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Concentration and Human Health Risk Assessment of Dichlorodiphenyltrichloroethane in Two Species of Fish Muscle from River Gongola Basin and its Dam, Dadinkowa, Gombe State, Nigeria

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ABSTRACT: The concentration of DDT and its metabolites in the two fish species (*Clarias gariepinus* and *Bagrus docmak*) from River Gongola basin and its dam, Dadinkowa in Gombe State Nigeria was determined and human health risk association from the consumption of contaminated fish was evaluated. Fish samples were collected for period of six months and the pesticide residues analysis was carried out by using QuEChERS method along with conventional method (Hand shaken technique) and gas chromatography with electron capture detector (GC-ECD). (DDTs) and its metabolites were observed with DDD were the predominant contaminants, followed by DDT. The predominance of DDE may be attributed to their current use in vector control and contamination from past usage. The estimated daily intakes (EDIs) of DDT and its metabolites from all fish species were much lower than the acceptable daily intakes (ADIs), indicating that consumption of fish is at little risk to human health at present. However, the cancer risk estimates in the area of concern and the hazard ratios (HRs) of DDTs below the threshold value of one, indicating daily exposure to these compounds is a potential concern.

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Keywords: Gongola River; *Clarias gariepinus; Bagrus docmak;* Health human risk

The organochlorine pesticide dichlorodiphenyl-trichloroethane (DDT) is one of the persistent pesticides in the environment, meaning that its effects last for a long period. Organochlorine pesticides have a high solubility in lipids and a low solubility in water, which means they tend to bioaccumulate and biomagnified in food chains (Harrad, 2009). They have public and environmental health implications. (ATSDR, 2002; UNEP, 2005; World Health Organization, 2010). Pesticides are spread throughout the environment by a variety of methods and agents. When pesticides are used, substantial amounts of the chemicals end up in the soil and the atmosphere. Precipitation washes pesticide residues out of the atmosphere, or they fall into the land and water. When infected leaves or crops fall to the ground or are washed away by rain, pesticide residues can accumulate in the soil. (ATSDR, 2002; Rathore and Nollet, 2012). Chemical contaminants such as pesticides find their way into water bodies, such as streams and rivers, and are eventually dumped into lakes and seas, where they are seldom broken down due to their physicochemical qualities. Nollet and Rathore (2012). As a result, water is the primary recipient of pesticide residues (ATSDR, 2002). DDT residues (p,p'-DDT, p,p'-DDT, and metabolites such as p,p'-DDD and p,p'-DDE) are soluble in fats and lipids of animals and plants, thus fish and other aquatic species can absorb them from the water and concentrate them in their fatty tissues when the water is contaminated. (World Health Organization, 2010). Because of its negative effects on human health and the environment, DDT is virtually universally banned for use in agriculture. Fish is the most important single

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source of high-quality protein providing about 17% of the annual protein consumed by world's population (Food and Agricultural Organization 2016). Contaminants ingested with food can build up in the tissues of fish. Furthermore, through diffusion across the gills and skin, fish can concentrate pollutants from the water (Gobas et al. 1999). Organochlorine pesticides (OCPs) have become a global issue due to their persistence, bioaccumulate potential, chronic toxicity, and potential negative effects on humans and wildlife (UNEP, 2001). It is well recognized that humans consume the majority of pesticide residues through the food chain (Martinez et al., 1997). Fish have been shown to biomagnified pesticides in the environment (Mackay and Fraser, 2000) and then transfer the poison to humans when eaten. Some of these substances have been linked to human cancers (IARC 2008) and have been shown to affect thyroid hormone concentrations (Meeker et al., 2007). Although OCPs have been prohibited or regulated in developed nations, they are still used for agricultural purposes and control of pest in underdeveloped countries like Nigeria, and as a result, they can be discovered in aquatic (Deribe et al., 2011) and terrestrial environments. The Gongola River and its Dams comprise numerous major tributes, including a densely inhabited area surrounded by varied agricultural operations, with an increasing tendency of pesticide use along the riverbank (Asferachew et al., 1998). Furthermore, Nigeria is one of the West African countries with a problem of outmoded pesticides that have accumulated since the 1960s, when the first imports began (Haylamicheal and Dalvie, 2009). Organochlorine chemicals such as DDT, DRINs, and lindane were mainly outlawed in most nations. According to this viewpoint, there is still a significant risk of pesticide exposure in the environment. As a result, water pollution is caused by intensive agricultural activity near the river and municipal waste discharges (Zinabu, *et al*., 2002). As a result, it is required to assess the water quality in terms of physical and chemical qualities, as well as the present status of DDT and its metabolites in several fish species from the River Gongola and its Dams. There was no research on the concentration and risk assessment of DDT and its metabolites along the River's and Dams' trend. As a result, the goals of this study are to determine the concentrations of DDT and its metabolites in two fish species and to analyze the possible dangers of DDT and its metabolites to human health posed by dietary consumption of these species.

MATERIAL AND METHODS

Area of Study: The Gongola River is the main drainage system, running approximately worth south towards the Benue river basin but with south toward the tributaries draining from west to east into the Gongola River (Figure 3.1), with Dadinkowa dam located about 5 km north of Dadinkowa village in Yamaltu Local Government Area of Gombe State in Nigeria's northeast. The location is located 37 kilometers to the east of Gombe, at longitude 10^{0} 19 19N and latitude 11^{0} 28 54. With a capacity of 800 million cubic meters of water and a surface area of 300 square kilometers, the dam is part of the River Gongola, whose drainage basin is located in north-eastern Nigeria. The Dam has two seasons: the rainy season, which is marked by significant rainfall and can result in flooding, and the dry season, which is marked by drier weather. The dry season is also marked by chilly, dusty, dry winds, which are followed by scorching heat. Around it is settlements dominated by farmers, businesses such as Ashaka Cement Company (one of West Africa's largest cement producers), and agricultural activity along the riverbanks. Dadinkowa is a rural environment with a 315-hectare field that is primarily used for irrigation and fishing. Consumer middlemen from Dadinkowa and the surrounding areas patronize the settlement's crops and seafood. Farmers use agrochemicals (fertilizer, herbicides, and germicides) to control pests and weeds and to improve crop yields as needed, without doing a follow-up study of how these agrochemicals (fertilizer, herbicides, and germicides) affect the environment of their settlements. Dadinkowa Dam is a three-fold dream, serving as a source of 34 megawatts of hydroelectric power, irrigation, and water supply for Gombe town and its immediate environs, as depicted in figures 1 and 2 respectively.

Fig1: Map of Gombe and Nigeria showing the study area

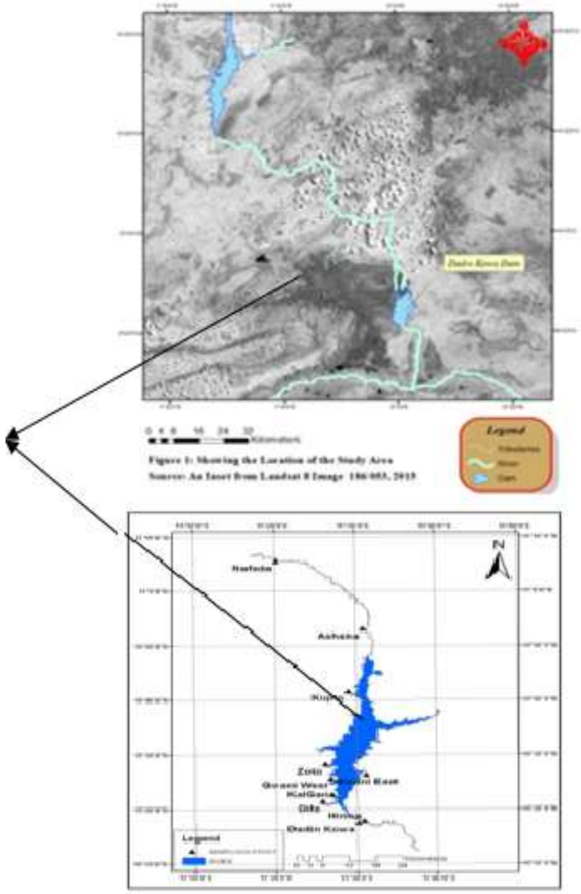

Fig 2: Map showing sampling point

Extraction of pesticides residue by QuEChERS method using conventional method: The method of Tatiana et al. (2018) was adopted and modified as follows: 2 g of homogenized fish muscle was weighted into a 50 ml centrifuge tube, 10 ml of 1:1 v/v acetone: hexane was added, and the mixture was immediately hand shaken for 1 minute, then 2 g of Na_2SO_4 was added and hand shaken again for 1 minute. After that, 4 g $MgSO_4$ and 1 g NaCl were added, shaken for 1 minute, and then centrifuged at 4000 rpm for 10 minutes at 4^{o}C. Four (4 ml) aliquots of the upper layer were placed into a 15 ml centrifuge tube containing 0.1 g PSA, 0.1 g C18, and 0.4 g $MgSO_4$, hand shaken for 1 minute, then centrifuged for 10 minutes at 5000 rpm. Gas chromatography with an electron capture detector was used to examine DDTs (p,p-DDT, p,p-DDE, and p,p-DDD) (Shimadzu GC-2014, Kyoto, Japan). To separate OCPs, a split less injection ENV-8 MS capillary column (30 m 0.25 mm i.d., 0.25 mm film thickness) was employed. Each sample was injected with one liter. The temperature in the column oven was originally set at 100 1C for 1 minute, then increased to 180 1C at 201C min1 and then to 260 1C at 41C min1 for 5 minutes. Temperatures for the injector and detector were 250°C and 310°C, respectively.

Health risk estimation: The USEPA's guidelines for possible risk assessment were utilized to assess the risk of pesticides found in each fish species on consumers. The reference dose (RfD) of any pesticide is the level of exposure that is likely to have no significant adverse effects, as determined by the EPA (USEPA, 2006). Hazard quotients (HQ, with equations (2)) were constructed using the estimated average daily intake (EDI, with equations (1)) to quantify the non-cancer risk of pesticide exposure.

Estimated daily intake: The estimated daily intake (EDI) was found by multiplying the average residual pesticide concentration (mg/kg) by the fish consumption rate (kg/day)

$$EDI = \frac{RC \times FC}{BW} \quad 2$$

Where RC = residual concentration, FC = food consumption; BW body weight (WHO, 1997; Fianko et al., 2011)

Hazard quotient (HQ): Hazard quotients were obtained by dividing the EDI with their corresponding reference dose (RfD)

$$HQ = \frac{Estimated\ daily\ intake}{Reference\ dose} \quad 3$$

Where HQ = hazard Quotient. (WHO, 1997; Fianko et al., 2011)

The food and agricultural organization (FAO, 2011) quotes the per capita consumption of fishes in Nigeria as 10 kg, while the body weight for adult population group is 70 kg.

RESULTS AND DISCUSSION

In general, fish's ability to metabolize organochlorine's is limited; as a result, pollutants build up in the fish, which is a mirror of pollution levels in the surrounding environment (Guo et al., 2008). Organochlorine insecticides have become commonplace pollutants with a wide spectrum of negative health effects in humans. Reproductive failures, immune system malfunction, endocrine disturbance, and breast malignancies are among the harmful effects. (Garabrant et al., 1992; Kolpin et al., 1998; Bouman, 2004; Ize-Iyamu et al., 2007; Adeyemi and Ukpo, 2008). Many organochlorine insecticides and their metabolites have been linked to a variety of negative human and environmental impacts, including birth abnormalities and reproduction. Despite the fact that DDT and its metabolites were banned since 2008 by Nigeria government, there were still traces of DDT and its metabolites persist in the environment and are known to accumulate in aquatic life. DDT, DDE and DDD have all been designated by National Food and Drugs Administration Control (NAFDAC) in Nigeria as potential human carcinogens by Nigeria. DDT and its metabolites are known to cause cancer. p, P'-DDE, p, P'-DDD, and p, P'-DDT were all detected in 100 percent of fish samples from both species during the wet and dry seasons. This has been confirmed in a number of studies, and according to Ize-Iyamu et al. (2007), Adeyemi and Ukpo (2008), and Ezemonye et al. (2009), it could be due to the persistence and long-range transport characteristics of DDT and its metabolites (2009). However, p, P'-DDE and p, P'-DDD shows the predominant congener accounting for 83 percent and 75 percent follow by p, P'-DDT which accounts for 12 percent and 16 percent and p, P'-DDD accounting for 5 percent and 9 percent in *Clarias gariepinus* during rainy and dry seasons respectively. While in *Bagrus docmak* p, P '-DDE accounts for 67 percent and 65 percent followed by p, P '-DDT and p, P '-DDD accounting for 22 percent and 11 percent and 23 percent and 12 percent respectively. Figure and Figure indicate the percentages during the rainy and dry seasons.

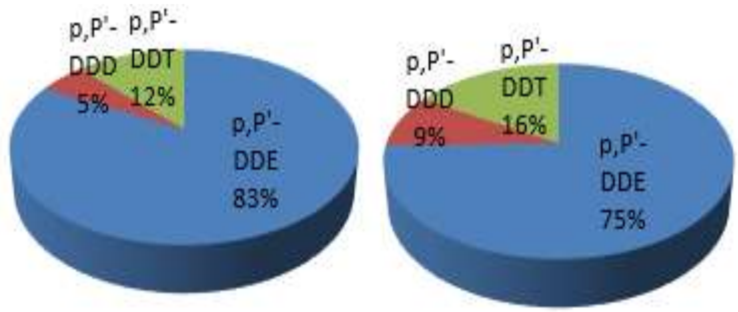

Fig 3 Show the percentage composition of DDT and its Metabolites in *Clarias gariepinus* during wet and dry season.

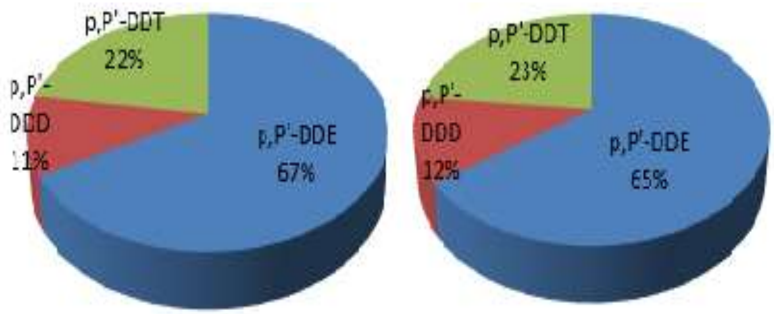

Fig 4. Show percentage composition of DDT and Metabolites in *Bagrus docmak* during wet and dry season

The quantities of DDT and its metabolites in all of the fish samples were substantially higher than the extraneous maximum residue limit (EMRL) of 1.0 g/kg (0.001 mg/kg), showing that pesticide residues in Clarias gariepinus had contaminated the aquatic environment severely. According to the findings, p, P'-DDE had the lowest and greatest mean concentrations in the rainy season, ranging from 0.15 mg/kg in the Nafada river to 2.8 mg/kg in the Dadinkowa dam. During the dry season, the minimum and maximum mean concentrations in Dadinkowa and Kalgari dams were 0.21 mg/kg and 0.53 mg/kg, respectively. p, P' DDD had mean concentrations in the rainy season ranging from 0.02 mg/kg in the Nafada river to 0.08 mg/kg in Kupto dam, whereas in the dry seasons, the mean concentrations varied from 0.02 mg/kg in Dadinkowa dam to 0.09 mg/kg in Zoto dam. In the wet season, the minimum and maximum mean concentrations ranged from 0.016 mg/kg at Dadinkowa Dam to 0.17 mg/kg at Zoto Dam, whereas in the dry season, the minimum and maximum mean concentrations ranged from 0.016 mg/kg at Dadinkowa Dam to 0.17 mg/kg at Zoto Dam. During the rainy season, the lowest and greatest mean concentrations of Bagrus docmak p, P' DDE were 0.15 mg/kg and 0.63 mg/kg in Gwani West and Kupto dam, respectively. During the dry season, the lowest mean concentration was 0.14 mg/kg at Zoto Dam, with the highest mean concentration of 0.63 mg/kg in the Nafada River. The mean concentration of p, P' DDD ranged from 0.03 mg/kg in Kalgari dam to 0.08 mg/kg in Ashaka river during the rainy season, while the lowest and greatest concentrations of 0.02 mg/kg and 0.15 mg/kg were reported in Zoto and Dadinkowa dams, respectively, during the dry season. During the rainy season, mean DDT concentrations ranged from 0.05 mg/kg in Dadinkowa dam to 0.22 mg/kg in Ashaka River, whereas the minimum and maximum mean concentrations ranged from 0.03 mg/kg in Kalgari dam to 0.53 mg/kg in Difa dam during the dry season. However, all of the results for the presence of DDT and its metabolites in the two fish species studied were lower than those reported by Akan et al (2009). This could be due to the pass's extended half-life, slow degradation, and long-term persistence in the environment. As a result, the total concentration of DDT and its metabolites was less than the Nigerian pesticide residue standard of 3.83 mg/kg and the maximum recommended limit of 5.0 mg/kg (FAO/WHO 1986). In comparison to this study, Ssebugere et al. (2009), Werimo et al. (2009), and Ezemonye et al. (2009) reported lower concentrations. The carcinogenic effect was assessed for the DDT and its metabolic residues detected in the selected muscle of the fish species and Hazard Ratio (HR) were calculated in all the locations during rainy and dry seasons. The slope factors were obtained from USEPA's integrated Risk Information system (IRIS). From the results obtained, the hazard ratio of the DDT and its metabolic was below one (1) which therefore suggest that, there is no potential risk to human health by the population for the consumption of the two varieties of fish within the ten different locations during rainy and dry seasons,

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