negative influences on an aldrin's effectiveness or its impact on the environment. They can move an aldrin to the target area or destroy its potentially harmful residues. Sometimes they can be detrimental, leading to reduced control of a target pest, injury of non-target plants and animals, and environmental damage. One of the major concerns today is the mobility of aldrin into groundwater and their bio-accumulation throughout the food chain. However, different soil characteristics (pH, clay, sand, organic matter, etc.), aldrin characteristics (water solubility, the tendency to adsorb to the soil, persistence, its resistance to being broken down over time, etc.), climatic factors, application methods and different handling practices of aldrin, for instance, can promote or prevent each process. The physical and chemical properties as well as natural processes influence their environmental risk. Apprehending the fate of aldrin ensures that applications are not only effective but are also environmentally friendly. The degree of environmental risks depends on four factors such as adsorption, degradation, mobility/volatility, and absorption. Furthermore, evidence of fate of some pesticides in other farming areas in Ghana has been documented by [5] [6] and it is enough backing to give cause for similar concerns in the Nanumba-North Municipality. This study aimed to assess the levels of contamination of aldrin and dieldrin in the area.

2.0 MATERIALS AND METHOD

2.1 Study area

The study was carry-out in the Nanumba North Municipality, which is located in the southeastern part of Northern Region of Ghana. The area lies between latitudes 8.5º N and 9.25º N and longitudes 0.57º E and 0.5ºE (Figure 1). According to the 2010 Population and Housing Census, the total population of Nanumba North Municipality is 141,584. Males constitute 49.4 percent and females represent 50.6 percent in the Municipality. The Municipality has a total land area of 2260.8 sq. Km. The predominant occupation is farming (www.ghanadistricts.com 2011).

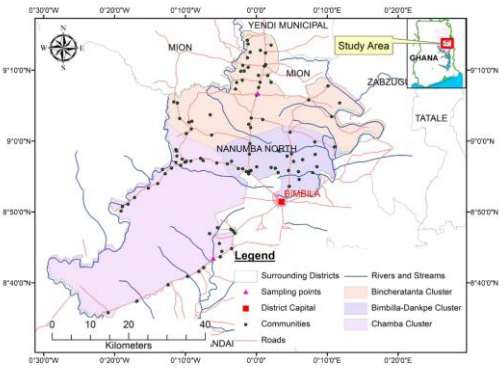

Figure 1: A map showing Nanumba North Municipality in the Northern Region of Ghana

2.2 Collection of samples

Water samples were collected from a River and two Dams. The Kumbo River was used while the Dams were Bincheratanga, and Waanpu. Grab sampling technique was used to collect thirty-six (36) water samples from four zones at three different depths. The depths used were (that is, 0-15cm, 15-30cm, and 30cm and below) using 500 mL pre-cleaned Teflon sample bottles with caps for analysis making a total of one hundred and eight (108) water samples. The Teflon sampling bottles were rinsed well with water to be sampled several times before they were carefully filled to over-flowing, to avoid trapping air bubbles in sealed bottles. Additionally, the Teflon sampling bottles were rinsed with the river and two dams' water before taking the water samples. The samples were labelled and transported to the laboratory within 24–48 hours on ice in clean ice chests and stored in the refrigerator at 4 °C until they were analysed for Aldrin and its metabolite. Also, all the selected farms were grouped into three Clusters that is, Bimbilla-Dankpe Cluster (BDS), Bincheratanga Cluster (BS) and Chamba Cluster (CS). Three quadrants of 70 × 70 m were marked out in each Cluster. In each quadrant, three (3) soil samples were collected randomly at depths 0–20 cm with a soil auger. The rectitude of taking this depth was because nutrient uptake by plants is usually reported to be within this horizon[1]. Additionally, one soil sample was taken from a nearby natural forest to act as a control (X1). This gave a total of ten soil samples for the study area. All soil samples were kept in well-labelled plastic polythene containers and transported to the laboratory for analysis. The soil samples were oven-dried at 105 °C to constant weight and sieved using 2 mm nylon mesh. Sampling of yams, maize, and ayoyo were done at the three different Clusters in the study area in August 2023, and labelled as the Bimbilla-Dankpe Cluster, Bincheratanta Cluster, and Chamba Cluster. In each Cluster three (3) samples of the selected crops were taken randomly. Additionally, one sample of each crop was taken from farmlands where pesticide application was observed not to be common to act as a control. These gave a total of twenty (30) crop samples for the study area. The samples were packed in black polyethylene bags and labelled accordingly and transported to the laboratory. In the laboratory, the samples were ground into powder. They were then packed in freezer bags and stored in a refrigerator at 4 ^{0}C for analysis.

2.3 Extraction of samples

After filtration of water samples through 0.45 mL fiberglass filters (WHATMAN) to remove debris and suspended material, 1000 mL portions of the filtered water samples were transferred into 2 L capacity separating flasks. A 30 mL of saturated sodium chloride solution (NaCl) was added to each to produce a salt-out effect to adjust the pH to 7. The samples were then thoroughly mixed by inverting the flask three to four times. A 100 mL of dichloromethane as extraction solvent was then added to each sample and vigorously shaken manually for 2–3 min while releasing the pressure intermittently. The phases were allowed to separate for 5 min and the dichloromethane extracts (organic layers) were separated from the aqueous layers. The extraction for each

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water sample was repeated twice with 100 mL of dichloromethane and the organic layers were put together and dried over anhydrous sodium sulfate through filter papers into 50 mL round bottom flasks. The extracts from the water samples were then concentrated on rotary vacuum evaporators (Buchi Ratovapor R-210, USA) to about 1 mL and subjected to silica clean-up. Ten grams (10 g) of the representative soil samples were weighed and quantitatively transferred into 250 mL separating flasks. 10 mL of acetonitrile was added to each of the soil samples in the flasks and ultra-sonicated (Becon FS400b) for 5 min. An additional 10 mL of acetonitrile was added, and the flasks closed tightly. The samples were placed on a horizontal mechanical shaker (Ika-Werke HS 501 Digital) and set to shake continuously for 30 min at 300 mot/ min. The contents were then allowed to stand for 10 min to sufficiently separate the phases or layers. 10 mL of the supernatants were carefully taken by pipette and dried over 2 g anhydrous magnesium sulfate through filter paper into 50 mL round bottom flasks. The concentrates were then adjusted to about 2 mL using the rotary film evaporator (Buchi Ratovapor R-210, USA) at 35 °C, and made ready for the silica clean-up step. All reagents and chemicals were of analytical grade and were used as received. Extraction of aldrin and its metabolites in yam, maize, and ayoyo samples were done. Each sample of 5.0 g was placed into a flask and 30 mL of acetone: methanol (1:1 v/v) extraction solvent was added. The content of the flask was shaken continuously on a mechanical flash shaker at 200 rpm for 3 h. The extract was filtered through a Buchner funnel fitted with Whatman filter paper under suction. The filtrate was transferred into a 500 mL separating funnel and 150 mL sodium sulfate solution was added. The mixture was partitioned with 30 mL of dichloromethane and vigorously shaken for 2 min releasing pressure intermittently. The phases were allowed to separate and the lower dichloromethane phase was collected into a flask. The aqueous layer was partitioned twice using 10 mL portions of dichloromethane each time. The dichloromethane extracts were combined and dried on 20 g of anhydrous sodium sulfate in a mini-glass column. The dried extract was concentrated to approximately 2 mL in a rotary evaporator at 37 ^{0}C and stored in a 2 mL sample vial. This was then taken for clean-up.

2.4 Clean-up of samples

Extracts clean-up was done, using polypropylene cartridge columns, packed with one-gram silica gel previously activated for 10 h in an oven at 130 °C, which has a 2 g layer of anhydrous sodium sulfate on top and conditioned with 6 mL dichloromethane. The concentrated extracts were then loaded onto the cartridges, and 100 mL round bottom flasks were placed under the columns to collect the eluates. A 20 mL dichloromethane was then used to elute the columns/cartridges afterward, and the total filtrates (eluents) collected were concentrated just to dryness using the rotary evaporator (Buchi Ratovapor R-210) set at 40 °C. The residues were re-dissolved in 1 mL ethyl acetate by pipetting and transferred into 2 mL standard opening vials before quantitation by gas chromatography (GC) (Varian Association Inc. USA) equipped with electron capture (ECD). Extracts clean up were done, using polypropylene cartridge columns, packed with one-gram silica gel previously activated for 10 h in an oven at 130 °C, which has a 1 cm thick layer of anhydrous magnesium sulfate on top and conditioned with 6 mL acetonitrile. The concentrated extracts were then loaded onto the columns/ cartridges, and 50 mL pear shape flasks were placed under the columns to collect the eluates. A 10 mL acetonitrile was used to elute the columns/cartridges afterward. The total filtrates (eluents) collected were concentrated to dryness using the rotary evaporator (Buchi Ratovapor R-210) set at 40 °C. The residues were re-dissolved in 1 mL ethyl acetate by pipetting and transferred into 2 mL standard opening vials before quantitation by gas chromatography (GC) (Varian Association Inc. USA) equipped with electron capture detector (ECD). All extracts were kept frozen until quantification was achieved. For the clean-up, a 15 g mixture of alumina and activated charcoal (12:1) slurry was packed with dichloromethane in a mini glass column and topped up with a 2 cm layer of anhydrous sodium sulphate. The column was conditioned with 5 mL of dichloromethane and the sample extract was loaded on the column. The sample vial was rinsed two times with 2 mL aliquots of dichloromethane and the rinsed was added to the column. The sample was eluted with 30 mL dichloromethane and elutes concentrated to approximately 2 mL using a rotary evaporator at 37 ^{0}C. The final extracts were refrigerated at 4 ^{0}C until GC analysis. The limit of quantification for aldrin and dieldrin detected in the samples in this study was 0.001 µg/L and 0.001 mg/kg dry weight respectively.

2.5 Data analysis

Statistical package for social sciences (SPSS) was used to generate the means, standard deviation, and standard error for aldrin and its metabolite. One-way analysis of variance (ANOVA) was performed to analyse significant differences in the concentrations of aldrin and its main metabolite detected from the samples.

3.0 Results and Discussion

3.1 Aldrin and its Main Metabolite in samples from Nanumba-North Municipality

An important finding of the study is the simultaneous presence of aldrin and its main metabolite dieldrin in the samples analysed. Aldrin and its metabolite dieldrin were present in the samples analysed at varying mean concentrations. In agricultural soils, the degradation of aldrin can be divided into three pathways: the oxidation pathway, the reduction pathway, and the hydroxylation pathway. In the oxidation degradation pathway of aldrin, dieldrin was the main metabolite and has a stable structure. The presence of chlorine-substituents on these molecules has been implicated in their persistence however, dieldrin was more stable than aldrin since the half-life of aldrin is 20-100 days, and that of dieldrin is 1.5-2.5 years. This suggested that aldrin gradually breaks down into dieldrin which was more stable. Therefore, the mean concentration of dieldrin was higher than the parent compound in the samples analysed. Perhaps the possible mechanisms for the low levels of aldrin from the agricultural soils were due to erosion, volatilisation, uptake by plants and

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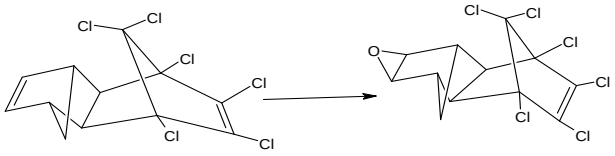
animals, and biodegradation. Since the representative half-life of aldrin was 20-100 days, then the estimated half-life for the transformation of aldrin to dieldrin in the agricultural soils was 12-60 days. The molar concentration of aldrin and dieldrin in the soil samples analysed were 7.876 picomoles, and 15.75 picomoles, respectively. Aldrin was the least frequently detected residue in the soil samples analysed, and occupied 30% with a mean value of 0.003 ± 0.001 µg/L. On the other hand, dieldrin occupied 70% with a mean value of 0.007 ± 0.004 µg/L. The mean concentrations of aldrin and dieldrin recorded in this study were below the MRL of 1.000 µg/L, and 0.030 µg/L for agricultural soils, (WHO 2017). The mean value of aldrin measured in this study was lower than the mean values of 0.01 µg/L and 0.01 µg/L reported by [3,4], in cocoa beans from Ghana. Similarly, the mean value of dieldrin measured in this study was lower than the mean values of 0.01 µg/L and 0.02 µg/L reported by [3 and 4], respectively in cocoa beans from Ghana. Perhaps, the key reason could be due to the differences in the sampling methods used as well as weather conditions. In general, the mean concentration of dieldrin at Chamba Cluster was higher than the mean values measured at Bimbilla-Dankpe and Bincheratanga Clusters.



Aldrin (7.88 picomoles) Dieldrin (15.75 picomoles)

Figure 2: Degradation of aldrin to dieldrin in the soil samples

Table 1: Aldrin and Dieldrin (µg/L) in soil samples in the Nanumba-North Municipality of the Northern Region of Ghana.

CLUSTERS		Aldrin and its metabolites (µg/L)	
		Aldrin	Dieldrin
0-20 cm	BDCS1	0.003	0.006
	BDCS2	0.004	0.007
	BDCS3	0.005	0.005
	BCS1	0.003	0.006
	BCS2	0.003	0.008
	BCS3	0.003	0.007
	CCS1	0.002	0.008
	CCS2	0.003	0.005
	CCS3	0.004	0.011
	x̄±SD	0.003±0.001	0.007±0.002
	XI	ND	ND
MRLs (WHO, 2017)		1.000	1.000

BDCS1**:** soil sample from Bimbilla-Dankpe cluster one; BDCS2**:** soil sample from Bimbilla-Dankpe cluster two; BDCS3**:** soil sample from Bimbilla-Dankpe cluster three; BCS1**:** soil sample from Bincheratanga cluster one; BCS2**:** soil sample from Bincheratanga cluster two; BCS3**:** soil sample from Bincheratanga cluster three; CCS1**:** soil sample from Chamba cluster one; CCS2**:** soil sample from Chamba cluster two; CCS3**:** soil sample from Chamba cluster three; Control X1; ND**:** Not detected.

Table 2.0: (a, b and c) Levels of aldrin and Dieldrin (µg/L) in water bodies in the Nanumba-North Municipality of the Northern Region of Ghana (ND: Not-detected)

Table 2.0a: Levels of aldrin and Dieldrin in Waanpu Dam

pesticide residues	Waanpu Dam				
	Depths(µg/L)				
	0-15cm	15-30cm	30cm and below	Mean±SD	WHO MRLs(µg/L)
Aldrin	ND	ND	0.004-0.007	0.002±0.001	1.000
Dieldrin	ND	ND	0.006-0.008	0.002±0.001	1.000

Table 2.0b: Levels of aldrin and Dieldrin in Bincheratanga Dam

pesticide residues	Bincheratanga Dam				
	Depths(µg/L)				
	0-15cm	15-30cm	30cm and below	Mean±SD	WHO MRLs(µg/L)
Aldrin	ND	ND	0.002-0.003	0.001±0.000	1.000
Dieldrin	ND	ND	0.002-0.004	0.001±0.000	1.000

*Corresponding Author: **Fusheini Yakubu**

Table 2.0c: Levels of aldrin and Dieldrin in Kumbo River

pesticide residues	Kumbo River				
	Depths(µg/L)				
	0-15cm	15-30cm	30cm and below	Mean±SD	WHO MRLs(µg/L)
Aldrin	ND	ND	0.002-0.003	0.001±0.000	1.000
Dieldrin	ND	ND	0.002-0.004	0.001±0.000	1.000

Table 3: Levels of aldrin and Dieldrin in food crop samples

pesticide residues	Food crops items				
	Selected food crops (µg/L)				
	Yam	Maize	Ayoyo	Mean±SD	WHO MRLs(µg/L)
Aldrin	ND	ND	ND-0.002	0.001±0.000	1.000
Dieldrin	ND-0.003	ND-0.002	ND-0.003	0.003±0.002	1.000

Table 4: Estimated period of decay of Aldrin and Dieldrin in soil samples in the study area

Aldrin and dieldrin in soil samples in the study area					
Name of pesticide	**Concentration of pesticide**	**Name of residue**	**Concentration of residue**	**Half-life of pesticide**	**Estimated period of decay**
Aldrin	0.003	Dieldrin	0.007	20-100 days	10-60 days

4.0 CONCLUSIONS AND RECOMMENDATIONS

4.1 CONCLUSION

From this study, the trends of aldrin and dieldrin residues distribution in water samples from the various depths indicated frequent occurrence and higher residue concentrations in water samples at depths 30 cm and below than depths 15-30 cm and 0-15 cm. Aldrin and dieldrin residues measured in water samples were below the WHO MRLs for drinking water. Additionally, the concentrations of aldrin and dieldrin residues in the soil samples analysed were generally low and below the WHO MRLs for agricultural soils. Similarly, aldrin and dieldrin residues measured in the food crops form the study sites were below the WHO MRL set for food crops. The presence of aldrin and dieldrin in the samples could be traced to wind drift or atmospheric transport of volatilized pesticides, direct overspray, direct spillage, pesticide misuse by farmers, run-off and leaching from application fields and surrounding areas during and after their applications. Aldrin and dieldrin detected showed significant differences at $p < 0.05$. However, the mean concentration of dieldrin at Chamba Clusters was higher than the mean values measured at Bincheratanga and Bimbilla-Dankpe. Therefore, at the moment aldrin and dieldrin may not pose significant risks to consumers.

4.2 Recommendations

In order to ensure sustainable and desirable environmental conditions in the Nanumba-North Municipality, the environmental protection agency must establish effective and protective measures such as integrated pest management practices and organic farming.

Extension officers should ensure routine monitoring of aldrin and dieldrin in the municipality for the prevention, control, and reduction of environmental pollution, so as to minimize health risks to the people.

DATA AVAILABILITY STATEMENT

The data that support the findings is Statistical package for social sciences (SPSS) and One-way analysis of variance (ANOVA).

CONFLICT OF INTEREST STATEMENT

Conflict of Interest: The authors declare that they have no conflict of interest.

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