Detection of Organochlorine Pesticide Residues in Lake Ziway and Health Risk Assessment

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Abstract
Organochlorine pesticides (OCPs) are chlorinated hydrocarbons extensively used for agriculture pest control and mosquito control from 1940s to 1960s. The use of OCPs for agricultural purpose was banned in many countries including Ethiopia. However, studies show that small scale farm holders in the central rift valley of Ethiopia use cocktail of pesticides including banned OCPs. This study was intended to detect the status of (OCP) residues in Ziway Lake, Oromia region of Ethiopia. For this study three commercially important fish species, sediment and succulent grass samples were randomly collected in replicate. The samples were extracted using USEPA (354 °C) method and the analysis was done using USEPA (8081) method. The health risk assessment was calculated based on USEPA (1996) guidelines while the data analysis was done using SAS 93.2 statistical software. The Laboratory result revealed that Dichlorodiphenyltrichloroethane (DDT) metabolites, (p,p'-DDT and p,p'-DDE) were detected in all the three sample types. Accordingly, mean concentration of p,p'-DDT and p,p'-DDE detected in the three sample types were, 52.08 and 4.67 ng/g in succulent grass, 46.84 and 0.93 ng/g in sediment and 19.83 and 11.17 ng/g in fish samples. Among the three fish species tested, the maximum DDT metabolite residue detected was in Ciprinu scarpio fish spices (35.65 ng/g) while the minimum was in Carassius carassius fish spices (27.05 ng/g). People who regularly consume fish from the lake above estimated mean Ethiopian daily consumption (0.027 kg day⁻¹), level are exposed to DDT while that those consume all species of fish from Lake below estimated mean, does not exposed to health problem. However, the low p,p'-DDE /p,p'-DDT ratio in sediment and succulent grass sample indicates that there is a recent pollution of the lake by DDT. Thus, Eco-system clearance from obsolete pesticides and control of illegal band organochlorine pesticide movement has to be given due attention.

Keywords
Organochlorine pesticide, Fish, Bio-accumulation, Health risk

Abbreviations
OCPs: Organochlorine Pesticides; USEPA: United states Environmental protection Agency; pp'DDT: p,p'-Dichlorodiphenyltrichloroethane (DDT); pp'-DDE: Dichloro diphenyl dichloroethylene; PSD: Sediment Particle size distribution; FAO: Food and Agricultural organization; ROCP: Residue organochlorine pesticides; HI: Hazard indices; LED: The lifetime exposure dose; RfD: Reference dose; EEFRI: Ethiopian Environment and Forest research institute

Introduction
Organochlorine pesticides (OCPs) are chlorinated hydrocarbons. These chemicals had been used extensively in the agriculture sector to control pests and in the health sector to control mosquito commencing 1940’s to 1960’s [1].

The legally-binding agreement between states was made and Stockholm convention (SC) was signed in 2001 and entered into force in 2004 by many countries with the overall objective of protecting human health and the environment from POPs. Ethiopia signed this agreement on 17 May 2002 and ratified the instrument on 2 July 2002. As neurotoxicants, Organochlorine pesticides were banned in many countries including Ethiopia who ratified Stockholm convention. However, DDT has been used for the purpose of disease vector control (malaria) in the country until 2011. These resulted in accumulation of obsolete (DDT) pesticide, poor storage facility, luck of destruction incinerators are among the problems facing the country. (UNEP-POPS-NIP-Ethiopia), [2] current inventory report on obsolete pesticide stores by Ministry of Environment Forest and Climate change shows reported significant quantity of obsolete pesticide stores [3]. Volatilization

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Materials and Methods

Sampling site description

The study area, Lake Ziway, is located in the geographical between 7° 52’ to 8° 8’ N and 38° 40’ to 38° 56’ E and has an altitude of 1636 m.a.s.l. The Lake is the fourth largest Lake in Ethiopia with an open water area of 434 km². It is the shallowest of the rift valley lakes with maximum and mean depth of 8.95 m and 2.5 m, respectively [8].

Sample collection, procedure and Analysis

Three composited replicate sediments samples were collected from three different locations in the lake (Figure 1). Each sampling point was 150 meters from the margin of the lake and the diagonal distance between each sampling location was 600 meters. Also, three composited succulent grass samples in the lake were collected from the same place where the sediment samples were collected in replicate. Among Fish spices in the lake; three species were identified based on their preference by local community. These were Oreochromis niloticus; Ciprinus carpio; Carassius carassius species. From each fish species ten individual fish was collected in replicate.

Sample preparation: The sediment samples were air dried and extraneous substance were removed grinded and passed through 2 mm sieve before subsequent labo-
Sample extraction and analysis: Prior to the organochlorine extraction and analysis; sediment size distribution (PSD) was analyzed by laser diffraction method using laser scattering particle size distribution analyzer while organic carbon of the sediment was determined by Weakley and Black method. The fish spices were analyzed for their lipid percentage by Soxhlet extraction method.

The Organochlorine pesticides (OCPs) in the samples were extracted by multi residue method. (EPA method 354 °C). 10 g of sample was placed into a beaker containing 10 g anhydrous sodium sulfate for sediment and succulent grass for fish 40 g of sodium sulfate used and mixed thoroughly. The sample mixture was transferred to an extraction thimble and placed in a Soxhlet extractor. The mixture was extracted with 300 ml of acetone: n-hexane (1:1 v/v) at 6-24 hours at 4-6 cycle/hr for 20 h.

The extracts were concentrated to 2 ml using vacuum rotary evaporator and nitrogen gas bubble. Each of the raw concentrated extract was then re-dissolved in 10 ml hexane. Column containing 3 gram of activated silica gel and 3 g sodium sulfate was conditioned with hexane. The sample passed through pre-conditioned column.

The sample which was trapped in the column was eluted with 80 ml of hexane, collected and concentrated to 5 ml by blowing nitrogen gas and the elute was transferred to 10 ml volumetric flask, diluted to the mark with hexane and transferred to GC/MS auto sampler vial. The analysis was done using 7890B GC with Inlet: Split/Split- fewer modes; DB-5 ms Column and 5977B MS Detector equipped with Mass Hunter Chromatographic system.

Health risk assessment

Consumption of contaminants in food was calculated based on its concentration in the fish, and on an estimate of the fish consumption rates. The lifetime exposure dose (LED) (mg kg⁻¹ day⁻¹) was obtained, and the hazard indices (HI) for each age class were estimated.

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LED = \text{Residue concentrations in fish tissue sample (mg/kg)} \times \text{fish consumption rate (kg/day)} / \text{Body weight (kg)}
\]

\[
HI = \frac{\text{Estimated dose}}{\text{Reference dose}}
\]

The following data’s were extracted from (USEPA) [9] and used for calculation:

• Hypothetical body weight (of 10 kg for children in age-class 0-1 year, 30 kg for children in age-class 1-11 years, and 70 kg for adults).

• Maximum absorption rate of 100% and a bioavailability rate of 100%

• The reference dose (RfD) of fish (µg kg⁻¹ day⁻¹) is 0.5

• Fish consumption rate in Ethiopia, estimated to be 0.027 kg day⁻¹ [10].

Data validation and analysis

Sample of known concentration was processed and quantified to control the equipment performance efficiency.

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Figure 2: Comparison of DDT and DDE concentration in Fish, sediment and succulent grass collected from Lake Ziway, Ethiopia.
Results and Discussion

Organochlorine pesticide residues in Lake Ziway

DDT and its metabolic form (pp’DDT and pp’-DDE) are present in sediment, succulent grasses and fish muscle tissue collected from Lakes Ziway. This condition is mainly due to the persistence of these group of compounds, even though their usage is discontinued.

Among the analyzed Organochlorine Pesticides only DDT and its metabolite were detected in the three samples (Fish, Sediment and grass). Analysis of variance showed non-significant difference among the three matrices in terms of total DDT contents. However, for the degraded product of DDT (PP’DDE), there was significant difference among the three sample types (P < 0.005). The mean concentration of p,p’-DDE; in fish (11.17 ng/g) > succulent grass (4.67 ng/g) > sediment (0.93 ng/g) respectively (Figure 2).

The low degraded DDT Product in sediment sample can be explained by low organic carbon content (0.26; 0.4 and 0.6%), and the sandy texture of the sediment. Generally, sandy and course texture has low organochlorine chemicals affinity which makes the chemicals unavailable for microorganism for degradation.

The result shows a highly significant difference (P < 0.0001) among the fish species in terms of total DDT, p,p’-DDT and p,p’-DDE (Table 1). The sum of DDT concentration in the fish was higher in Carassius carassius spices (35.65 ng/g) followed by Oreochromis niloticus (30.29 ng/g) and Carassius carassius (27.07 ng/g). There was significant difference between Oreochromis niloticus and Carassius carassius and between Ciprinus carpio and Carassius carassius. The highest p,p’-DDE was recorded in Ciprinus carpio followed by Oreochromis niloticus and Carassius carassius in that order. This can be explained by the lipid content of the fish’s, the mean concentration of high p,p’-DDE (15.25 ng/g > 9.97 > 8.31) among the fish species (1.29 > 0.72 > 0.52)% Lipid.

The low p,p’-DDE/p,p’-DDT ratio of Oreochromis niloticus/Carassius carassius shows less than 50% of fresh DDT was degraded. This value is an indication of recent contamination of the fish species by fresh DDT from non point source pollution including dry deposition and wet deposition of DDT from obsolete pesticide store in the rift valley the finding agrees with the report of other researchers such as Daniel.

Health risk assessment

Comparison between lifetime estimated dose and the reference dose shows that there is no vulnerable population sub-group. Feeding fish to children would not consequently expose them to systemic toxicity. The estimated dose of DDT for adults would not exceed reference dose (with hazard index below 1) (Table 2), consumption of all species fish does not poses a health risk for a human health based on the current findings.

However, the fish fillet is still prepared and served as soup daily by the local fishermen families. For those who eat fish daily and much more than the estimated mean Ethiopian daily consumption (0.027 kg day⁻¹), the DDT intake will thus be considerably higher [10]. Therefore, children from the fishermen families may be the most exposed and vulnerable group among the local people.

Conclusion and Recommendation

The findings of this study shows that the lake eco-system is under threat and need to be protected from pollution by obsolete pesticide. In conclusion, my results suggested the need for eco-system clearance from obsolete pesticides store to reduce which could be the case for the lake contamin-

Table 1: Concentration of ΣDDT (ng/g), p,p’-DDT (ng/g) and p,p’-DDE (ng/g) in different fish species sampled from Lake Ziway, Ethiopia.

| Fish species                | Mean ΣDDT (ng/g) | p,p’-DDT (ng/g) | p,p’-DDE (ng/g) |
|----------------------------|-----------------|----------------|----------------|
| Oreochromis niloticus      | 30.30a          | 20.33a         | 9.97a          |
| Ciprinus carpio            | 35.65b          | 20.38b         | 15.25b         |
| Carassius carassius        | 27.05c          | 18.78c         | 8.31c          |
| P-value                    | < 0.0001        | < 0.0001       | < 0.0001       |

Values in the same column with different letters are significantly different by the Duncan’s multiple range test (α = 0.05).

Table 2: Estimated dose values and hazard indices of ROCP exposures in fish at different trophic levels (O. niloticus, Ciprinus carpio and Carassius carassius) sampled in Lake Ziway, Ethiopia.

| DDT mg/kg | Fish species              | RfD (µg kg⁻¹ day⁻¹) [9] | Estimated dose (µg kg⁻¹ day⁻¹) 0-1 years | Hazard index 0-1 years | Estimated dose (µg kg⁻¹ day⁻¹) 1-11 years | Hazard index 1-11 years | Estimated dose (µg kg⁻¹ day⁻¹) Adult | Hazard index Adult |
|-----------|---------------------------|-------------------------|------------------------------------------|------------------------|-------------------------------------------|------------------------|--------------------------------------|------------------|
| 0.030     | Oreochromis niloticus     | 0.5                     | 0.081                                    | 0.011571               | 0.162                                     | 0.054                  | 0.023                                |                  |
| 0.036     | Ciprinus carpio           | 0.5                     | 0.0972                                  | 0.013886               | 0.1944                                   | 0.0648                 | 0.0278                               |                  |
| 0.027     | Carassius carassius       | 0.5                     | 0.0729                                  | 0.010414               | 0.1458                                   | 0.0486                 | 0.0208                               |                  |

N.B body weight of 10 kg for children in age-class 0-1 years, 30 kg for children in age-class 1-11 years, and 70 kg for adults Numbers in bold indicate hazard indices greater than one.
Illegal movement of banned organochlorine pesticides has to be given due attention. People who live around Lake Ziway, and regularly consume fish from the lake above estimated mean Ethiopian daily consumption (0.027 kg day\(^{-1}\)), level are exposed to DDT while those consume all species of fish from Lake Ziway below estimated mean Ethiopian daily consumption level (0.027 kg day\(^{-1}\)), does not expose to health problem.

This finding can contribute the state of environment report regarding the area with respect to POPs which is a global issue. Detail assessment on the impact of obsolete pesticides on pollinators; birds, and soil beneficiary microorganisms and farm land is needed to be investigated.

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