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Organochlorine pesticides in rain, rivers and groundwater in the Lake Naivasha basin and implications for their management

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ABSTRACT

Organochlorines (OCs) have a high persistence in the environment and toxicity to humans and wildlife. Despite their ban in several countries, they are still found in agricultural areas. Their presence in drinking water resources (rivers, rain and groundwater) in Lake Naivasha area, which has had a booming horticultural industry in the last three decades was investigated. Twenty-two water samples from the rift floor around Lake Naivasha were collected in June 2017 and analysed for organochlorines. Findings show that all the water samples are contaminated with various organochlorine pesticides and metabolites. The abundance of OCs in water resources can be ranked as follows, rain> river >groundwater in which 18 compounds were identified; alpha, beta, gamma and delta HCH, endrin, aldrin, heptachlor, chlordane, b-endosulfan, methoxychlor, *p,p'*-DDE, *p,p'*-DDD, endrin aldehyde, endosulphan sulphate, *p,p'*-DD and endrin ketone. Their concentrations range from 0.1 to1 µg/L. The OCs with high concentrations are aldrin, endrin aldehyde and endosulphan sulphate. Twelve banned compounds were found present, several decades after their ban (1986 - 2014) shows either occasional usage in the region or the residues drift from use elsewhere. Despite the low concentrations, many of these pollutants can exceed acceptable daily intake (ADI) for humans. Thus, the National Environment Management Authority, Kenya needs to set OCs guidelines on drinking water quality and concerted efforts with agriculture, water and health ministries is needed to ensure public health safety.

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1. Introduction

Freshwater resources globally are threatened by increase in use of various chemicals in the agricultural sector. Because a significant amount of inputs such as pesticides and fertilizers are required to ensure high yields. Studies report presence of pesticides in surface water and groundwater close to agriculture lands in different parts of the world [1-7]. High levels of pesticides adversely impact ecosystems and human health. For example, organochlorines, a group of chlorinated compounds widely used as pesticides, are classified as persistent organic pollutants

(POPs) with high persistence and toxicity in the environment. They are banned in many countries; however, some are still used in developing countries for agricultural and public health purposes [8]. Currently about 31 pesticides are banned in Kenya and 718 products are registered for use, of these about 28% are not approved in Europe because of their potential human or environmental health effects [9, 10]. Studies have shown that over 98% of sprayed insecticides and 95% of herbicides reach a destination other than the target, namely; air, water, bottom sediments and food [11, 12].

Their fate in the environment depends on their physicochemical properties (persistence, solubility, and vapour pressure), application frequency and concentration as well as on environmental characteristics (rain, temperature, soil properties) [13]. Organochlorines reach rivers or lakes through runoff from agricultural areas and infiltration through soils thereby affecting many aquatic and terrestrial species. For example, microorganisms, invertebrates, plants and fish are badly affected by pesticides in the environment [14-16]. In addition, many of the organochlorine molecules are carcinogenic and neurotoxic and are therefore detrimental to public health [17, 18]. Epidemiological studies expose the etiological relationship between Parkinson's disease, prostate, lung and breast cancers, and Hodgkin's lymphoma with organochlorine pollutants [19]). Though such studies have been established in various parts of the world, comparable assessments for the developing world where commercial agriculture is growing are missing. Moreover, studies on pesticide pollution in groundwater are still few globally. Over the years the quantity of pesticides used has increased considerable, In Kenya between 2008 and 2013 a total of 54,516 tonnes insecticides, fungicides, and disinfectants were imported [20, 21]. As commercial agriculture and uses of pesticides increases, non-point source pollution will have a considerable influence on the quality of groundwater.

During the past decades, Naivasha basin in Kenya has had significant growth in the agricultural industry. Started in the 1980s, it has been associated with the degradation of soil and water quality [22-24]. Different pesticides are used in the agricultural industry [25-27] and health sector [28], however, there is no systematic monitoring and set guidelines for drinking water quality. This poses an exposure risk to the health of the residents and the ecosystem. Previous studies have reported some organochlorines in the lake, sediment and fishes [29]. However, groundwater is used by over 50% of the residents of Naivasha and there is still a gap in knowledge in the status of organochlorines in groundwater. The objective of this study was to carry out an investigation to determine the organochlorines present and their concentrations in groundwater, rivers and rainfall in the Naivasha basin.

2. Materials and Methods

2.1 Study Area

The Lake Naivasha basin is located in the central Kenya rift, which is part of the eastern arm of the East African rift, formed due to the tectonic and volcanic processes initiated in the Miocene [30, 31] (Figure 1). Thus the study area is characterized by faulted blocks and alkaline volcanic rocks such as basalts, comendites, trachytes and tuffs [30, 31]. Weathering of these rocks has created rich volcanic

soils; this, together with availability of freshwater in the area, makes it agriculturally productive. Since the 1980s this area has been at the centre of horticultural growth contributing at least 1.44% of the country's GDP [22, 32]. In two decades (1985-2005), the population grew by 300%, in part due to the growth of the flower farming industry, tourism, and fishing industries [22].

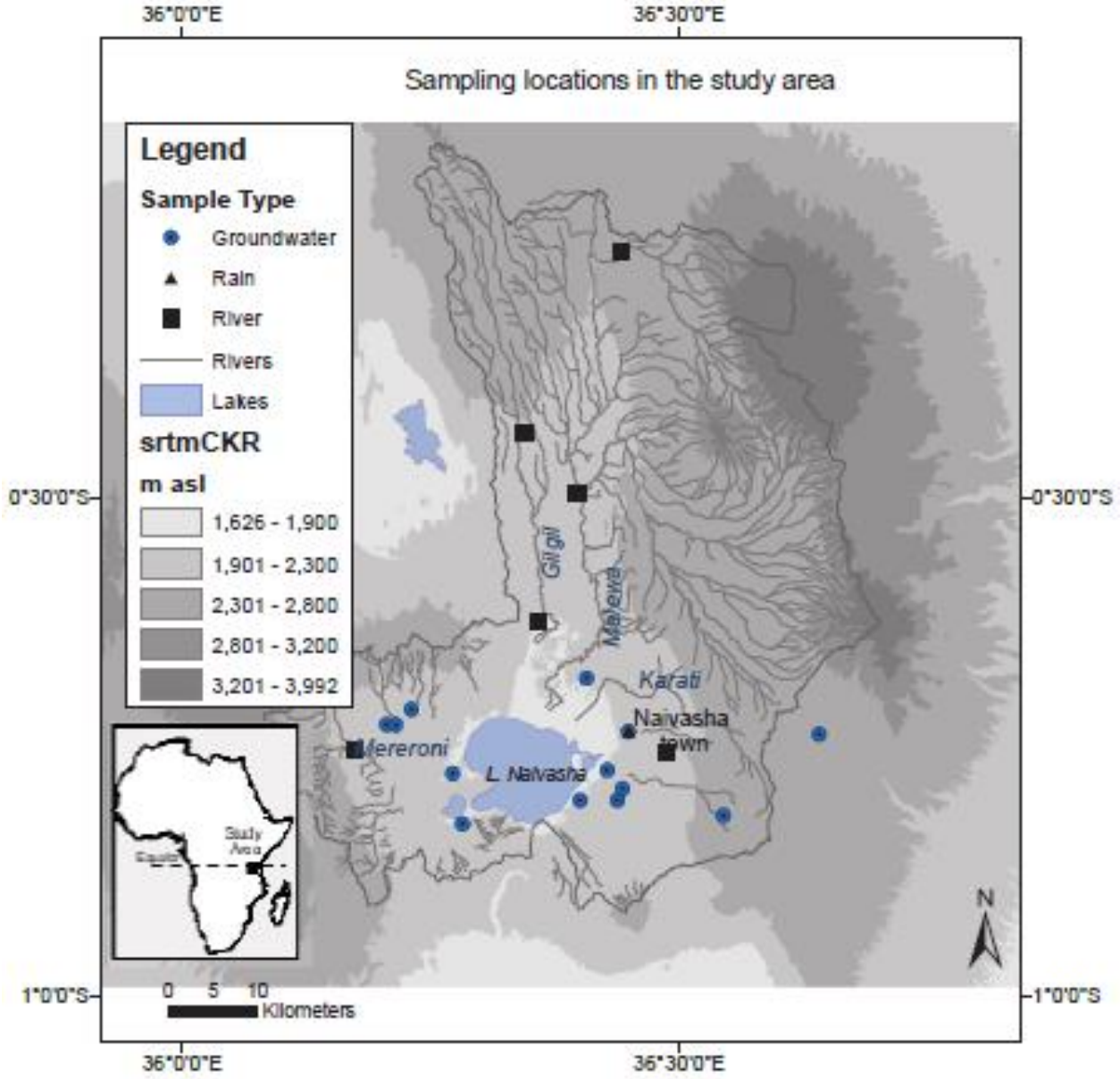

Figure 1: Map of study area showing location of samples from groundwater, rivers and rain.

The climate of the area ranges from humid in the escarpments which receive over 2400 mm of rainfall annually to semi-arid in the rift floor which receives about 600 mm/year, and evaporation is three times higher than precipitation on the rift floor. Four main rivers drain the high eastern and western escarpments into the Lake Naivasha basin. The water resources in the study area, excluding the lake, include four main rivers (Melewa, Gilgil, Karati, Mereroni) (Figure 1), and groundwater from over 300 shallow and deep boreholes that are used for domestic, agriculture, energy generation and industrial

purposes. In the semi-arid rift floor, irrigation agriculture is practiced using groundwater, lake and river water to grow flowers and horticultural vegetables, while in the humid escarpments agriculture is mainly rain-fed and wheat, carrots, peas, potatoes and tea are grown. To achieve high yields and good quality of produce, synthetic pesticides and fertilizers are used [25]. Lake Naivasha is a Ramsar site, a wetland of international importance since 1995 [33] and also a biodiversity hotspot with several species of birds and animals attracting a large number of tourists annually.

2.2 Sample Collection and Preparation

Twenty-two water samples (15 groundwater, 6 rivers, 1 rainfall) were collected in duplicates in 1 L amber glass bottles from rain, rivers and groundwater sources around the lake in June 2017. The samples were kept cool during the fieldwork duration and were transported to and refrigerated in the Organic Chemistry Laboratory, Department of Chemistry, University of Nairobi.

2.3 Extraction of Pesticides from Water Samples

Extraction of pesticides from water samples was done by solvent–solvent extraction procedure which was adopted from US EPA Method 3510C [34]. The pH of the 1.0 L of sample was measured and pH recorded. 50 ml of 0.2 M dipotassium hydrogen phosphate buffer was added to the sample. The pH was adjusted to 7.0 by carefully adding drops of 0.1 M hydrochloric acid or 0.1 M sodium hydroxide solutions. The neutral solution was then treated with 100 g of sodium chloride to salt out the pesticides to the organic phase, then this was followed by addition of 60 ml dichloromethane. The mixture was shaken further while releasing pressure and allowed to settle for 30 minutes to enhance separation into two phases. The lower organic layer was then collected into a pre-cleaned and dry 250 ml conical flask and the extraction process was repeated twice each time with 60 ml dichloromethane. The combined extracts were dried using activated anhydrous Na_2SO_4 and 2 ml of isooctane added as keeper then concentrated to about 3 ml using LABCONCO rotary evaporator. The concentrated extracts were then put in vials and stored in a fridge at -4 ^oC, awaiting the clean-up process.

2.4 Sample Clean-up

Sample clean-up was done using an alumina chromatographic column of 25 cm x 1.5 cm diameter packed with 1 g of activated anhydrous sodium sulphate (drying agent) followed by 15 g of deactivated alumina and finally another 1 g layer of activated anhydrous sodium sulphate. The column was pre-conditioned with 15 ml of HPLC grade hexane and the liquid was then discarded. The extracts were each introduced into the column and eluted with 165 ml of HPLC grade-hexane into a round-bottom flask. Each was then rinsed four times with 2 ml portions of HPLC grade-hexane. 2 ml of iso-octane was added to each cleaned sample as a keeper then the samples were concentrated to about 1ml using a rotary evaporator. The extracts were transferred into a clean pre-weighed auto vial and concentrated to 0.5 ml under a gentle stream of white spot nitrogen, after which the samples were ready for analysis.

2.5 GC Analysis and Quantification of the Extract

Analysis of pesticides was carried out using GC-MS (Agilent HP 6890) combined with an auto sampler (Agilent 7683 series injector). The HP 19091J-102 capillary column of 25 µm x 20 µm internal diameter x 0.33 µm film thickness coated with cross-linked 5% phenyl methyl siloxane was used. The carrier gas used was helium at a flow rate of 1.0 ml/min. Oven temperature was 90 ˚C (1 min), 90 ºC to 185 ˚C (at 35 ˚C/ min and hold time of 0 min), 185 ˚C to 220 ˚C (at 2 ˚C/min and hold time of 0 min), 220 ˚C to 280 ˚C (at 5 ˚C/min and hold time of 0 min), 280 ˚C to 320 ˚C (at 25 ˚C/min and hold time of 0 min). A volume of 1 µL, was injected in splitless mode at 250 ºC.

2.6 Identification, Quantification and Data Analysis

Reference standards ranging from 1 pg/ml to 1000 pg/ml were 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 was 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 analytes were also obtained in the same way. Descriptive statistics were carried out on the data on organochlorine pesticides and their concentrations are presented in graphs and boxplots showing the means, minimum and maximum values.

3. Results and Discussion

Eighteen compounds of organochlorines and their metabolites were identified in the water samples. They consist of alpha, beta, gamma and delta Hexachlorocyclo-hexane (HCH), endrin, aldrin, heptachlor, chlordane, b-endosulfan, methoxychlor, *p,p'*-DDE, *p,p'*-DDD, endrin eldehyde, endosulphan sulphate, *p,p'*-DD and endrin ketone. Their concentrations range from below detection limit to 1 µg/L (Figure 2). The least concentrations of organochlorines in water are for g-chlordane and α-HCH which are below 0.1 µg/L with a maximum of 0.3 µg/L. *β*-HCH has the highest concentration of the HCH's, and the alpha, beta, gamma and delta HCH were found in all the samples apart from 1 borehole.

When comparing the three water sources (rain, river, and groundwater), OC concentrations are slightly higher in rain and lower in groundwater (Figure 3). Of the six river samples, Malewa and Gilgil rivers have higher concentrations of OCs, while the organochlorines detected in groundwater are those applied in the agriculture industry as fungicides (the 5 isomers of HCH), and insecticides (aldrin, chlordane, endrin, heptachlor, dieldrin, endosulfan). Aldrin and dieldrin are restricted to termite control in the building industry (PCPB, 1998; 2010). Most frequently detected pesticides in the waters

are aldrin, endrin aldehyde, *p,p'*-DDE and endosulfan sulphate (Figure 3). These pesticides are commonly used in developing countries due to their low cost and effectiveness against various pests [35, 36]. Our findings show that there is no specific order of abundance of specific organochlorine group and its metabolites in one water source which could be related to differences in the date of application and half-lives that vary from days to several years as reported in [37]. Other studies have shown that insecticides and herbicides are common groundwater contaminants in the developing world [38, 39] as farmers strive to optimise crop yields and profits.

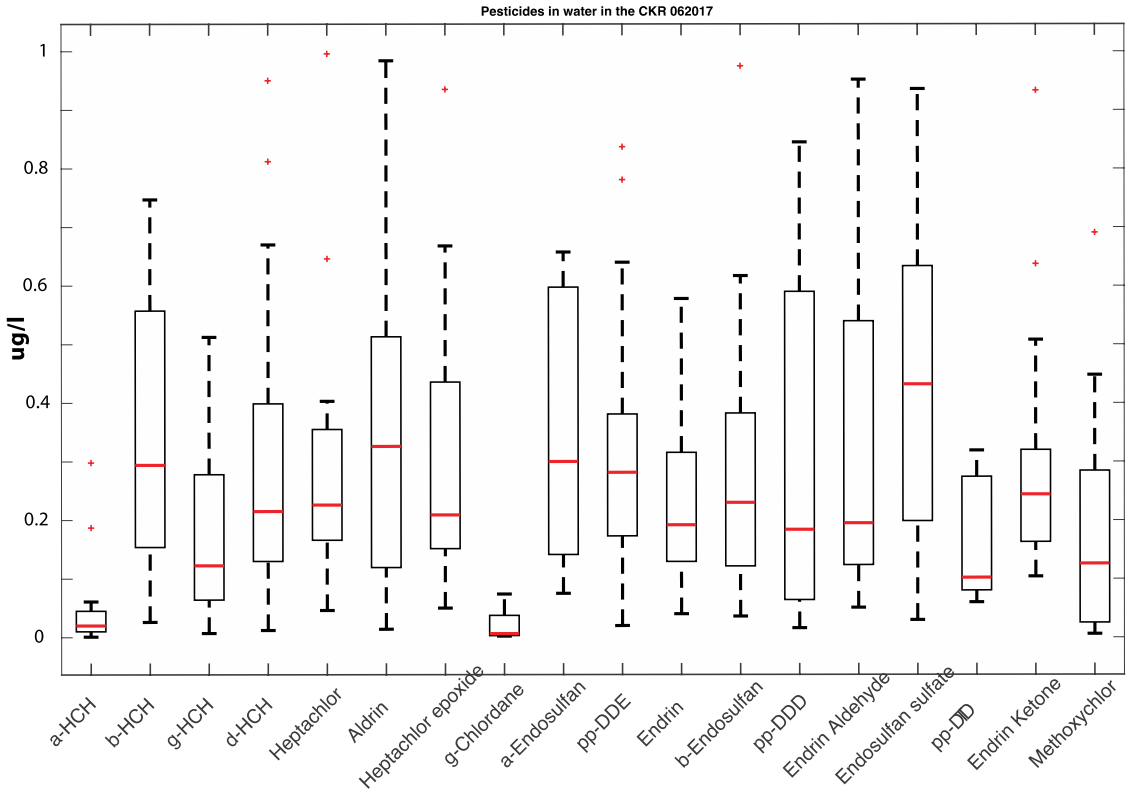

Figure 2: Box plots showing the total ranges of concentrations of pesticides and their metabolites (18) found in the water sources (rain, rivers and groundwater) in Lake Naivasha basin.

Twelve out of the eighteen organochlorine pesticides found in the water are listed amongst the 31 banned in Kenya by the Pest Control Products Board (Table 1). These organochlorines are used as insecticides and fungicides. They include chlordane, heptachlor, 5 isomers of hexachlorocyclo-hexane (HCH), endrin and DDT (Dichlorodiphenyl Trichloroethane) since 1986, while aldrin and dieldrin are banned since 2004, and endosulfan was banned in 2011. The by-products of lindane, α-Hexachlorocyclohexane (α-HCH) and *β*-HCH were detected in the water but not lindane which was banned in 2011 in Kenya. Dichlorodiphenyldichloroethane (DDD), a metabolite of DDT, and DDE, a product of dehydrohalogenation of DDT, were also found, but not DDT. However, DDT was banned for agriculture use in Kenya in 1986 but its use is restricted to indoor residual spraying only for mosquito control.

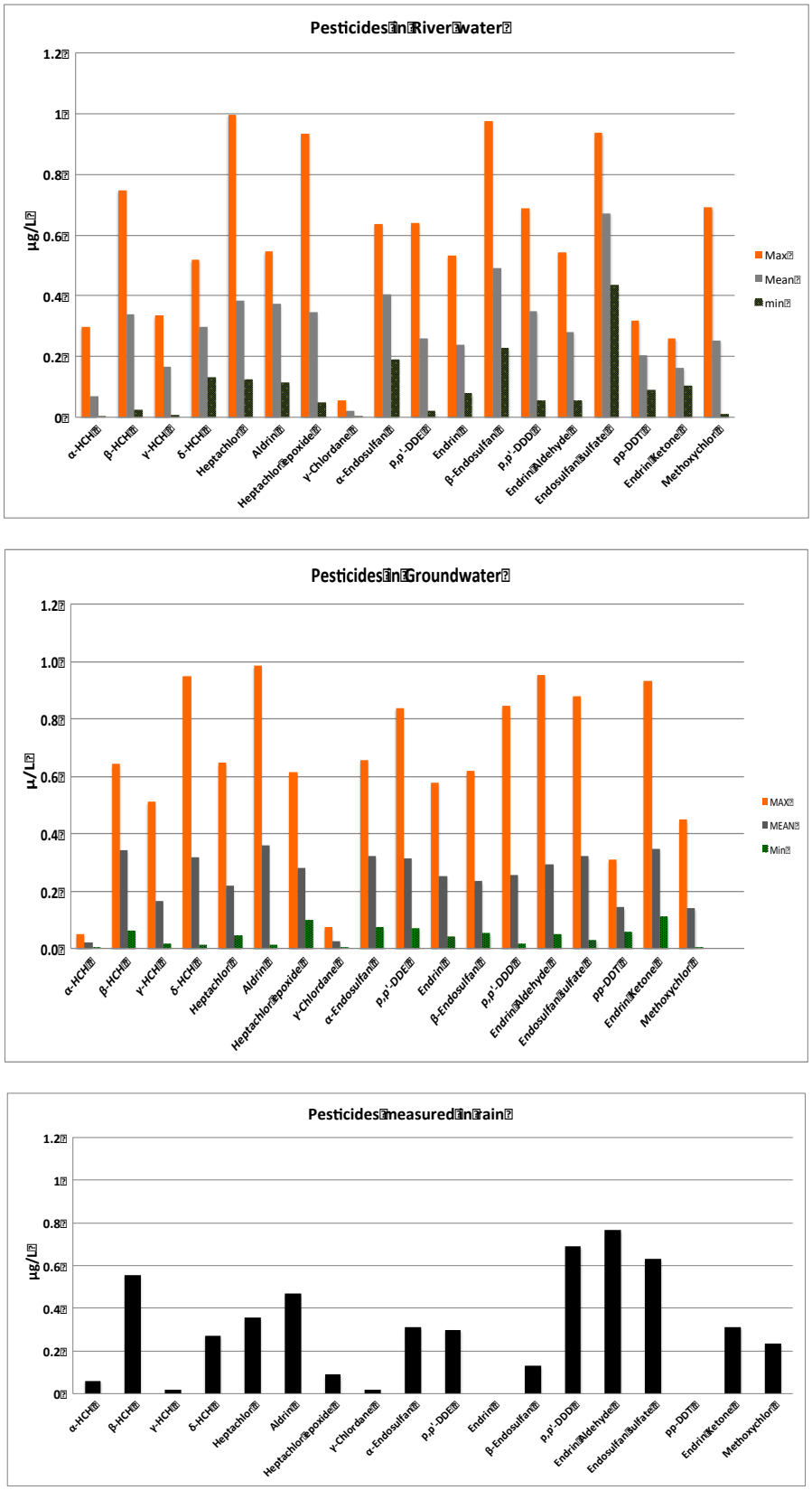

Figure 3: Concentrations of pesticides in the different water sources: river water (upper panel), groundwater (middle panel) and rainfall (lower panel).

The presence of banned OC metabolites several decades after their ban in Kenya shows that either they are persistent, or occasionally used elsewhere in the region /neighbouring countries and are dropped via the atmosphere. The persistence of OCs in tropical environments is shown to be lower with half-lives of 3-7 months for DDT [40, 41] unlike in temperate regions where they stay for decades [42, 43]. In general the concentrations detected in the waters are low and complementary studies by Gitahi and others [25], established that fewer number of organochlorines are used in the study area as compared to organophosphates, carbamates, and pyrethroids. However, the presence of banned compounds in rain water can be interpreted as ongoing usage that is potentially coupled with atmospheric transport and deposition, which would occur through volatilization of pesticides after application in the vicinity. This inference is supported by studies by Majewski and others [44] which showed that the occurrence and atmospheric concentration of pesticides were most closely related to their use within 40 km of the Mississippi River from New Orleans, Louisiana to St. Paul, Minnesota.

Table 1: List of pesticides banned in Kenya since 1986.

	Name	**Usage**	**Year of Ban**
1.	Chlordane,	Insecticide	1986
2.	Heptachlor	Insecticide	1986
3.	5 isomers of hexachlorocyclo-hexane (HCH)	Fungicide	1986
4.	Endrin	Insecticide	1986
5.	DDT (Dichlorodiphenyl Trichloroethane)	Disease vector control, banned for agriculture	1986
6.	Aldrin	Insecticide	2004
7.	Dieldrin	Insecticide	2004
8.	Endosulfan	Insecticide	2011
9.	Lindane	Insecticide	2011

Once applied, the temperature and low rainfall conditions in the rift floor can cause organochlorines to degrade naturally in soil through the mechanistic pathways of dechlorination, dehydrochlorination, isomerization and oxidation [45, 46], while transport of the compounds to groundwater and surface water occurs through processes of leaching and runoff, respectively. Once in the aquifer, their degradation is likely to be much lower than the half-lives reported for the near surface, particularly within the bedrock. This is because of limited microbial populations and activity [47, 48], combined with more limited nutrient and oxygen availability within groundwater [49]. Overall, groundwater is considerably less contaminated than surface water. Residues in river water samples occur when the pesticides are carried into the rivers either through the wind, runoff or through the adhered suspended particulate, while compounds that spread into the air are incorporated in rain droplets. It has been established that the application of pesticides in tropical countries characterized by high temperatures and heavy rainfall lead to a higher spread of contamination over watersheds than in the temperate regions [50].

Even though the concentrations measured are low, in the microgram per litre range, many of these pollutants can exceed an acceptable daily intake (ADI) for humans. A soil sample from the area had much higher concentrations (50 times [51]) leading the authors to speculate that the OCs quantities

will be higher in the animals grazing in affected areas or aquatic organisms and raptors (birds of prey). The exposure pathways to humans are through drinking contaminated water, milk, meat and crops derived from this area. Though these results are from a single sampling campaign in June 2017, these are the first findings of organochlorines in groundwater and rain water in the Naivasha basin where previous studies focused on surface water sources.

4. Conclusions

This paper set out to determine the status of organochlorine pesticides in the drinking water sources in the Naivasha basin, some of which are also sources for drinking water supplies. A total of 18 organochlorine compounds and their metabolites were measured in the water sources (rivers, rain and groundwater). The three organochlorines with the highest concentrations are aldrin, endrin aldehyde, endosulphan sulphate (between 0.7 and 0.9 µg/L), and these are used in the agriculture sector. The low concentrations compared to the WHO [52] guidelines on drinking water quality could be because of faster degradation of OCs in the tropics. However, though these concentrations are generally low, in the microgram per litre range, many of these OCs can exceed an acceptable daily intake (ADI) for humans. Their presence in rainwater, despite their ban several decades ago (1986 - 2014) shows either active use in Naivasha or atmospheric transport from the vicinity of study area. These findings are contrary to previous studies by [29] where OCs were not detected.

Findings from this study suggest that there is need for the state ministries responsible for sectors such as agriculture, health and water where OCs are used to collaborate to set up regular monitoring of use / sale of banned pesticides, air and water quality to keep the residents safe. Long term monitoring of seasonal changes in the compounds and concentrations of organochlorines in all the three water sources especially used for domestic purposes should be set up. Future research should set up long term monitoring of the various pesticides, organochlorines, organophosphates, carbamates and pyrethrins which are used much more frequently. Additional measures such as minimising the usage of OCs through seeking alternatives such as bio-pesticides and bio-control should be explored. The most urgent intervention by the National Environmental Management Authority is to develop drinking water quality guidelines for pesticides and their metabolites, which are not yet established in Kenya, to ensure reduced exposure to organochlorines and their metabolites. Additionally, the Pest Control Products Board in Kenya, the National Environment Management Authority and other relevant authorities need to regularly engage with the aim of ensuring harmful Pesticides are not sold in the country and protecting Kenyans from exposure to harmful compounds.

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