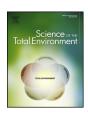
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Fish consumption from urban impoundments: What are the health risks associated with DDTs and other organochlorine pesticides in fish to township residents of a major inland city



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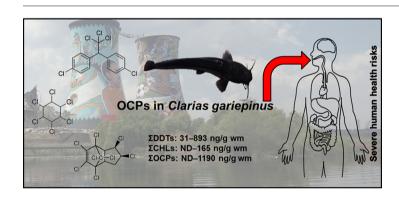
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HIGHLIGHTS

• Organochlorine pesticides (OCPs) found in urban area

- Clarias gariepinus accumulated OCPs in muscle tissue.
- The cancer risk associated with consumption was greater than acceptable levels.
- Non-cancerous risk was up to a 1000 times that what is considered safe.

GRAPHICAL ABSTRACT



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ABSTRACT

Organochlorine pesticides (OCPs) in South Africa have for the most part been banned, except dichlorodiphenyl-trichloroethane (DDT) which is still used as malaria vector control. The aim of this study was to determine OCP residues in the aquatic fauna of one of South Africa's most populated areas, Soweto. Risk to human health through OCP exposure via fish consumption was investigated. *Clarias gariepinus* was chosen as bioindicator because it is an apex predator that is in abundance, but is also a valued food source. Dichlorodiphenyltrichloroethanes (DDTs), hexachlorocyclohexanes (HCHs), and chlordanes (CHLs) were detected in the fish tissue with the DDTs being the most prevalent at all sites. Of the three locations, Fleurhof, Orlando, and Lenasia, the latter location's fish had the highest Σ OCP load, ranging between 81 and 1190 ng/g wm. The DDTs were determined to be from historic use, whereas the CHL levels indicated more recent inputs. Although the possibility of illegal use cannot be excluded completely, the presence of OCPs outside of their allowed areas of use indicate that these compounds not only stay in the aquatic systems long term, but may be of concern in areas previously not considered high risk areas. The OCP residues in *C. gariepinus* from the study area pose an extremely high risk to human health when consumed, and has a cancer risk as high as 1 in 10. This potential problem should be kept in consideration when developing national health and conservation strategies.

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1. Introduction

The unregulated use of organochlorine pesticides (OCPs) in South Africa was banned in 2002 as per the Stockholm Convention (Bouwman, 2004). However the ban on one of these, namely DDT (dichlorodiphenyltrichloroethane), was lifted for the use of malaria vector control (MVC) by indoor residual spraying (IRS), under strict legislation, overseen by provincial government. A detailed overview on malaria vector control in South Africa is described by Brooke et al. (2013). There have been an increasing number of reports on the accumulation of DDT and other organochlorine pesticides (OCPs) in the aquatic wildlife (Barnhoorn et al., 2009; Gerber et al., 2016; Viljoen et al., 2016). Aquatic ecosystems are especially at risk as most OCPs are more stable in sediments, therefore increasing its persistence (Doong et al., 2002; Yohannes et al., 2013a). OCPs are transferred from the sediment via different mechanisms into aquatic organisms and magnify up the food chain (Van der Oost et al., 2003). High bioaccumulation of these OCPs have been reported in aquatic predators at the top of the food chain, such as the tigerfish (Hydrocynus vittatus Castelnau, 1861) (McHugh et al., 2011; Wepener et al., 2012; Gerber et al., 2016) and sharptooth catfish (Clarias gariepinus (Burchell, 1822)) (Barnhoorn et al., 2009, 2015). These aforementioned studies were all in subtropical areas of South Africa, where spraying of DDT for MVC is still allowed and results showed very high concentrations of not only DDTs, but also various other banned OCPs, such as chlordane and HCHs, in the tissues of these species. The study areas were of a rural nature and included important conservation areas and their surroundings with low human population densities. In the present study we determined the levels of OCPs in aquatic organisms in ecosystems within surrounding densely populated areas of South Africa. The demographic includes many unemployed people that rely on subsistence fishing to supplement their diet.

The study area is in the urban area of Soweto (South Western Townships), one of South Africa's most densely populated areas (6400 people/km²) (StatsSA, 2011). Soweto is found south west of the city of Johannesburg and is South Africa's largest township with a high unemployment rate with some people reliant on subsistence fishing as a means of obtaining food. There is no direct spraying of OCPs, particularly DDT in this area. However, the fact that long range air transport of OCPs is well documented (Gong et al., 2015; Pozo et al., 2006), it could potentially result in the exposure of this region to these persistent pollutants. The study area is also relatively close to sources where OCPs were manufactured in the past. The production of DDT and other OCPs in Gauteng occurred in the Wadeville and Chloorkop areas, and in the neighbouring North West province, at the town of Brits (DEA, 2011). These locations are approximately 35 (east), 40 (north east) and 75 km (north) from the selected study area, respectively. Manufacturing of various OCPs, such as lindane, heptachlor, aldrin, dieldrin, endrin and DDT was discontinued in South Africa in the 1980s and the formulation of DDT in 2010 (DEA, 2011). In addition, a study of the middle Vaal River, into which our study area drains, reported surprisingly high levels of OCPs in fish species (Wepener et al., 2011).

Clarias gariepinus was chosen as an indicator species because it is abundant in South Africa, it is resilient, it is the apex aquatic predator in the sampling area, and most importantly, is a valued food source (Rouhani and Britz, 2011). They are targeted by local fishermen not only in rural areas but are specifically targeted by fisherman in the study area (personal communication: W Pheiffer). Their position on the food web and preference for bottom dwelling in the aquatic systems makes it ideal to study exposure to pollutants, as well as the bioaccumulation and bio-magnification of organic chemical pollutants, allowing for investigation into possible transfer of pollutants to humans.

The aim of this study was to determine DDT and other OCP residues in *C. gariepinus* muscle tissue from impoundments in the urban area of Soweto, South Africa post the cessation of use and formulation. These findings are compared to other African studies on *C. gariepinus* or relevant apex predators. This paper most importantly investigates the

potential risk posed to health of the local human population through consumption of OCP contaminated *C. gariepinus*.

2. Materials and methods

2.1. Study area and sampling

Clarias gariepinus was sampled during Austral summer (October) of 2013 from three impoundments in the upper Klip River catchment (Fig. 1) in Soweto and Lenasia. The Klip River drains the Witwatersrand area in the Gauteng province of South Africa. It is the largest tributary of the Vaal River. These rivers together supply >12 million people of potable water in Gauteng (DWAS, 2004). Fleurhof Dam (26°12′03.49"S 27°54′31.87″E) is the most northern site, representing the upper region of the study area. Orlando Dam (26°15′21.63″S 27°55′18.97″E) is located in the centre of Soweto, and Lenasia Dam (26°18'8.33"S 27°50' 10.8"E) is located in the southern stretches of the Klip River. The fish were caught using fyke and gill nets (110 mm mesh). Nets were left in for 5 h and checked periodically every half hour. Fish weighing less than 1 kg were released. Following capture, fish were euthanized by severing the spinal cord behind the head (NWU Ethics approval: AREC-130913-015). The mass (g) and standard length (SL; mm) of each fish were recorded. Epaxial muscle samples were removed, placed in pre-cleaned (acetone/hexane) aluminium foil, and kept frozen at — 20 °C. Epaxial muscle was selected for consistency in sampling between specimens and because the epaxial muscle is the part of the fish primarily consumed by humans. For comparative purposes a group of ten fish from the Mooi River system in the North West province (part of different catchment as the study area) were depurated for half a year at the Water Research Group's aquarium at the North-West University (Potchefstroom Campus). Fish were kept under standard aquarium conditions (CCAC, 2005). These reference fish were processed in the same manner as the experimental fish.

2.2. Organochlorine pesticide determination

2.2.1. Materials

An OCP standard reference mixture (all isomers of chlordane and nonachlor, oxychlordane (collectively CHLs); all isomers of DDT, DDD and DDE (collectively DDTs; group referred to as DDx when DDT is excluded); dieldrin, endrin and aldrin (collectively drins); hexachlorobenzene (HCB); hexachlorocyclohexanes (α -, β -, γ -, δ -isomers, collectively HCHs); and all isomers of heptachlor and heptachlor-epoxide (collectively heptachlors)) were obtained from Dr. Ehrenstorfer GmbH (Germany). Analytical grade organic solvents (acetone, hexane, and dichloromethane), and anhydrous sodium sulphate (pesticide residue and PCB analysis grade) were obtained from Kanto Chemical Corp. (Tokyo, Japan). Florisil (60–100 mesh) from Kanto Chemical Corp. (Tokyo, Japan) was activated in an oven at 180 °C for 8 h.

2.2.2. Extraction of OCPs in muscle

Analysis of OCPs in muscle samples followed an adapted version of the method by Yohannes et al. (2013b). Sample (10 g wet mass; wm) was homogenized with anhydrous sodium sulphate and placed in precleaned (acetone/hexane) cellulose extraction thimbles. Samples were spiked with PCB#77 as internal standard. Extraction was carried out with 150 mL hexane:acetone (3:1 v/v) for 6 h using a Soxtherm (S306AK Automatic Extractor, Gerhardt, Germany). Extracts were concentrated to approximately 2 mL using a rotary evaporator and diluted to 10 mL with hexane. Lipid content was determined gravimetrically using an air dried 10% aliquot of the extract. The remaining extract after concentrated to 5 mL was passed through gel permeation chromatography columns (Waka Gel Bio-Rad) with dichloromethane (DCM): hexane (1:1 v/v) as mobile phase, collecting only the fraction containing the OCPs. The fraction collected was concentrated and cleaned-up on a glass column packed with 6 g 5% activated Florisil, and eluted with

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