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Unparalleled perils of Dichlorodiphenyltrichloroethane (DDT) and its metabolites in Nanumba-North Municipality, Ghana

By

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Abstract

The aim of this study was to assess the levels of contamination of DDT and its metabolites 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. On the whole, 148 samples were analysed, consisting of ten (10) soil samples, 108 water samples and 30 food crop items. The study revealed the presence of p,p'-DDT and its metabolites at varying mean concentrations, with p,p'-DDD recording significant concentrations in soil samples. The mean concentrations of p,p'-DDT and its metabolites in the samples analysed from the study site were generally below their respective MRLs (WHO, 2017) of 1.000 µg/L. p,p'-DDT and its metabolite (p,p'-DDE, and p,p'-DDD) were not found in the water and food crop items but are still present in 90% of the soil samples. Additionally, p,p'-DDT and its metabolites 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 appropriate authorities in the municipality such as the EPA should provide proper tutelage to farmers on the best way of pesticide utilization that can zip up the ambition of sustainable agricultural production and desirable environmental conditions.

Keywords: *p,p'-DDT; Nanumba-North Municipality; GC-ECD; pests and diseases*

1.0 Introduction

Organochlorine compounds are highly hydrophobic, soluble in water, and have a strong affinity for organic matter and carbon. They are commonly found in soils and groundwater from pesticide formulation and storage sites (F.Yakubu et al., 2023).

The subject that has preoccupied the minds of scientists in the last few decades is *the unparallel perils of DDT and its metabolites in the environment.* In order to, do justice to the above subject and to remain faithful on the chosen subject, there is a sense to take a historical journey on the factors that animated chemists to synthesis this potential toxic chemical. DDT, or dichloro-diphenyl-trichloroethane, has a significant history and impact on the environment. Originally developed as an insecticide in the 1940s, DDT became widely used for agricultural and public health purposes. DDT is a broad spectrum with a non-systemic contact insecticide of activity because of its ability to control vectors that transmit malaria, sleeping sickness, yellow fever, typhus, and other insect-transmitted diseases. It was banned in several countries in the early 1970s because of ecological considerations. DDT was designated as a persistent organic pollutant in 1997 by the Governing Council of the United Nations Environment Programme (UNEP, 1997). DDT and its metabolites are persistent in the environment and resistant to complete degradation by microorganisms, although photochemical degradation does occur. The persistence of DDT is substantially lower in tropical climates than in temperate ones (IPCS, 1989). However, its environmental consequences soon became apparent. DDT has been linked to several adverse effects, including the decline of bird populations and the thinning of eggshells, which led to reproductive issues. It also bioaccumulates and persists in the environment, making it a long-term concern. The harmful effects of DDT on human health have also been documented, with studies showing links to cancer, reproductive disorders, and developmental issues. Due to these concerns, international and national efforts have been made to regulate and restrict the usage of DDT. However, DDT and other persistent organochlorines have

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been detected in water bodies and soils such as Lake Bosomtwi, Volta Lake, Tano and Vea Dams and Irrigation Sites in the Upper East Region of Ghana (Darko *et al.*, 2008, Ntow 2007, Pelig-Ba 2011, Abagale 2020), and it is enough backing to give cause for similar concerns in the Nanumba North Municipality. Studying the environmental impact of DDT is crucial due to the unparalleled perils it poses to the environment. DDT is a highly toxic pesticide that has been widely used in the past. However, research has shown that DDT has devastating effects on wildlife and ecosystems. The persistent nature of DDT means that it can stay in the environment for a long time, accumulating in organisms and causing harm to entire food chains. Additionally, DDT has been linked to the decline of certain bird species, such as the bald eagle, due to its negative impact on their reproductive systems. Understanding the environmental consequences of DDT is vital for developing effective strategies to protect ecosystems and promote sustainable pest control methods. Exposure to DDT and its metabolites poses significant health risks to both humans and wildlife. The effects of DDT and its metabolites on human health include increased risks of cancer, reproductive problems, and neurological disorders. Furthermore, long-term exposure to DDT and its metabolites can have severe consequences, with studies suggesting adverse effects on immune function and hormone regulation.

Similarly, while extensive research has been done on the effects of DDT and its metabolites in other parts of the world, less is known in the Nanumba North Municipality of the Northern Region of Ghana. In order to fix this expertise gap, there is the need, to assess the levels of contamination of DDT and its metabolites in the Nanumba-North Municipality of the Northern Region of Ghana.

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).

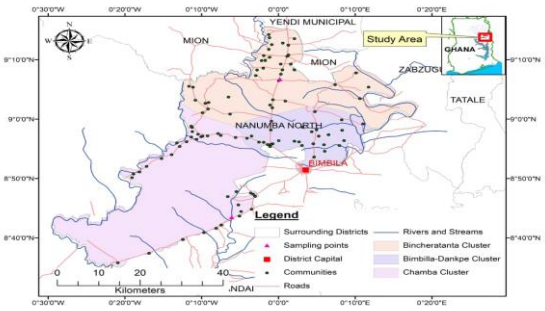

Figure 1: A map showing Nanumba North Municipality in the Northern Region of Ghana

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 DDT and its metabolites. 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 (Aiyesanmi and Idowu, 2012). 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 DDT 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 DDT and its metabolites 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 p,p'-DDT and its metabolite. One-way analysis of variance (ANOVA) was performed to analyse significant differences in the concentrations of p,p'-DDT and its metabolite detected from the samples.

3.0 Results and Discussion

3.1 p,p'-DDT and its metabolites (p,p'-DDE, and p,p'-DDD) in samples from Nanumba-North Municipality

p,p'-DDT and its metabolite (p,p'-DDE, and p,p'-DDD) were not found in the water and food crop items but were detected in 90% of the soil samples. However, in the water and food crop samples in the Nanumba-North Municipality, the absence of DDT and its metabolites may implicate that their concentration levels were below the limit of detection of the applied analytical method. Perhaps, the absence of parent compound and its metabolite in the water and food crop samples may be attributed to the highly less soluble nature of DDT (1 µg L^{-1} at 25^{o}C). It is quite intriguing to note that, the initial attack on p,p'-DDT in soil samples appears to be centred on the aliphatic trichloro ethyl group of the molecule and proceeds in either one or two directions, depending on the prevailing environmental condition (Figure 2.1). Under aerobic conditions, p,p'-DDT undergoes dehydrochlorination to yield p,p'-DDE. Under anaerobic conditions, transformation of p,p'-DDT to p,p'-DDD by reductive dechlorination is considered to be the dominant reaction. This

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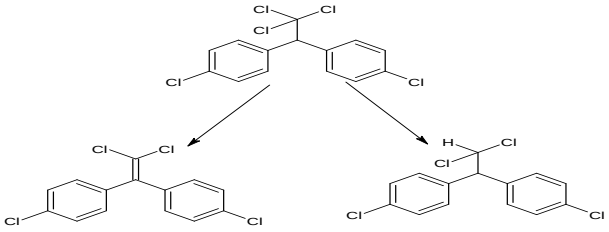
suggested that in the study area p,p'-DDT in the environment gradually breaks down into p,p'-DDD and p,p'-DDE which are more stable. The order of percentage of the parent compound and its metabolites occurring in the soil samples was p,p'-DDD > p,p'-DDE > p,p'-DDT. This implies that the transformation rate of the p,p'-DDD was faster than p,p'-DDE in the samples. The potential reason why the levels of p,p'-DDT and its metabolites in the samples were low was genuinely down to environmental factors such as sunlight, high temperature, low relative humidity and volatilization. Photochemical reactions, by UV or visible light, may degrade DDT according to two types of processes, known as direct photolysis and indirect photodegradation. The intensity and spectrum of sunlight reaching the soil depends on time of the day, season, latitude, altitude and state of the atmosphere for example clouds of dust absorbing light, with shortest wavelength (λ) around 290 nm. When soils are exposed to sunlight photodegradation occurs and depends on soil characteristics and on the photodegradation mechanism. In fact, soil characteristics plays a critical role in photons penetration and its light absorption and scattering by particles. However, both vary with wavelength and moisture content. Perhaps p,p'-DDT, and its metabolites in the samples decreased with decrease organic carbon content. Besides, soil organic carbon influences adsorption and deposition of p,p'-DDT, and its metabolites in the samples, and since the organic carbon and organic matter in the sampled soil was low this could account for the low levels of p,p'-DDT, and its metabolites. Another possible reason could be, p,p'-DDT and its metabolites levels in the samples were associated with organic matter content of the soil and could be attributed to p,p'-DDT, and its metabolites molecules having significant tendency of binding to organic matter in soil, similar to fats or lipids of plants and animals as reported by Swackhamer and Hites (1988) and Bentzen *et al*., (2008). Furthermore, decrease in soil pH could decrease the concentrations of p,p'-DDT, and its metabolites, and since the pH in the sampled soil was low this could potentially account for the low levels of p,p'-DDT, and its metabolites. Lastly, the possible mechanisms for low levels of p,p'-DDT from the agricultural soils were due to erosion, volatilisation and uptake by plants and animals. This findings was in agreement with Senyo (2015), p,p'-DDD > p,p'-DDE > p,p'- DDT, but contrary to Wang *et al.,* (2006) and Manz *et al.,* (2001), p,p'-DDE > p,p'-DDD > p,p'-DDT and p,p'- DDE > p,p'-DDT > p,p'-DDD, respectively. Since the half-life of p,p'-DDT is 35 years then, the estimated time for the transformation of p,p'-DDT to p,p'-DDE, and p,p'-DDD in the agricultural soils were 23 and 35 years (Figure 4). The molar concentrations of p,p'-DDT, p,p'-DDE, and p,p'-DDD in the selected agricultural soils were 8.463 picomoles, 21.87 picomoles, and 44.021 picomoles. The mean concentration of p,p'-DDT, and its metabolites recorded in this study were below the MRL of 1.000 µg/L, (WHO, 2017) of agricultural soils. In general, the mean concentration of p,p'-DDT at the Bimbilla-Dankpe Cluster was higher than the mean values measured at Bincheratanga and Chamba Clusters (Table.1). According to Alloway and Ayres, (1999), p,p'-DDT is not very toxic to humans but its LD_{50} in rats is 110 mg/kg. It has been shown that a human test population group ingested 35 mg daily for an extended period without any ill effects but its fatal dose is estimated to be 500 mg/kg for humans, (Alloway and Ayres, 1999). The current trend of higher p,p'-DDD, and p,p'-DDE relative to p,p'-DDT was also observed by Ntow (2005) in sediment samples from the Volta Lake and Darko *et al.,* (2008) in sediment from Lake Bosomtwi.



p,p'-DDE = 21.87 picomoles p,p'-DDD = 44.021 picomoles

Figure 2.1: Degradation of* p,p'-*DDT to form* p,p'-*DDE (by elimination of HCl, left), and* p,p'-*DDD (by reductive dichlorination, right)

Table 1: p,p'-DDT, and its metabolites (µg/L) in soil samples in the Nanumba-North Municipality of Northern Region of Ghana.

CLUSTERS		p,p'-DDT, and its metabolites (µg/L)		
		p,p'-DDT	p,p'-DDE	p,p'-DDD
0-20 cm	BDCS1	0.003	0.006	0.020
	BDCS2	0.004	0.007	0.021
	BDCS3	0.005	0.005	0.024
	BCS1	0.003	0.006	0.020
	BCS2	0.003	0.008	0.020
	BCS3	0.003	0.007	0.020
	CCS1	0.002	0.008	0.002
	CCS2	0.003	0.005	0.002
	CCS3	0.004	0.011	0.003

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x̄±SD	0.003±0.001	0.007±0.002	0.015±0.009
XI	ND	ND	ND
MRLs (WHO, 2017)	1.000	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 DDT and its metabolites (µg/L) in water bodies in the Nanumba-North Municipality of the Northern Region of Ghana (ND: Not-detected)

Table 2.0a: Levels of p,p'-DDT and its metabolites in Waanpu Dam

pesticide residues	**Waanpu Dam**				
	Depths(µg/L)				
	0-15cm	15-30cm	30cm and below	Mean±SD	WHO MRLs(µg/L)
p,p'-DDT	ND	ND	ND	0.000±0.000	1.000
p,p'-DDE	ND	ND	ND	0.000±0.000	1.000
p,p'-DDD	ND	ND	ND	0.000±0.000	1.000

Table 2.0b: Levels of p,p'-DDT and its metabolites in Bincheratanga Dam

pesticide residues	**Bincheratanga Dam**				
	Depths(µg/L)				
	0-15cm	15-30cm	30cm and below	Mean±SD	WHO MRLs(µg/L)
p,p'-DDT	ND	ND	ND	0.000±0.000	1.000
p,p'-DDE	ND	ND	ND	0.000±0.000	1.000
p,p'-DDD	ND	ND	ND	0.000±0.000	1.000

Table 2.0c: Levels of p,p'-DDT and its metabolites in Kumbo River

pesticide residues	**Kumbo River**				
	Depths(µg/L)				
	0-15cm	15-30cm	30cm and below	Mean±SD	WHO MRLs(µg/L)
p,p'-DDT	ND	ND	ND	0.000±0.000	1.000
p,p'-DDE	ND	ND	ND	0.000±0.000	1.000
p,p'-DDD	ND	ND	ND	0.000±0.000	1.000

Table 3: Levels of p,p'-DDT and its metabolites in food crop samples

pesticide residues	**Food crops items**				
	Selected food crops (µg/L)				
	Yam	Maize	Ayoyo	Mean±SD	WHO MRLs(µg/L)
p,p'-DDT	ND	ND	ND	0.000±0.000	1.000
p,p'-DDE	ND	ND	ND	0.000±0.000	1.000
p,p'-DDD	ND	ND	ND	0.000±0.000	1.000

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Table 4: p,p'-**DDT metabolites in soil samples in the study area**

p,p'-DDT and its metabolites 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**
p,p'-DDT	0.003	p,p'-DDE	0.007	35 years	23 years
		p,p'-DDD	0.015		35 years

4.0 CONCLUSIONS AND RECOMMENDATIONS

4.1 CONCLUSION

From this study, p,p'-DDT and its metabolite (p,p'-DDE, and p,p'-DDD) were not found in the water and food crop items but were present in 90% of the soil samples analysed. The presence of p,p'-DDT, and its metabolites in the soil samples could be as a results of p,p'-DDT use by farmers in the study area. The mean concentrations of p,p'-DDT and its metabolites in the samples analysed from the study site were generally below MRLs (WHO, 2017) of 1.000 µg/L, respectively. p,p'-DDT and its metabolites 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. However, the mean concentration of p,p'-DDT at the Bimbilla-Dankpe Cluster was higher than the mean values measured at Bincheratanga and Chamba Clusters.

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 conduct regular DDT monitoring in the municipality to prevent, control, and reduce environmental pollution, thereby minimizing health risks to the population.

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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