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Biomagnification of persistent organic pollutants along a high-altitude aquatic food chain in the Tibetan Plateau: Processes and mechanisms☆

Jiao Ren [a, c], Xiaoping Wang [a, b, *], Chuanfei Wang [a, b], Ping Gong [a, b], Xiruo Wang [d], Tandong Yao [a, b]

[a] Key Laboratory of Tibetan Environment Changes and Land Surface Processes, Institute of Tibetan Plateau Research, Chinese Academy of Sciences, Beijing 100101, China
[b] CAS Center for Excellence in Tibetan Plateau Earth Sciences, Beijing 100101, China
[c] University of Chinese Academy of Sciences, Beijing 100049, China
[d] School of Environment, Guangzhou Key Laboratory of Environmental Exposure and Health, Guangdong Key Laboratory of Environmental Pollution and Health, Jinan University, Guangzhou 510632, China

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ABSTRACT

Biomagnification of some persistent organic pollutants (POPs) has been found in marine and freshwater food chains; however, due to the relatively short food chains in high-altitude alpine lakes, whether trophic transfer would result in the biomagnification of POPs is not clear. The transfer of various POPs, including organochlorine pesticides and polychlorinated biphenyls (PCBs), along the aquatic food chain in Nam Co Lake (4700 m), in the central Tibetan Plateau, was studied. The POPs levels in the water, sediment and biota [plankton, invertebrates and fish (*Gymnocypris namensis*)] of Nam Co were generally low, with concentrations comparable to those reported for the remote Arctic. The composition profiles of POPs in the fish were different from that in the water, but similar to their food. DDEs, DDDs, PCB 138, 153 and 180 displayed significant positive correlations with trophic levels, with trophic magnification factors (TMFs) ranged between 1.5 and 4.2, implying these chemicals can undergo final biomagnification along food chain. A fugacity-based dynamic bioaccumulation model was applied to the fish with localized parameters, by which the simulated concentrations were comparable to the measured data. Modeling results showed that most compounds underwent net gill loss and net gut uptake; only when the net result of the combined gut and gill fluxes would be positive, bioaccumulation could eventually occur. The net accumulation flux increased with fish age, which was caused by the continuous increase of gut uptake by aged fish. Due to the oligotrophic condition, efficient food absorption is likely the key factor that influences the gut POPs uptake. Long residence times with half-lives up to two decades were found for the higher chlorinated PCBs in *Gymnocypris namensis*.

1. Introduction

Persistent organic pollutants (POPs), such as organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs), are ubiquitous in the environment (Pozo et al., 2006; Koblizkova et al., 2012). The levels of POPs are higher than expected in remote regions, which are attributed to "cold trapping" effects (Carrera et al., 2002; Wania and Mackay, 1993). In cold environments, including polar regions and high-altitude mountains, low temperature enhances the accumulation of POPs in both terrestrial and aquatic ecosystems (Cropp et al., 2011; Muller et al., 2011). Biomagnification of both legacy and emerging POPs along food webs in the Arctic has been found (Hoekstra et al., 2003; Kelly et al., 2009). Then, by consuming the contaminated fish, human health can be affected; this has been shown in Inuit infants (Dallaire et al., 2003). Similar to the Arctic, there is evidence of the accumulation of various POPs in penguins and fish in the Antarctic (Cipro et al., 2013; Corsolini et al., 2003).

Previous studies have also reported high levels of POPs in biota

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* Corresponding author. Key Laboratory of Tibetan Environment Changes and Land Surface Processes, Institute of Tibetan Plateau Research, Chinese Academy of Sciences, Beijing 100101, China.
E-mail address: wangxp@itpcas.ac.cn (X. Wang).

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from remote alpine lakes (Demers et al., 2007; Gallego et al., 2007). On the one hand, mountainous regions are often found in the vicinity of industrialized and densely populated regions (Kallenborn, 2006). On the other hand, due to the extremely cold mountain climate, the cold-trapping effect is also evident on the altitudinal scale (Wania and Westgate, 2008). Dichlorodiphenyltrichloroethane (DDT) and higher chlorinated PCBs concentrations in fish from European mountainous lakes increased by 2–3 orders of magnitude along an altitudinal transect, from 400 to 2800 m (Grimalt et al., 2001). "Cold trapping" effect was taken as the cause of this accumulation. Additionally, dietary pattern, growth rate, age and lipid content of fish are all suggested as the important factors that influence the bioaccumulation of fish (Campbell et al., 2011; Demers et al., 2007; Vives et al., 2004); however, the key factors that truly control the bioaccumulation of fish from high altitude lake are still largely uncertain.

Apart from this knowledge gap, whether the biomagnification of POPs occurs along the food chains in alpine lakes is also unclear (Wania and Westgate, 2008). Studies from the Canadian Rocky Mountains found that the trophic levels of fish only explained a small amount of the variation in POPs (Campbell et al., 2011). Similar results were observed for fish from lakes in the French Pyrenees; their POPs contents (after lipid-normalized) did not correlate with the trophic levels of the fish (Blais et al., 2006). These studies indicated that the relatively short food chains in alpine lakes and the lower trophic levels of fish resulted in weak or no biomagnification of POPs. However, in contrast, Villa et al. (2011) found evidence of DDT and PCBs biomagnification in biotic samples from a sub-alpine Lake in Italy (Lake Como). The confliction involved in previous studies demonstrated that more researches should be conducted regarding the food web biomagnification of POPs in high mountain lakes.

The Tibetan Plateau (TP), with an average elevation above 4000 m, has the highest and largest high-altitude lakes in the world. Recently, the highest levels of mercury (Hg) contamination in wild fish of China were found in the Tibetan lakes (Zhang et al., 2014). Although high Hg concentrations in the wild fish were observed, due to the relatively short food chain the enrichment of Hg through the food chain was negligible (Zhang et al., 2014). Apart from Hg, many kinds of POPs, such as OCPs, PCBs (Yang et al., 2010), PBDEs (Yang et al., 2011), hexabromocyclododecanes (HBCDs) (Zhu et al., 2013) and perfluorinated compounds (Shi et al., 2010), have also been detected in fish from Tibetan lakes. However, it is unknown whether biomagnification and trophic transfer of these POPs occur in the aquatic ecosystems of TP. Tibetan high-altitude lakes are typically oligotrophic, and have a high pH and alkalinity, which is different from many European and Arctic lakes. In the unique environment hundreds of endemic fish species breed (Wu and Wu, 1992). The organisms in lakes of the TP typically have relatively low growth and metabolic rates, which further complicate the transfer dynamics and extend the residence time of POPs along the food chains.

In this study, we investigated the biomagnification of legacy POPs (OCPs and PCBs) along a typical high-altitude aquatic food chain from Nam Co Lake, located in the central of the TP. After developing a food chain framework and assigning biota to their trophic levels, we determined the concentrations of OCPs and PCBs in both the environment medium (lake water and sediment) and food web (plankton, invertebrates and fish). We verified whether biomagnification along the food chain occurred and quantified the extent of trophic transfer of each pollutant; and applied a fugacity-based bioaccumulation model in the Tibetan fish species (naked carp, *Gymnocypris namensis*) to clarify the mechanisms involving in the biomagnification.

2. Materials and methods

2.1. Site description and sampling

Nam Co Lake (30°30′–30°56′N, 90°16′–91°01′E) is the third largest lake on the TP, with an area of 2015 km^2 and elevation of 4718 m; it is situated to the north of the Nyainqentanglha Mountains [see Fig. S1 in the Supplementary Material (S)]. Due to the high altitude, the lake environment is severely cold, with an annual average temperature of 0 °C. The lake is ice-covered for more than 4 months each year (December to April). Previous surveys found Chlorophyll *a* levels of 0.46 μg L^{-1} in the water of Nam Co, which demonstrates the low primary productivity and oligotrophic conditions (Liu et al., 2010). There are few species of aquatic biota and the ecosystem structure is relatively simple in the lake (Xu and Kang, 2010).

An integrated sampling campaign was carried out in July 2014. In total, lake water (n = 15), sediment (n = 12), plankton (n = 14), invertebrate (n = 23) and fish (n = 38) samples were collected. An overview of the species collected is given in Table S1. Lake water (0–1 m depth, 200 L) and surface sediment (0–10 cm) were sampled at sites that were more than 10 km away from the shore, to avoid local contamination. Water samples were filtered with a glass fiber filter (GFF 0.7 μm, Whatman), to get the total suspended particulate matter (SPM), and then pumped through an XAD-2 resin column, to collect the dissolved phase compounds. A plankton trawl was used to sample the phytoplankton (10–100 μm) and zooplankton (>100 μm). Different classes of invertebrates, including aquatic insects, shrimp and snails (including *Gyraulus* and *Radix*) were collected. To consider the top predators in the food chain, different sizes of *Gymnocypris namensis*, the dominant fish species in Nam Co, were sampled (Table S2). Fish age was estimated using the von Bertalanffy growth function (Text S1) (Bertalanffy, 1938). All samples were transported to the laboratory under ice cold conditions and then stored at −20 °C until extraction.

2.2. Extraction and analysis for POPs

About 25 g wet weight (ww) of fish muscle was used for extraction. For plankton and invertebrates, individuals of the same species were pooled and 5 g ww of tissue was used for extraction. The extraction and cleanup methods are presented in Text S2 for each sample type. Briefly, the samples were Soxhlet extracted and then one tenth of each extract was used to determine the lipid content of the organisms gravimetrically. The rest of each extract was cleaned on an alumina/silica column. Concentrated sulfuric acid and a gel-permeation chromatography column were used to remove the lipids in the samples. Suppliers of the materials and solvents are given in Table S3. Finally, POPs were analyzed on a gas chromatograph with an ion-trap mass spectrometer (GC-MS, Finnigan Trace GC/PolarisQ) operating under an MS–MS mode. Further information on the chromatographic conditions is provided in Text S3. The following compounds were measured and quantified: hexachlorocyclohexanes (HCHs, including α-HCH, β-HCH, and γ-HCH), DDTs (*o*,*p*′-dichlorodiphenyldichloroethylene (DDE), *p*,*p*′-DDE, *o*,*p*′-dichlorodiphenyldichloroethane (DDD), *p*,*p*′-DDD, *o*,*p*′-DDT, and *p*,*p*′-DDT) and PCBs (PCB 28, PCB 52, PCB 101, PCB 138, PCB 153, and PCB 180). The terms ∑HCHs, ∑DDTs and $\sum_6$PCBs are used when referring to the sum of every group of contaminants.

2.3. Quality assurance/quality control (QA/QC)

Strict QA/QC measures were implemented to monitor the

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