Comparative exposomics of ~~P~~persistent ~~O~~organic ~~P~~pollutants (PCBs, OCPs, MCCPs and SCCPs) and ~~P~~polycyclic ~~A~~aromatic ~~H~~hydrocarbons (PAHs) in Lake Victoria (Africa) and Three Gorges Reservoir (China)

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Editor: Adrian Covaci

Abstract

Exposomics is assessment of organism exposure to high priority environmental pollutants in an ecosystem using OMIC technologies. A virtual organism (VO) is an artificial property-tool (OMIC) reflecting exposomic process in compartments of real organisms. The exposomics of aquatic organisms inhabiting Lake Victoria (L.V.) and Three Gorges Reservoir (TGR) were compared using VOs. The two reservoirs are heavily depended on for food and water both in Africa and China. The target priority pollutants in the reservoirs were polyclic aromatic hydrocarbons (PAHs) and persistent organic pollutants such as polychlorinated biphenyls (PCBs), organochlorine pesticides (OCPs), medium chain chlorinated paraffins (MCCPs) and short chain chlorinated paraffins (SCCPs). The VOs showed that in a period of 28 days, aquatic organisms in TGR were exposed to total (∑) PAHs of

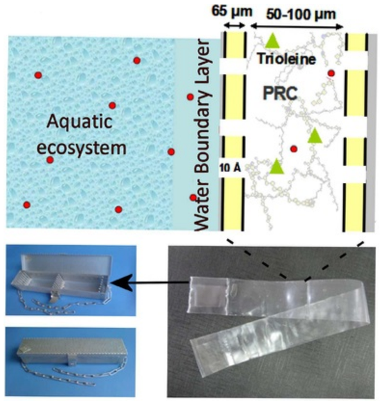
8.71×10^{-6} mg/L, PCBs of 2.81×10^{-6} mg/L, OCPs of 2.80 ~~x~~ × 10^{-6} mg/L, MCCPs of 8.9×10^{-10} mg/L and SCCPs of 1.13×10^{-7} mg/L. While in a period of 48 days, organisms in L. V. were exposed to total (∑) PAHs of 7.45×10^{-6} mg/L, PCBs of 4.70×10^{-6} mg/L, OCPs of 3.39×10^{-8} mg/L, MCCPs of 4.6×10^{-10} mg/L and SCCPs of 3.6×10^{-9} mg/L. The exposomic levels in TGR after 28 days were higher than those in Lake Victoria after 48 days. In both reservoirs, bioaccumulation levels are above set standards for aquatic organisms. The sources of the pollutants into the reservoirs were diagnostically determined to originate from anthropogenic processes such as petrogenic, diesel emissions, biomass burning, coal combustion, electronic wastes, traffic emissions and historic uses.

Keywords: Exposomics; Virtual ~~O~~organism; Lake Victoria; Three Gorges Reservoir; Persistent organic pollutants; Polycyclic ~~A~~aromatic ~~H~~hydrocarbons

~~1.~~1 Introduction

EXPOsOMICs is the study of high priority environmental pollutants exposure to an organism in an ecosystem using ‘OMIC’ technologies such as proteins (proteomics) to monitor the pollution status (Vineis et al., 2017). The ‘OMIC’ technology that uses a virtual organism (VO) as an artificial property-tool to reflect exposomic processes in compartments (fat, protein, and membranes) of real organisms has been previously reported (Karacık et al., 2013b).

A VO is a passive sampler whose principal working mechanisms were developed by Huckins et al. (1999, 2002a). This theory is used to determine the amount of freely dissolved nonpolar organic chemicals in water (bioavailable) for in situ trace level monitoring. The target compounds are accumulated in the VOs from their freely dissolved form in water. The transient cross section of the polymeric membrane of the VOs, with maximal diameters of about 10 Å, does not restrict passage of the compounds under study (see Fig. 1). Transport mechanism of the compounds from water phase to the VO is limited either by diffusion under water boundary layer (WBL) control or by the resistance in the membrane of the VO. The WBL is a thin hydro-dynamically complex region area between the VO membrane and the water body from the bulk water. Under membrane control, a VO acts as a permeate sampler, while under WBL, it acts as a diffusion sampler. More details about the working mechanisms of VOs are reported in our recent publications (Temoka et al., 2016; Temoka et al., 2017).



~~Fig. 1.~~**Fig. 1** Virtual Organisms for the lipidome are designed from a lay-flat PE semi-permeable membrane tubing enclosing a thin film of natural-like triolein with a high molecular weight (MW. 885 g/~~mole~~mol) natural lipid. Stable isotopes labeled PRCs are used to calculate the volume sampled by the VO. Seven VO are placed in stainless steel container which are then deployed in an aquatic ecosystem.

alt-text: Fig. 1

Booij et al. included performance reference compounds (PRCs) to compensate for environmental factors that can affect uptake kinetics of SPMDs (Booij et al., 1998; Booij et al., 2007). Thus, a high turbulence leads to release of more PRC and therefore a higher sampling rate. PRCs are analytically non-interfering compounds. They are absent in the environment and are spiked into passive samplers during their production (Booij et al., 2007; Huckins et al., 2002b; Huckins et al., 2002c). The PRC-method relies on isotropic exchange so that the uptake of the target analyte into the sorbent phase is presumably synchronized with the dissipation of preloaded PRCs out of the sorbent phase. Thus, PRC loss rate data can be used to adjust VO-derived estimates of ambient concentrations to reflect site-specific environmental conditions of an exposure (Exposomics). Furthermore, persistent organic pollutants (POPs) uptake kinetics of triolein VOs is similar to that of rainbow trout fish when monitored in similar environments (Lu and Wang, 2003). In this study, a VO is similar to a semi-permeable membrane device (SPMD) deferring only in the amount of triolein used (mass fraction of 20% triolein of a standard SPMD is replaced with a mass fraction of 44% which helps to gain more pollutant accumulation and sensitivity) (Temoka et al., 2017).

Polycyclic aromatic hydrocarbons (PAHs) and persistent organic pollutants (POPs) such as polychlorinated biphenyls (PCBs), organochlorine compounds (OCPs), medium chain chlorinated paraffins (MCCPs) and short chain chlorinated paraffins (SCCPs) have been classified as high priority environmental pollutants by the world health organization (WHO, 2019). These chemicals are of global concern due to their potential for long-range transport, persistence in the environment, ability to bio-accumulate and bio-magnify in ecosystems, as well as their significant negative effects on human health and the environment. Human exposure to POPs and PAHs (even to low levels) may cause increased cancer risk, reproductive disorders, alteration of the immune system, neurobehavioural impairment, endocrine disruption, genotoxicity and increased birth defects (WHO, 2019). Reservoirs are great sources of food to humans hence their contamination by PAHs and POPs poses a risk to human health. The need to monitor reservoirs for POPs and PAHs is necessary in order to develop policies in their management.

The exposomics of Three Gorges Reservoir (TGR) on Yangtze River in Asia have been determined (Table S1) and organisms living in this aquatic environment found to be at a risk (Wang et al., 2009a). TGR has a maximum water depth of 175 m (average of 90 m), total water capacity of 393 billion m^3, total coverage area of 1084 km^2, total length of 670 km, and average width of 1.1 km (Li et al., 2018) (Fig. S1). This very big manmade reservoir can only be compared to Lake Victoria (L.V.) in Africa via comparable matrices like water because the biota is not similar and thus not comparable. VOs are then a good alternative for both biota and pure water (Wang et al., 2015). Continuous monitoring of TGR ecological health is important as the dam can affect the over 300 fish species of Yangtze River, prevent upstream spawning of fish species, and disrupt natural processes of aeration hence affect chemical composition of the dam and the river.

Lake Victoria (Fig. S2) is the second largest freshwater lake in the world. It has a surface area of 68,800 km^2 and a maximum depth of 79 m (average of 40 m). The lake has several shorelines with numerous islands, bays, channels and wetlands. For the past five decades, the lake has undergone dramatic liminological transformations and fish declines as well as changes in trophic status, stratification pattern, water temperature, seasonality, and fish biodiversity (Natugonza et al., 2016; Omwoma et al., 2014). The lake has several inlets but only one outlet (White Nile) which starts its journey to the Mediterranean Sea from the source of the Nile in Jinja District, Uganda near the Nalubaale hydropower station (originally known as Owen Falls Dam). The White Nile, flows through Uganda, South Sudan, Sudan, Egypt and into the Mediterranean Sea.

This is the first time Exposomics studies are being carried out in Lake Victoria and as such, the sampling sites were chosen at the discharge point of the lake, Nalubaale hydropower station (Nalubaale dam). The purpose of this sampling site was to allow reflection of the exposomic levels in the White Nile. Previously, active sampling in Lake Victoria had indicated that its water, sediments and biota are contaminated with POPs as shown in Table S1. However, there is no data indicating the amount of these POPs that are freely dissolved in Lake Victoria waters leading to their mobility into other aquatic systems. Furthermore, analysis of sediment and fish samples from River Nile in Egypt shows contamination by the mentioned POPs (El-Kady et al., 2007; Magdy, 2017). The sources of the detected POPs in Egypt have not been determined. The current research determines if the POPs detected in L. V. could be a point source of contamination to Nile River through the White Nile. Nile River majorly comprises of the White Nile from L. V. and Blue Nile from Ethiopia.

Though the exposomics of pollutants in Lake Victoria was yet to be determined, the priority pollutants had been detected in different compartments within the aquatic environment (Table S1). Since the two systems are highly depended upon by human population for food, we determined and compared the current exposomics levels of the two systems. In this study, we have adopted the PRC-method that is well explained by Temoka et al. (2017). The priority pollutants under the present study include the PAHs, PCBs, OCPs, MCCPs and SCCPs.

~~2.~~2 Materials and methods

~~2.1.~~2.1 Sampling

VOs were deployed on 23rd February 2016 at Nalubaale power station in L.V. (latitude: 0.44, longitude: 33.18) and were collected on 13th March~~,~~ 2016. The second sampling period was between 16th October 2017 and 3rd December~~,~~ 2017. The sampling sites are as shown in Fig. S1 and the cages are as shown in Fig. S3. At every site, four cages were deployed and within each cage, 5 VOs were inserted. The cages at TGR were installed in a similar manner on 23rd June~~,~~ 2016 and collected on 21st July~~,~~ 2016 at Zigui county (latitude: 30.92, Longitude: 110.74). The sampling days in TGR were derived from Wang et al. (Wang et al., 2009a) who had indicated that SPMDs could equilibrate with exposomic chemicals after 24 days. However, in L.V. this information was yet to be determined hence the sampling period of less or ~~more than~~ ≥24 days was considered (19 days and 48 days). Furthermore, the summer season was chosen as the sampling period in TGR due to climatic conditions of L.V. which lies on the equator with negligible seasonal changes.

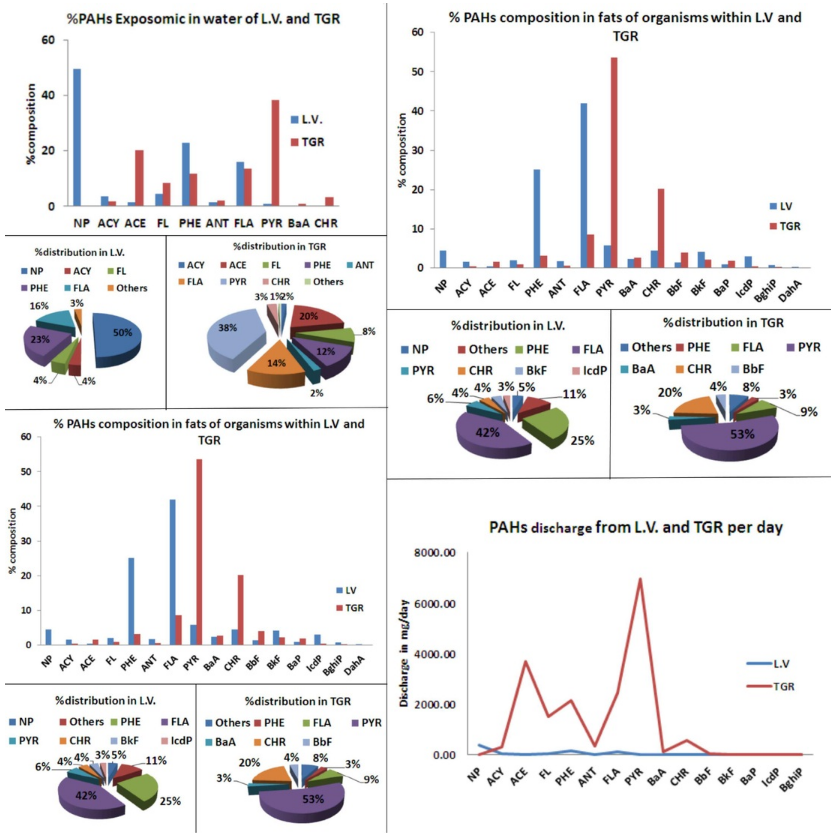
~~2.2.~~2.2 Sample preparation and analysis

In this study, SPMD was replaced with VOs since the mass fraction of 20% triolein of a standard SPMD was replaced with a mass fraction of 44% which helps to gain more pollutant accumulation and sensitivity previously demonstrated in former studies (Temoka et al., 2017). Information on VO preparation, extraction, clean-up and analysis have been previously reported (Karacık et al., 2013a; Wang et al., 2009a; Wang et al., 2013). However, we describe briefly this information (*VO preparation, extraction, clean-up and analysis and quality assurance/control*) in supporting files. The flow rates of the sampling sites on TGR (24,088 m^3/s) and L.V. (1201 m^3/s) were used in determining the exposomic levels in the respective reservoirs. Detailed information on how exposomic levels were calculated has been previously published (Temoka et al., 2017). Freight amounts/levels were determined as the total amount of the chemical being transferred from the reservoir per sampling period.

~~3.~~3 Results and discussion

~~3.1.~~3.1 Comparative exposomics of PAHs in TGR and L.V~~.~~

The PAH amounts of aquatic organisms exposure in both L.V and TGR are shown in Table S2 and it is analyses in Fig. 2. The levels of PAHs (7.4×10^{-6} mg/L in L.V. and 8.7×10^{-6} mg/L in TGR~~)~~) that are exposable (bioavailable) to aquatic organisms are within drinking water standards for humans (2.0×10^{-4} mg/L) (WHO, 2003a). Although PAHs have an elaborate metabolism by aquatic organisms (Abdel-Shafy and Mansour, 2016), such levels can be teratogenic with low body weights and birth defects can occur (Abdel-Shafy and Mansour, 2016). Bioaccumulation of the PAHs in adipose tissues of organisms is possible also as evidenced by VOs fat accumulations of 236.11 ng/g after 48 days in L.V. and 1~~,~~142.93 ng/g after 28 days in TGR (Table S2). For instance, fish samples from Lake Victoria (Alice et al., 2012) and TGR (Floehr et al., 2015) have been reported to bio-accumulate the PAHs in their adipose tissues. In addition, the present study reveals the ability of the reported PAHs to be suspended (exposable) in waters leaving L.V. hence their ability to be a point source to the Nile River waters.



~~Fig. 2.~~Fig. 2 Comparison and distribution of PAHs within Lake Victoria and TGR (sampling in L.V. = 48, TGR = 28 days).

alt-text: Fig. 2

WHO Standards (WHO, 2003b) states that PAHs are known to be carcinogenic and the concentrations of the six representative PAH compounds (fluoranthene, 3,4~~-~~-benzfluoranthene, 11,12~~-~~-benzfluoranthene, 3,4~~-~~-benzpyrene, 1,12~~-~~-benzopyrene and indeno [1,2,3~~-~~-cd] pyrene) should not, exceed 2.0×10^{-4} mg/L. The current study reveals a value (2.37×10^{-4} mg/L) that slightly exceeds this limit. In addition, bioaccumulation of pollutants by aquatic organisms is of concern especially for organisms higher in the food webs such as human beings. Although our calculated water concentrations values of ~~less than~~ <1 ng/L are below EU and USEPA standard levels (Table S2), the accumulated levels in the lipid (ng/g) of the VOs of 3~~-~~–230 ng/g are way above the standard values set by the EU in food and feeds (Table S2).

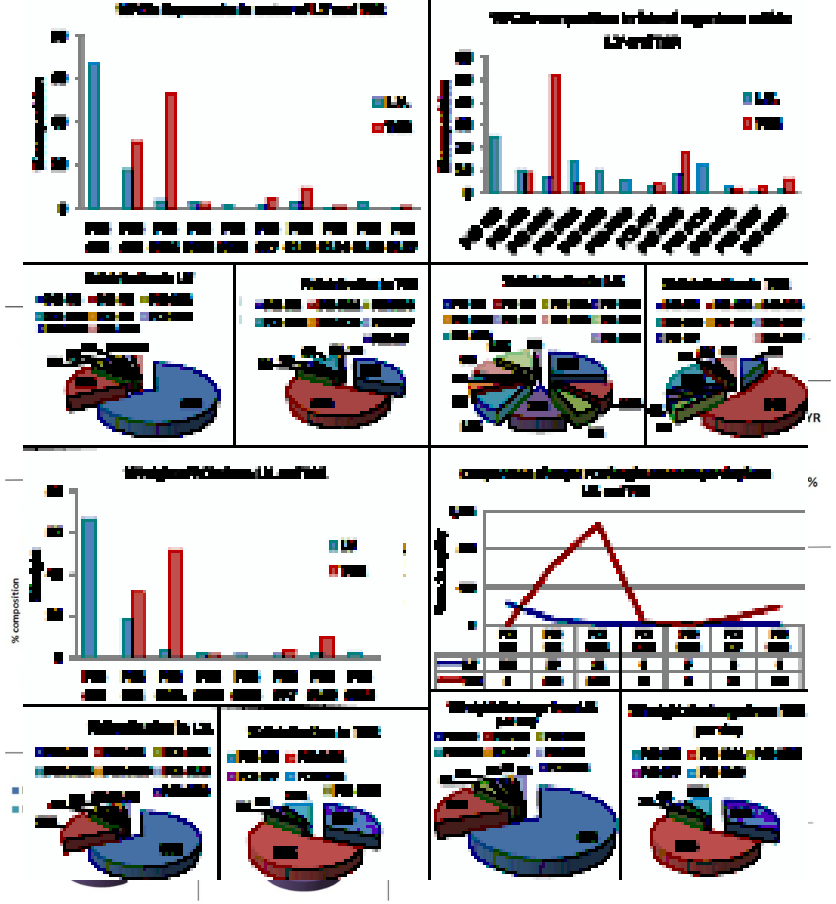
Some of the natural sources of PAHs include volcanic eruptions, biomass burning and diagenesis (Wang et al., 2007). Anthropogenic sources include coal and wood burning, petrol and diesel oil combustion, industrial processes and liquid fuel spills (da Silva and Bícego, 2010; Mostert et al., 2010). The release of PAH mixtures to the environment has a correlation to the molecular concentration ratios which are characteristic of a given emission source (Tobiszewski and Namiesnik, 2012). As such, PAH diagnostic ratios provide an important tool for identification of pollution emission sources. The emission profile for a given source normally depends on processes producing the PAHs (Manoli et al., 2004). At low temperatures such as during wood burning, PAHs with a low molecular weight will be formed. During combustion of fuels by engines which exhibits high temperatures, higher molecular weights PAHs are released (Mostert et al., 2010) and organic compounds are cracked into reactive radicals that further react to form stable PAHs (pyrosynthesis). The resultant PAHs from the pyrosynthesis process are less alkylated with more aromatic rings (Hwang et al., 2003). It has been demonstrated that PAH diagnostic ratios may clearly show intra-source variability but inter-source similarity (Galarneau, 2008). In L.V., PAHs could be originating from petrogenic, petrol emissions, pyrogenic, traffic emissions, combustion of grass, wood and coal processes whereas in the TGR, the PAHs could be from pyrogenic, petrol emissions, petrogenic, fossil fuel combustion, petrogenic, petroleum combustion and/or traffic emissions (Table S3).

Petrogenic sources in L.V. reservoir could be attributed to inefficient engines in automobiles and/or spillages in petrol stations. For instance, in Kisumu town on the shores of L.V., runoff water from all petrol stations is deposited into the reservoir directly (Omwoma et al., 2015). Furthermore, inefficient exhaust in most automobiles could lead to pyrogenic PAHs sources. Use of firewood in homesteads around the reservoirs especially in their catchments also contributes to PAHs sources (Lisouza et al., 2011).

It is noted that the total PAH exposure levels reported in this study for TGR (8.71 ng/L) are similar to previously reported values in the same reservoir using SPMDs (Wang et al., 2009b; Wang et al., 2013). For L.V., the generated data could be used as baseline values for subsequent monitoring studies of bioavailable PAHs. PAHs Exposomics, fat concentration and their subsequent discharge from TGR is higher than in L.V. These differences are demonstrated in Fig~~ure~~. 2.

~~3.2.~~3.2 Comparative Exposomics of PCBs in TGR and L.V~~.~~

The indicator PCBs (#28, #52, #101, #138, #153 and #180) that originate from sources like industrial chemicals (Omwoma et al., 2015), were yet to equilibrate with the sampling VOs in the short first sampling campaign (19 days) in L.V. This resulted to values below our limit of quantification (LOQ) (Table S4). However, an increase in the sampling period from 19 days to 48 days resulted in detectable exposure levels to VOs in L.V. (Table S4). In TGR, PCBs #28, #118, #153, and #180 were below the LOQ. All the mono-ortho PCBs (#105, #118, #123, #156, #157, #167, and #189) and non-ortho PCBs (#77, #81, #126, and #169) with similar toxicity patterns as dioxins were found to be bioavailable in L.V. (Table S4). However, in TGR, mono-ortho PCBs, #118 and #123 had values below the LOQ whereas only PCB #77 was detectable for non-ortho PCBs. In general, most PCB values reported in TGR were below the LOQ. However, high values recorded for PCBs #52, #101 and #105 especially the freight amounts makes the total PCBs/period in TGR to be higher than in L.V. (Table S4 and Fig. 3). Since these PCBs are considered as highly toxic to aquatic organisms and may be correlated to cause obesity (CHEMWATCH, 2010; Ferrante et al., 2014) the high amounts demonstrated in TGR should be a major concern. The value reported for dioxin-like PCBs in TGR of 0.14 ng/L (summation of dioxin like PCBs in Table S4) is within the same range of data as that previously determined using SPMDs at Zigui county in China (0.08~~–~~–0.51 ng/L) (Wang et al., 2009a).



~~Fig. 3.~~Fig. 3 Comparison and distribution of PCBs within Lake Victoria and TGR (sampling in L.V. = 48, TGR = 28 days).

alt-text: Fig. 3

Although the reported total PCB values for aquatic exposure (4.70×10^{-6} mg/L for L.V. and 2.81×10^{-6} mg/L for TGR) are below standard values that can affect aquatic life (3.0×10^{-3} mg/L) (NEMA, 2006), bioaccumulation that is evidenced by high concentrations in fat content of the VOs (L.V. = 1.51×10^{-5} and TGR = 5.68 ~~x~~ × 10^{-5} mg/g) are of major concern. The high exposure levels of PCB #28 in L.V. results in a higher total PCBs level in L.V. than TGR (Table S4 and Fig. 3). Furthermore, it can be deduced from the results that although the other congeners are in high amounts in TGR, their exposure levels are very low (Table S4 and Fig. 3). As a result, the freight amounts from TGR are higher than those of L.V. (Table S4 and Fig. 3).

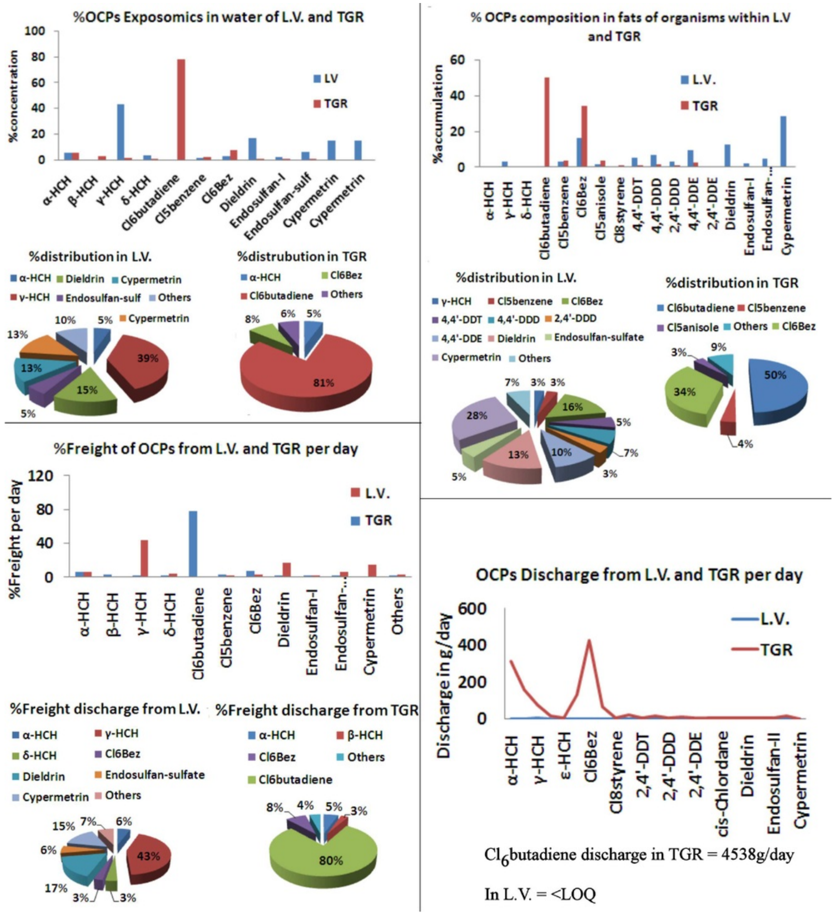
Environmental conditions of a reservoir such as temperature, pH, dissolved oxygen, and water flow rates determine bioavailability of PCBs (Alkhatib and Weigand, 2002; Daniel and Ronald, 2005; Sergio et al., 1997). Therefore, the differences in temperature (L.V. = 24~~o~~°C and TGR = 23~~o~~°C), pH (L.V = 6.56 and TGR = 8.2) and water flow rates (L.V. = 1201 m^3/s TGR 24088 m^3/s) significantly affected bioavailability of the reported PCBs. As such, despite TGR freight levels being higher than in L.V., the latter records higher water and fat concentrations. Freight levels are the total amount of the chemical being transferred from the reservoir per sampling period.

Dechlorination of PCB congeners (e.g. from PCB #66 to PCB #28, PCB #92 to PCB #52 and PCB #141 to PCB #101; Table S4) is an indication of their historical uses (USEPA, 2007). PCB dechlorination does not remove PCBs; however, it only alters the composition of the PCB congeners. Dechlorination transforms the PCB into forms that are susceptible to mineralization processes. It should be noted that some old transformers and other equipment that contained PCBs are still in use today, and therefore they may release PCBs to the environment. Furthermore, contaminated upland sites and runoff from many industrial environments continue to deposit PCBs to aquatic systems. PCBs were also used in caulk, paint, sealants, gasket materials, and numerous other industrial applications, a few of which did not use Aroclor formulations but relied on individual PCB congeners or non-Aroclor mixtures (Magar et al., 2005; USEPA, 2007). Because of their stable and persistent nature, historic PCB contamination can still be widely found. Therefore, there is a high probability that the detected PCBs are from historic uses in both reservoirs. Electronic waste that includes old transformers, capacitors, voltage regulators, switches, re-closers, bushings, electromagnets, fluorescent light ballasts, cable insulation, oil used in motors and hydraulic systems could all be contributing to historic uses.

The diagnostic ratio of PCB #28/PCB #153 has previously been used to determine the sources of PCBs contamination (USEPA, 2007). In general, values above 0.3 indicate strong contamination from landfills and surface runoffs (Magar et al., 2005). In this study, the calculated ratio of PCB #28/PCB #153 for L.V. was 2.5 while for TGR; both congeners were below quantification level. This indicates that the main source of PCBs into Lake Victoria aquatic system could be through landfills where there is deposition of electric wastes. It should be noted that there is no plant that manufactures PCBs in Africa. Nevertheless, there is a widespread use of PCB-containing transformers and other PCB-containing devices. Apart from transformers, importation of electric waste to Africa is a major source of exposure as the imported products are finally dumped in various landfilling dumpsites (UNEP, 2009b). Recently, several unmanaged landfills (30 in Kisumu, 22 in Homabay and 14 in Mbita) were reported to drain waste directly into Lake Victoria (Omwoma et al., 2015); Kisumu, Homabay and Mbita are towns on the Kenya side of the lake.

~~3.3.~~3.3 Comparative Exposomics of OCPs in TGR and L.V~~.~~

During the first sampling campaign of 19 days in L.V, the VOs recorded high concentrations of hexachlorobenzene (0.79 pg/L in water, 0.42 ng/g in fat and a freight release into White Nile of 1.42 g/day) (Table S5). However, after 48 days of sampling, the VOs had equilibrated with this pollutant at concentration of 0.95 pg/L in water, 0.50 ng/g in fat and the outlet freight of 4.73 g/day. In TGR, the bioavailability of hexachlorobenzene was several times higher as reported 205.86 pg/L in water, 108.72 ng/g in fat and a freight of 11,996.34 g/day. Generally, TGR recorded higher OCPs levels than L.V. (see Fig. 4 for analysis).



~~Fig. 4.~~Fig. 4 Comparison and distribution of OCPs within Lake Victoria and TGR (sampling in L.V. = 48, TGR = 28 days).

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Hexachlorobenzene is banned globally under the Stockholm Convention on Persistent Organic Pollutants (UNEP, 2014). It is a former organochlorine fungicide but also a product of industrial processes and incineration activities. It has been cited as a probable carcinogen to living organisms (UNEP, 2009a). The high presence of this chemical and its ability to flow out of both reservoirs requires an immediate investigation on its use in the catchments of the two reservoirs. Before it~~'~~'s ban in the USA, the chemical was widely used as a seed-treatment fungicide for onions, sorghum, wheat, and other grains (ATSDR, 2000). It was also used as a chemical intermediate in dye manufacture and synthesis of other organic chemicals, in the production of pyrotechnic compositions for the military, as a raw material for synthetic rubber, a plasticizer for polyvinyl chloride, and as a wood preservative (ATSDR, 2000). Hexachlorobenzene can also bioaccumulate in fat and biomagnify in tissues of living organisms that are high in the food chain (ATSDR, 2000). However, it is possible that the compound could have found its way into the reservoirs from atmospheric deposition as recently demonstrated on Shergyla Mountain, southeastern Tibetan Plateau (Zhu Nali et al., 2014).

Dichlorodiphenyltrichloroethane (DDT) metabolites such as 4,4′~~-~~-Dichlorodiphenyldichloroethane (4,4′~~-~~-DDD), 2,4′~~-~~-Dichlorodiphenyldichloroethane (2,4′~~-~~-DDD) and 4,4′~~-~~-Dichlorodiphenyldichloroethene (4,4′~~-~~-DDE) were present in significant amounts in waters flowing into the White Nile and in TGR (Table S5). This could be attributed to the use of DDT as a pesticide to control malaria mosquitoes in areas surrounding the Lake Victoria Basin. We determined current use of DDT in the riparian area of the two reservoirs using the ratio of (DDE + DDD) / DDTs. Recent application has been reported to give values <0.5 while past application gives values >0.5 (Doong et al., 2002; Wang et al., 2015). The value of this ratio was 0.508 for L.V. in the second sampling campaign of our study and 0.89 for TGR suggesting historical usage of DDT where technical DDT gradually degraded to its stable metabolite DDE (Willett et al., 1998). Furthermore, the ratio of 2,4′~~-~~-DDT/4,4′~~-~~-DDT with a value of 0.2 for L.V. and 0.25 for TGR indicates that the source of DDT could be of industrial origin rather than from fresh applications of DDT-containing dicofol (Qiu et al., 2005)~~.~~. Though China stopped importation and use of DDT in 2005, it continues to produce and use dicofol using DDT (Andrew, 2009; Qiu et al., 2005). This could probably be the source of fresh applications envisaged in our findings. Furthermore, the continued use of DDT in India (Davé, 2016) could be imported to China by the monsoon winds. It is noted that DDT has been detected in air over Taihu Lake in China (Qiu et al., 2005).

Gamma-Hexachlorocyclohexane (lindane) and the other isomers of hexachlorocyclohexane (HCH) showed high concentrations in the second sampling campaign that took 48 days in L.V. (Table S5). The inability to detect lindane in the first sampling campaign for L.V. could be attributed to the insufficient time to accumulate amounts beyond its LOQ. Therefore, detectable values were recorded when the sampling period was extended in the second campaign.

In the present study, the total OCP exposure levels (2.79 ng/L) in TGR are in the same range of data as those reported recently by the use of SPMDs (2.33~~–~~–3.60 ng/L) (Wang et al., 2009b). However, it was noted that all the reported values for OCPs were within levels tolerable levels set for aquatic life (Table S5).

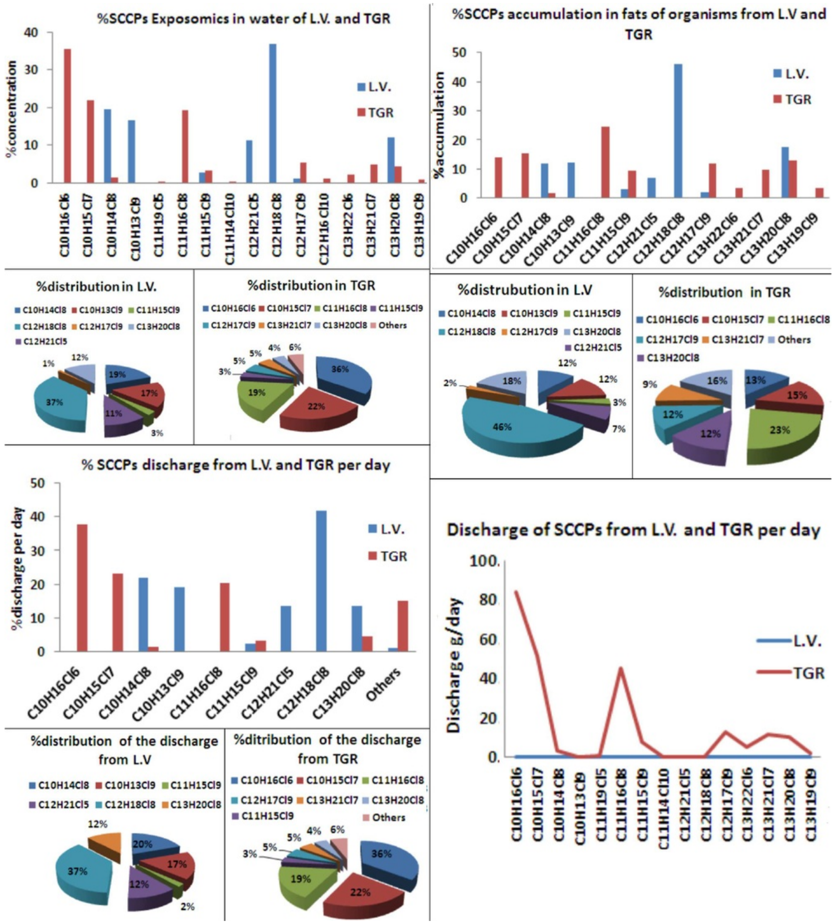
Among the congeners of HCH, β-HCH has the lowest water solubility, vapor pressure and, is the most stable thus resistant to microbial degradation (Zhou et al., 2007). As such, this congener was below our LOQ in our study for both sampling campaigns in L.V. Nevertheless, we used the relative abundances of α-HCH and γ-HCH (10% and 75%, respectively for L.V.) to determine whether the observed concentrations of HCH mixtures originate from historical applications (Buser and Muller, 1995). TGR had these abundances at 5.4% and 1.3%, respectively, indicating that their presence may not be entirely from historical applications. Historical applications is also supported by the ratios of β-HCH/(α-HCH + γ-HCH) with a very small value since β-HCH concentrations in water were below our limit of quantification. Fresh applications would have the value of β-HCH/(α-HCH + γ-HCH) higher than 0.5. Otherwise, lower values than 0.5 indicate HCH aging (Zhang et al., 2007). Although TGR has a value of 0.41 for the above ratios, which is closer to the value of fresh applications, there is need to investigate further the source of lindane in this catchment. Normally, the ratios of α-HCH/γ-HCH in technical mixtures of HCH range from 3.0 to 7.0 and zero for lindane. Therefore, the ratio value of α-HCH/γ-HCH of 0.13 for L.V. suggests that the source of the detected pollutants was from both technical mixture of HCH and lindane (Zhu et al., 2015). Lindane is widely used as an agricultural insecticide and as a pharmaceutical in the treatment of lice and scabies (USEPA, 2006).

Since endosulfan II converts to endosulfan I over time, our results indicate that the source of the observed endosulfan I (0.68 ng/L) was from historical applications as endosulfan II concentrations were below the LOQ value for L.V. This is particularly the case as an industrial mixture of endosulfan would contain a ratio of 2~~-~~–3 for endosulfan I/endosulfan II (Estellano et al., 2008). However, the ratio of 1.8 for TGR may also indicate fresh applications of endosulfan I in the catchments of this reservoir.

~~3.4.~~3.4 Comparative exposomics of MCCPs and SCCPs in TGR and L.V~~.~~

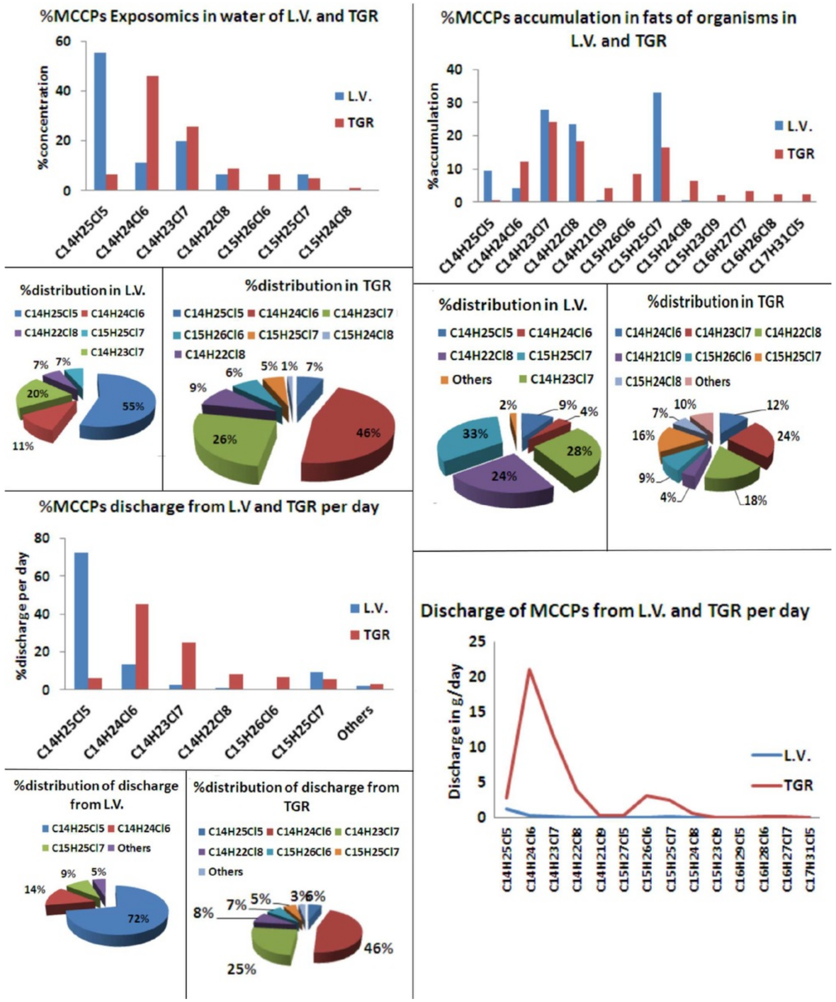
Chlorinated paraffins are industrial chemicals that can be subdivided into short-chain (SCCP), medium-chain (MCCP) and long-chain (LCCP) (EPA, 2014; Gluge et al., 2018). Three major MCCPs ($C_{14}H_{24}Cl_6$, $C_{14}H_{23}Cl_7$, and $C_{14}H_{22}Cl_8$) and SCCPs ($C_{10}H_{16}Cl_6$, $C_{10}H_{15}Cl_7$ and $C_{11}H_{16}Cl_6$) were detected in significant amounts (Tables S6 and S7). The high amounts of the compounds can be attributed to the use of these chemicals in plastics (mainly PVC), rubbers, paint and sealants, where they act as plasticizers with flame retardant properties (EPA, 2014). Further to this, the chemicals can also be found in metal cutting fluids, where the chlorinated paraffins act as lubricants which prevent sliding metal surfaces from seizing under conditions of extreme pressure.

Chlorinated paraffins can affect aquatic invertebrates (in particular *Daphnia magna*) which appear to be a sensitive group in terms of toxicity. Concentrations that have been experimentally determined for no observed effect on *Daphnia magna* include 0.005 mg/L for SCCPs and 0.010 mg/L for MCCPs (EPA, 2014). Values recorded in this study for L.V. and TGR were 4.5×10^{-10} and 8.0×10^{-10} mg/L for ∑MCCPs, and 3.6×10^{-9} and 1.13×10^{-7} mg/L for ∑SCCPs, respectively (Tables S6 and S7). The values of the pollutants are way below the aforementioned standards hence they present no major acute threat to aquatic organisms within L. V. and TGR. However, the levels in TGR are higher than those in L.V. (Figs. 5 and 6).



~~Fig. 5.~~Fig. 5 Comparison and distribution of SCCPs within Lake Victoria and TGR (sampling in L.V. = 48, TGR = 28 days).

alt-text: Fig. 5

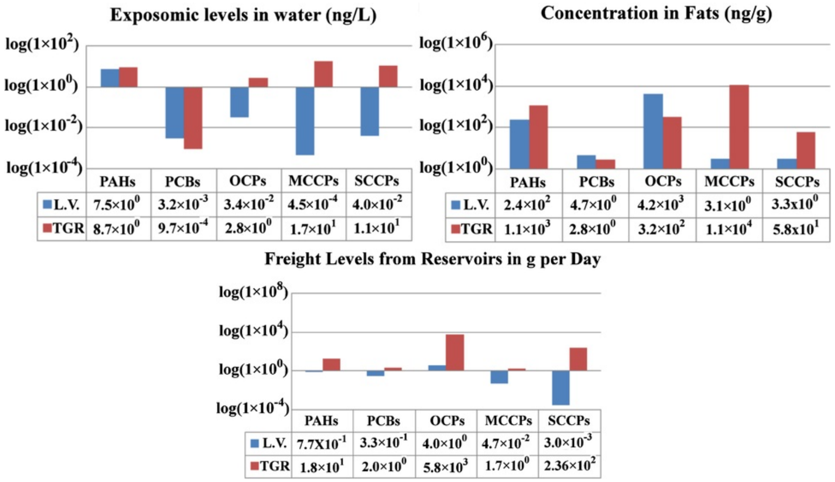


~~Fig. 6.~~Fig. 6 Comparison and distribution of MCCPs within Lake Victoria and TGR (sampling in L.V. = 48, TGR = 28 days).

alt-text: Fig. 6

~~3.5.~~3.5 3.5. General comparative exposomics of TGR and L. V.

The level of exposure by aquatic animals to priority pollutants such as PAHs, OCPs, MCCPs and SCCPs is higher in TGR than in L.V. (Fig. 7). Consequently, the accumulated amounts in fat tissues by aquatic animals could be higher in TGR than in L.V. as indicated by the recorded values in VOs. However, different environmental conditions in the two reservoirs make PCBs more bioavailable to aquatic animals in L.V. than in TGR (Fig. 7). The phenomenon of environmental effect on bioavailability of chemicals is measured by performance reference compounds (PRCs) factored in VOs (Temoka et al., 2017). The rate at which PRCs diffuse from VOs into water is used to determine mathematically the environmental effect on POPs and PAHs accumulation by organisms. As such, organisms exposed to chemicals in an environment with a high PRCs leaching ability are less likely to accumulate the chemicals. For instance, in this study, although results for TGR show that the system has high content of PCBs exhibited by high levels of freight amounts, L.V. shows higher exposure levels combined with high fat accumulation levels (Fig. 7). It is also noted that differences in other environmental factors such as temperature (L.V. = 24 ~~o~~°C and TGR = 23~~o~~°C), and pH (L.V = 6.56 and TGR = 8.2) might have contributed significantly to the observed differences (Alkhatib and Weigand, 2002; Daniel and Ronald, 2005; Sergio et al., 1997).



~~Fig. 7.~~Fig. 7 Comparative exposomics of sums PAHs, PCBs, OCPs, MCCPs and SCCPs in Lake Victoria and Three Gorges Reservoir

alt-text: Fig. 7

~~4.~~4 Conclusions

The level of exposure by aquatic organisms to priority pollutants such as PCBs, PAHs, OCPs, MCCPs and SCCPs is high in the Three Gorges Reservoir (TGR) (China) than in Lake Victoria (L.V.) (Africa). In both reservoirs, bioaccumulation of the priority pollutants is higher than the set standards (WHO, EU and USEPA) for aquatic reservoirs. The main source of PCBs into L.V. was identified as landfills deposited with electric wastes while TGR could be majorly from Historic uses. OCPs in L.V. are identified to originate from both current and historical uses while in TGR, there is strong evidence of current uses in the catchment. The high amounts of MCCPs are attributed to the use of these chemicals in plastics, rubbers, paint and sealants in both reservoirs. Environmental conditions of a reservoir can change the bioavailability of priority pollutants. In this study, despite TGR having higher freight levels of PCBs than L.V., the lower concentrations in L.V. are more bioavailable than the high levels of TGR. Banning the use of DDT should include it is use in the manufacture of other pesticides. The fresh applications of DDT diagnosed in TGR could be due to the use of dicofol, a pesticide manufactured using DDT. Atmospheric depositions can affect reservoirs far away from point of pesticide application. The cancerous hexachlorobenzene detected in this study could be a result of atmospheric deposition as there is no evidence of its use in the catchments of the reservoirs. Electronic waste should be sorted out and processed professionally instead of dumping it into landfills. These wastes could be responsible for most of the chemicals detected in the reservoirs. Runoff water from towns bordering reservoirs should be treated before deposition. This water is a major source of reservoir contamination especially in Africa.

Acknowledgement

We thank the management of the Nalubaale Power station for accepting us to deploy samplers at the dam. Silke Bernhöft from Helmholtz Zentrum Muenchen is appreciated for extraction and laboratory clean-up of samples. The reviewers of this manuscript are highly appreciated for their critical contribution. The AvH group linkage program titled: Ecosystem Services: Lake Victoria versus Three Gorges Reservoir, An Institutional Partnership project between German, Chinese and Kenyan Institutions, is highly appreciated for funding. National Research Fund, Kenya (2016/2017 FY) is highly appreciated for partial funding.

~~Appendix A.~~Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2019.133789.

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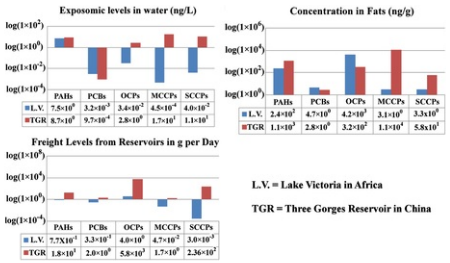
~~Appendix A.~~Appendix A. Supplementary data

Multimedia Component 1

Supplementary material

alt-text: Image 1

Graphical abstract



alt-text: Unlabelled Image

Highlights

- Exposomics is assessment of organism exposure to pollutants using OMIC technology.
- Virtual organisms were used for comparative exposomics in two Reservoirs.
- Levels in Three Gorges Reservoir were higher than those in L. Victoria.
- In both reservoirs, fat bioaccumulations are above set standards for aquatic life.

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Have we correctly interpreted the following funding source(s) and country names you cited in your article: "AvH; National Research Fund, Kenya".

Answer: Yes

Query:

Supplementary caption was not provided. Please check the suggested data if appropriate, and correct if necessary.

Answer: it is correct

Query:

Please provide the volume number and page range for the bibliography in Li et al., 2018.

Answer: The article is in press and the doi is: https://doi.org/10.1016/j.ecohyd.2018.08.005