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Bioaccumulation and human health risk assessment of DDT and other organochlorine pesticides in an apex aquatic predator from a premier conservation area

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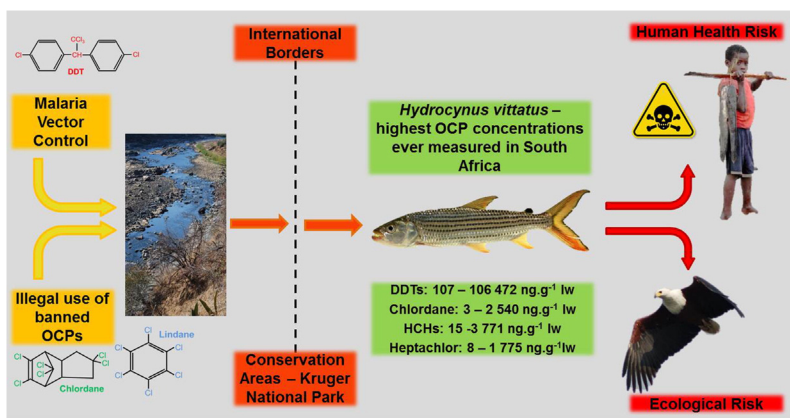
HIGHLIGHTS

- First record of OCPs in top predator tigerfish from rivers in Kruger National Park
- Pesticides used for vector control bioaccumulate at very high levels in fish from conservation areas
- Both rivers flowing through the Kruger National Park are polluted by OCPs originating from the catchment.
- OCP bioaccumulation is the highest reported for Africa.
- Significant human cancer risk is associated with the consumption of fish from these rivers.

GRAPHICAL ABSTRACT



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ABSTRACT

With the second highest gross domestic product in Africa, South Africa is known to have a high pesticide usage, including the highly persistent and banned group of organochlorine pesticides (OCPs). South Africa is also one of few countries to still actively spray DDT as malaria vector control. The aim of the study was to determine the degree to which aquatic biota in selected rivers of the world renowned Kruger National Park (KNP) are exposed to by use of OCPs in the catchments outside the KNP and how this exposure relates to human health. Tigerfish (*Hydrocynus vittatus*) are economically important apex predators and was selected as bioindicator for this study. Fish were sampled from the KNP sections of the Luvuvhu, Letaba and Olifants rivers during the high and low flow periods from 2010 to 2011 within the KNP and 19 OCPs were determined in muscle tissue using GC-ECD techniques. Significant flow related and spatial OCP bioaccumulation was observed. Tigerfish from the Luvuvhu River displayed the highest OCP bioaccumulation. Concentrations of the majority of the OCPs including the DDTs were the highest levels ever recorded from South African freshwater systems and in many cases the concentrations were higher than most contaminated areas from around the world. The concentrations found in *H. vittatus* muscle also exceeded maximum residue levels in edible fat as set by the European Union. The health

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Multivariate analysis
Tigerfish

risk assessment also demonstrated that the levels of OCPs pose very high cancer risks to the local populations consuming tigerfish, as high as 2 in 10 increased risk factor. This is of concern not only when managing the water resources of the conservation area but also for surrounding communities consuming freshwater fish. Contaminants enter the park from outside the borders and pose potential risks to the mandated conservation of aquatic biota within the KNP.

1. Introduction

South Africa has the second highest gross domestic product in Africa and is also one of the top four pesticide importers in Africa (Quinn et al., 2011). These pesticides include the highly persistent and lipophilic group of hydrocarbon compounds known as organochlorine pesticides (OCPs) which are used extensively in industry as well as agriculture throughout the world (Dallaire et al., 2013). Environmental contaminants such as OCPs are known to be ubiquitous anthropogenic pollutants in aquatic ecosystems (Sarkar et al., 2008) and because of their high persistence and the fact that they are highly lipophilic, OCPs are known to bioaccumulate in the fatty tissues of aquatic organisms (Dallaire et al., 2013), specifically those of fish. Organochlorine pesticides are known to have deleterious effects on aquatic ecosystems, and as such monitoring the bioaccumulation of these pollutants is essential for the assessment of potential impacts (Wepener et al., 2011; Yohannes et al., 2013a, b).

Tigerfish (*Hydrocynus vittatus*) were selected as the indicator organism as they are an important freshwater fish species due to their high economic value and role in sustaining the livelihoods of many communities (Smit et al., 2009). They are abundant and widely distributed within the sampling area, easy to sample and target, occupy the top position in the aquatic food chain (McHugh et al., 2011) and relatively long lived (up to 20 years, Gerber et al., 2009; Soekoe et al., 2013). As apex predators they have been shown to accumulate pollutants, which include DDTs, various heavy metals and organohalogens (Bouwman et al., 1990; Du Preez and Steyn, 1992; Mhlanga, 2000; Wepener et al., 2012).

Notwithstanding the high volume of pesticide use in South Africa (Dabrowski et al., 2014), there is limited reporting of bioaccumulation data. By virtue of its position on the western border of South Africa, the rivers that flow through the Kruger National Park (KNP) transport pollutants from the industrial and agricultural heartland (Gerber et al., 2015a) into this premier conservation area. Various studies have shown that rainfall and therefore seasonal runoff has a very strong influence on the water and sediment quality of rivers in the KNP (Wepener et al., 2000; Gerber et al., 2015a). The previous assessment of OCPs in the KNP was by Heath and Claassen (1999), who recorded low concentrations of various OCPs in a number of fish species sampled from the various rivers during the period 1992 to 1993. Recent studies in upstream reaches of the Olifants and Luvuvhu rivers adjacent to the KNP have, however, highlighted that concentrations of a number of OCPs in both abiotic (Gerber et al., 2015b) and biotic (Barnhoorn et al., 2009, 2010) compartments are at concerning levels. This is of concern to conservation managers but also to local communities bordering the KNP and its rivers. Inland water bodies throughout central and southern Africa are considered to be important sources of artisanal fisheries (Andrew et al., 2000) with rural communities relying on rivers where large inland lakes are not available (Coetzee et al., 2015). The threats of OCPs especially DDTs to human health in the region have been highlighted in other studies outside the KNP (Barnhoorn et al., 2009; Bornman et al., 2010). The consumption of potentially contaminated freshwater fish could be a primary source of human exposure to OCPs in the area.

The aim of this study was to evaluate the distribution and current levels of various OCPs, including DDTs in *H. vittatus* tissue from selected rivers of the KNP by describing the temporal changes in OCP bioaccumulation related to abiotic concentrations during different flow periods in the rivers. This paper further investigates the potential risks posed to the human health of local rural communities by OCPs and provides a baseline for managers to make informed decisions and thereby take effective measures aimed at mitigating the potential ecological and health risks posed by the OCPs.

2. Materials and methods

2.1. Study area and sampling

Sampling of tigerfish was undertaken between May 2010 to June 2011 at sampling sites along the Olifants, Letaba and Luvuvhu rivers within the KNP (Fig. 1). The Letaba River site was included for comparative purposes to assess whether the Letaba River may act as a refuge for the large *H. vittatus* population found at Olifants River Gorge in the Olifants River. Tigerfish were sampled in the Olifants River Gorge S23° 59′ 25.2″ E31° 49′ 33.3″ located at the confluence of the Olifants and Letaba rivers, the Letaba River site (S23° 56′ 32.9″ E31° 43′ 53.5″) located in the Letaba River before its confluence with the Olifants River. The Luvuvhu River site (S22° 27′ 04.3″ E31° 04′ 47.7″) is located downstream of the confluence of the Mutale and Luvuvhu rivers and before the confluence of the Luvuvhu and the Limpopo rivers (Fig. 1).

Tigerfish were sampled using standard angling techniques. Following capture, fish were sacrificed by severing the spinal cord just behind the head. Fish were weighed (g) and measured (mm) for standard length (SL) and axial muscle sample was removed, placed in aluminium and frozen at −20 °C until further analysis. Tigerfish were sampled at the end of alternating low (LF) and high flow (HF) periods between 2010 and 2011. During these surveys the top 5- to 10 cm of inundated surface sediments were collected using an Eckman grab sampler for OCP analysis. The uppermost sediments were collected as it is considered that these sediments have been recently deposited are closely associated with the water column and therefore pose the most direct significant risk to the aquatic biota at the selected sites. For detailed sampling and treatment methods refer to Gerber et al. (2015b).

2.2. OCP bioaccumulation

2.2.1. Materials

Pesticide grade organic solvents (diethyl ether, acetone and *n*-hexane), anhydrous sodium sulphate and florisil (60–100 mesh) were obtained from Kanto Chemical Corp. (Tokyo, Japan). The florisil was activated in an oven at 130 °C for 12 h. A standard mixture (DDTs, HCHs, chlordanes, drins, heptachlors and hexachlorobenzene (HCB) at 10 μg mL^{-1} was purchased from Dr. Ehrenstorfer GmbH, Germany.

2.2.2. Analysis of OCPs

2.2.2.1. Muscle tissue. Muscle tissue analyses were done following the method of Yohannes et al. (2013a). Acetone/hexane pre-washed extraction thimbles were filled with 10 g of fish fillet which was homogenised together with anhydrous sodium sulphate. The sample was extracted for 6 h with 150 mL mixture of hexane:acetone (3:1 *v*/*v*) in a Soxtherm apparatus (S306AK Automatic Extractor, Gerhardt, Germany). The extract was concentrated to approximately 2 mL using a rotary vacuum evaporator, and diluted to 10 mL using hexane. Gravimetric lipid determination was done using an aliquot of 20% of the extract and the remaining 80% was subjected to a clean-up process after more of the