โลหะหนักในปลาน้ำจืดในแม่น้ำพองและซี Heavy Metals in Freshwater Fish along Pong and Chi Rivers

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บทคัดย่อ

โลหะหนักบางชนิดที่สะสมในปลาอาจทำให้เกิดผลต่อสุขภาพ การศึกษาการสะสมของโลหะหนักในปลาน้ำจืด ที่สุ่มจากแม่น้ำพองและแม่น้ำชีระหว่างปลายปี พ.ศ. 2545 - ต้นปี พ.ศ. 2546 จุดเก็บตัวอย่าง 8 จุด A, B, C, D, E, F, G, H เรียงตามลำดับจากต้นน้ำบริเวณเขื่อนอุบลรัตน์สู่ปลายน้ำฝายมหาสารคามความยาวของลำน้ำประมาณ 180 กิโลเมตร ทำการตรวจวัด Cd, Co, Cr, Cu, Ni, Pb และ Zn ในกล้ามเนื้อและเนื้อเยื่อไขมันของตัวอย่างปลา ที่สุ่มจับมาได้โดยใช้อะตอมมิกแอบซอบซัน ค่าเฉลี่ยของปริมาณโลหะหนักที่ตรวจพบในเนื้อเยื่อไขมันสูงกว่าในกล้ามเนื้อ การจัดลำดับปริมาณโลหะหนักที่พบสูงไปต่ำเป็นดังนี้ Zn > Cu > Cr > Co > Ni > Pb > Cd จุดเก็บตัวอย่างที่พบปริมาณ โลหะหนักสะสมในปลาสูงกว่าจุดอื่น ๆ ได้แก่ B และ H แสดงให้เห็นว่าควรมีการติดตามศึกษาเกี่ยวกับโลหะหนัก สะสมในปลาโดยเฉพาะในจุดที่มีการตรวจพบโอกาสของการสะสมสูง

Abstract

Certain trace metals in fish may lead to health problems through the food chain. This work aimed to study the accumulation of some heavy metals in freshwater fish along the Pong and Chi rivers. The fish were randomly sampled during late 2002 until early 2003 from 8 monitoring sites, marked A, B, C, D, E, F, G, H, located downstream between Ubolratana reservoir and Mahasarakham weir covering a river length of about 180 km. Cd, Co, Cr, Cu, Ni, Pb and Zn, were determined from samples of the muscle and adipose tissue of the fish by using atomic absorption. In comparison, the levels of heavy metals in the adipose were higher than those found in the muscle of the fish. The mean concentrations in these tissues were listed from high to low levels as follows: Zn > Cu > Cr > Co > Ni > Pb> Cd. Sites B and H showed high potential bioaccumulation of heavy metals in these fish during the highly contaminated season. Pollutant stress in fish should thus be emphasized with an establishment of future biomonitoring at potential contamination sites.

คำสำคัญ: โลหะหนัก ปลา แม่น้ำ

Keywords: heavy metal, fish, river

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Introduction

Heavy metals from various sources released into natural waters can be a serious threat because of bioaccumulation and biomagnifications in the food chain (Rashed, 2001). Fish are often at the top of the aquatic food chain and may concentrate large amounts of some metals from water (Mansour and Sidky, 2003). Furthermore, fish are one of the most indicative factors in freshwater systems, particularly for the estimation of the risk potential from human consumption. Heavy metals taken up by an organism are distributed to different organs of the fish because of the affinity between them. The muscle, liver, gonads and gills are the organs of fish that have been mostly used in studies of heavy metals. Fish liver is the storage organ and thus was mostly used because of the fact that it accumulates the highest level of heavy metals (Amundsen et al., 1997; Papagiannis et al., 2004; Farkas et al., 2003; Yilmaz, 2003; Yilmaz et al., 2007). Muscle was included in these studies as a food source. However, these studies showed differences in the levels of different metals. Begum et al. (2005) suggested that there is a need for continuous monitoring of heavy metal concentrations in edible freshwater fish in Bangladesh because the lead levels found in 3 species of fish in Dhanmondi Lake were twice the maximum level regulated by the European Union at 0.2 mg/kg wet weight or 0.96 μ g/g dry weight. Farkas et al. (2003) measured the liver concentrations of cadmium in freshwater fish populating a lowcontamination site to exceed the tolerable levels for human consumption. Our previous study reports the accumulation of heavy metals in the liver of herbivorous and carnivorous fish along the Pong and Chi rivers (Sripanidkulchai et al., 2003) suggesting

that the highest liver Pb level in both types of fish was at a site of previous contamination (site B in Figure 1). This study follows on by using the adipose tissue and muscle of fish along Pong and Chi rivers which are usually consumed by people.

The Pong river is one of the most important water resource of the Northeast Region of Thailand, and particularly for Khon Kaen Province. The Pong River runs downstream towards the Chi River, which runs through provinces, merges rivers and flows to the Mekong River. Toxic moieties affecting the Pong River may generate significant incidences of pollution outbreaks. Incidences of pollution of the Pong River by accidental releases of chemical residues from the industries located along the river were reported in 1993 and 1994 (Khon Kaen University, 1994; 1995). A national alert has called for rehabilitation of the environmental quality of the Pong River. Some parts of the river where the water quality was classified as class 3-4 witnessed incidences of suspicious fish deaths in these areas. All heavy metals are potentially harmful to most organisms at some level of exposure and absorption. Certain heavy metals such as Cd, Co, Cr, Cu, Ni, Pb and Zn are known to be related to industry. The pollution of fresh water fish with heavy metals is an issue both from a hygienic and from an eco-toxicological point of view. Industries and communities situated on/or nearby riverbanks are usually major causes of waste generation.

Reports using levels of heavy metals in fish to indicate and assess the contamination of heavy metals in water were found to be very useful for environmental monitoring (Yilmaz, 2003; Szefer et al., 2003). There is little or no information regarding heavy metal accumulation in fish habitats along the Pong and Chi Rivers, particularly with respect to the distribution of heavy metals. The distribution of heavy metals in the fish could result in bio-transference and bio-magnification of the hazardous heavy metals to humans, because of our culture of fish consumption (Mansour and Sidky, 2003). It is also suspected that the water might be receiving some heavy metal(s) from various sources, not only from the industrial effluent but also from households and animal farms along the river. The study of heavy metals such as Cd in living organs usually emphasizes the analysis of the content in the liver and/or some other related tissues (Sirisangtrakul and Sripanidkulchai, 2004). In our study, the muscles and adipose tissues of freshwater fish were of particular interest as sources of food.

In this study, the levels of Cd, Co, Cr, Cu, Ni, Pb and Zn accumulating in the muscle and adipose tissue of fish samples from 8 sites of the Pong and Chi Rivers were determined. The fish samples were treated as both wet samples and dried samples because of the local consumption culture which likes both wet and dried fish. The streams have potentially been polluted by different types of industry, agricultural drainage and domestic waste water. The fishes, due to random sampling, were subsequently determined for species at the Department of Fisheries, Faculty of Agriculture, Khon Kaen University. It was aimed to observe the levels of heavy metals in the muscle and adipose tissue. The adipose tissue, not reported by any previous study, was thought to be a part of the lipophilic part of the body which might possibly store up some heavy metals. It might not be possible to explain the possible ecological effects of the metal levels found in fish from this study but it was intended that the

pollutant stress in fish would be emphasized and extended to be a future biomonitor.

Study sites

The Pong river originates from Phu Kradueng Mountain and Chi river sub-watersheds. It flows through Phu Kradueng District in Loei Province to Ubolratana reservoir and Nong Wai irrigation weir, as shown in Figure 1. The downstream watershed area comprises the Pong River and its tributaries from Ubolratana reservoir downstream to Mahasarakham weir on the Chi River and a short river section of the Chi River from Ban Tha Pra to its confluence with the Pong River. The downstream watershed area is divided into various sub-sections: Ubolratana reservoir to Nong Wai irrigation weir, Nong Wai irrigation weir to Ban Na Piang; Ban Na Piang to Mahasarakham weir (site H in Figure 1) and Ban Tha Pra downstream to the Chi and the Pong River confluence. Major tributaries which were predetermined to be sampling sites in this study included Huay Sai (site A as shown in Figure 1), Huay Kum Mum, Huay Chod and Chod lagoon (site B in Figure 1) and Huay Sue Ten and Sue Ten lagoon (site C in Figure 1). Nong Wai irrigation weir (#2 in Figure 1) is located about 34 km from Ubolratana dam (#1 in Figure 1). The community near Nong Wai weir, namely Nong Bua Noi (site D in Figure 1) was one of the predetermined sampling sites in this study.

Downstream from Nong Wai irrigation weir, the Pong River flows to join the Chi River. Between Nong Wai irrigation weir and 150 km downstream to Mahasarakham weir (site H), sampling from two lagoons, Tung Taew lagoon (site E) and Bua Noi lagoon (site G) were collected. Ta Kra Serm (site F) is located between sites E and G, and the last site is at Mahasarakham weir (site H). Numerous communities along the rivers are dominated by agriculture with mainly paddy or rice fields which start planting rice in April or May, when the use of fertilizers starts and increases until harvesting time in November and December. Other sources of potential pollution affecting the Pong and Chi rivers can be detected as discharges from various sources including industries, particularly sites B, E, F and G, farms, soils used for agricultural activities, waterweeds and community (domestic) wastes (Khon Kaen University, 1995).

Material and Methods

Sample Collection

A survey prior to sample collection was performed to determine the sites and the duration of sampling. The sampling was carried out during the dry season between April and November of 2002 when the water level of the river would be lowest and, thus, the highest concentration of most of the contaminants and pollutants would be expected. November is the end of the cultivation and the beginning of harvesting season, and this month would usually be the time when the land would peak with fertilizers and other agricultural substances. The sites for sample collection were chosen with the most likely high potential for pollutants as shown in Figure 1.

Fish samples were randomly caught from the 8 predetermined sites of the Pong and Chi rivers by using a beach seine from a boat and identified by Assist. Prof. Prapas Chalorpunrut, Department of Fisheries, Faculty of Agriculture, Khon Kaen University. Mature full-sized individuals of each fish species were categorized by a fishery scientist to minimize variations in metal concentration due to size of the organisms and separated into herbivorous and carnivorous. The types and pictures of the fish are shown in Figure 2.

Preparation and Analysis of Samples

The adipose tissue and muscle of the fish samples were separated, weighed and subjected to heavy metals analysis as previously described (Sirisangtrakul and Sripanidkulchai, 2004):

Briefly, each sample was heated at 70 °C until a constant weight was obtained, then ground to a powder. The sample was digested in conc. nitric acid (ultrapure) until completion and hydrogen peroxide (BDH Laboratory Supplies, Poole, England) was added to bleach the digested sample. The digested sample was dispersed in de-ionized water and filtered.

The standards, purchased from BDH Laboratory Supplies, England, were 1000 ppm solutions, each, of Cd, Co, Cr, Cu, Ni and Pb. Zn standard solution (1000 ppm) was from Merck, Germany. The sample was analyzed for Co, Cr, Cu, Ni, Pb and Zn by using flame atomic absorption (AAS Spectr. AA200, Varian, Australia) at λ_{max} of 243, 358, 327, 232, 217 and 214 nm, respectively. Cd content was determined by graphite furnace atomic absorption by using Spectra AA 640Z (Variance, Australia) at λ_{max} of 229 nm. The metal levels were calculated in micrograms per gram of wet weight and dried weight of the tissues used. Each sample was analyzed in triplicate.

Results and Discussions

The random sampling of fish in habitats along the Pong and Chi Rivers at sites A, B, C, D, E, F, G and H resulted in 3 species of herbivorous fish, namely *Trichogaster trichopterus* Pallas, Henicorhynchus siamensis Sauyage, Osteochilus hasselti Valenciennes and 2 species of carnivorous fish, namely Hampala dispar Smith, Channa striata Bloch, as shown in Figure 2.

Mean levels of heavy metals in the adipose and muscle of herbivorous fish habitats in the Pong and Chi Rivers collected from sites A-H are shown in Figure 3. From the adipose tissue of herbivorous fish samples, the ranges of Cd, Co, Cr, Cu, Ni, Pb and Zn found were 0.002 - 0.037, 0.74 - 8.00, 1.12 - 33.33, 5.88 - 96.52, 0.36 - 8.51, 0.25 - 1.99, 36-245 μ g/g wet weight and 0.004 -0.166, 3.38 - 39.98, 2.93 - 148.10, 29.11 -482.58, 0.94 - 37.80, 0.66 - 9.94 and 160 -988 μ g/g dried weight, respectively. The ranges of the levels of Cd, Co, Cr, Cu, Ni, Pb and Zn in the muscle of herbivorous fish samples were 0.002 -0.014, 0.29 - 4.17, 0.51 - 25.51, 2.77 - 17.59,0.15 - 4.86, 0.05 - 0.91, 26.8 - 88.9 µg/g wet weight and 0.006 - 0.066, 1.45 - 19.77, 2.3 -118.2, 12.4 - 93.19, 0.69 - 22.5, 0.02 - 4.57 and 133 - 487 µg/g dried weight, respectively. The highest levels of each heavy metal from the muscles of the herbivorous fish were as follows. Co was found at similar levels at site C (3.61 or 19.77 μ g/g of wet and dried weight) and site E (4.17 or 19.30 μ g/g of wet and dried weight). Cr had the highest content at site E (25.51 μ g/g wet weight or 118.2 μ g/g of dried weight). Cu presented the highest value at site C (17.03 μ g/g wet weight or 93.19 μ g/g of dried weight) and Ni at site E (4.86 μ g/g wet weight or 22.5 μ g/g of dried weight). Pb was highest at site F (0.91 μ g/g wet weight or 4.57 μ g/g of dried weight).

Figure 4 shows the mean levels of heavy metals found in the adipose tissue and muscle of

carnivorous fish sampled from sites A-H. The range of the levels of Cd, Co, Cr, Cu, Ni, Pb and Zn in the muscles of herbivorous fish samples were 0.002 -0.022, 0.41 - 1.65, 0.37 - 14.05, 4.32 - 14.07,0 - 1.38, 0 - 1.42, 24.27 - 45.10 µg/g wet weight and 0.007 - 0.08, 1.88 - 7.98, 1.56 -16.55, 21.08 - 71.43, 0.01 - 7.02, 0.01 - 7.02 and 118.4 - 215.0 µg/g dried weight, respectively. The highest levels of the heavy metals in the muscles of the carnivorous fish from each site were as follows. Site B had Cr (14.05 and 16.15 μ g/g of wet and dried weight), Cu (14.07 and 71.43 μ g/g of wet and dried weight), Ni (1.38 and 7.02 μ g/g of wet and dried weight) and Pb (1.38 and 7.02 mg/g of wet and dried weights). Site D had Cr (4.28 and 16.55 μ g/g of wet and dried weights) and Zn (45.10 and 215.0 μ g/g of wet and dried weights). Site F had Cd (0.022 and 0.082 μ g/g of wet and dried weights). The ranges of the levels of Cd, Co, Cr, Cu, Ni, Pb and Zn in the muscles of carnivorous fish samples were 0 - 0.021, 0.24 -2.01, 0.12 - 3.94, 2.40 - 8.50, 0.19 - 1.27,0.09 - 0.53, 7.91 - 35.56 µg/g wet weight and 0.008 - 0.088, 2.01 - 8.60, 2.26 - 14.50, 14.50- 49.95, 1.05 - 5.55, 0.77 - 3.90 and 86.34 -164.40 µg/g dried weight, respectively. For all observations from muscles of carnivorous fish, the highest levels of the heavy metals from all sites were as follows. Site A had Cr (3.08 and 13.55 μ g/g of wet and dried weights). Site B had Cr (2.98 and 14.50 μ g/g of wet and dried weights) and Ni (1.14 and 5.55 μ g/g of wet and dried weights). Site D had Cd (0.021 and 0.088 µg/g of wet and dried weights) and Co (2.01 and 8.60 μ g/g of wet and dried weights). Site F had Cr (3.94 and $13.10 \,\mu$ g/ g of wet and dried weights) and Cu (8.50 μ g/g or

35.89 μ g/g of wet and dried weights). Site G had Cu (2.40 and 49.95 μ g/g of wet and dried weights), Pb (0.19 and 3.90 μ g/g of wet and dried weights) and Zn (7.91 and 164.40 mg/g of dried weights). Mean concentrations in the muscles and adipose tissue of the fish samples could be summarized in sequence from highest to lowest levels, as follows: Zn > Cu > Cr > Co > Ni > Pb > Cd. A similar order was also found by a study of Yilmaz et al. (2007). Zn, being an essential trace element which is also one important micronutrient for humans, frequently occurs in nature together with other metals of which Cd is one the most common (Papagiannis et al., 2004). The presence of heavy metals, particularly transition metals, in nature is a complex phenomenon which might need more investigations. The level of Pb from most sites found in the muscle and adipose tissue of herbivorous and carnivorous fish exceeded the limits of FAO/WHO (1989) and EU but not for Cd. The sampling sites and duration employed in this study were intended to observe the highest potential contaminations and at least Pb was the result of such intention.

Adipose tissue uptakes lipid-soluble compounds rather than water-soluble compounds, and thus could be one of the distribution depots of some heavy metals. This study has shown that adipose tissue accumulated these heavy metals at a higher level than the muscle. Concentrations of heavy metals detected in the muscle and adipose tissue when compared to those found in the liver of these fish reported by Sripanidkulchai et al. (2003) suggested that each heavy metal might have accumulated in different tissues/organs at a different distribution profile. From our previous study, it was obvious that the liver accumulated the highest levels of these metals. From this present study, the adipose tissue accumulated higher levels of metals than the muscle but lower than the liver. In contrast to the livers of the carnivorous fish which obviously stored Cd and Pb to a higher extent than those of the herbivorous, the adipose tissue and the muscle could not show good correlation of heavy metal accumulation for comparison. However, the results might suggest some awareness for fish consumption is necessary as the adipose tissue is also used for some products. Keeping in mind that the fish were caught during the dry season and the period with peak concentration of chemicals in the rivers, the results obtained represent the highest potential of heavy metal accumulation in these fish. In normal circumstances, lower levels should be expected.

The observed variability of heavy metals in different species depends on feeding habitats, ecological needs, metabolism, age, size, length of the fish and their habitats. The feeding habits investigated in this study involved differentiating the fish into herbivorous and carnivorous. Herbivorous fish are expected to be more susceptible to the pollutants than carnivorous when compared to environmental exposure while carnivorous fish are susceptible to the accumulation of heavy metals. Moreover, it was observed that dry to wet weight of the fish sample gave an overall ratio of 4.8 when compared by using data of each heavy metal found and this figure was the same as that reported by Begum et al. (2005).

To identify the source of such contamination, it is important to further study and elaborate on the source of the pollution, the habitat and behavior of the fish. It might be interesting to compare the results obtained from various tissues and from various species of fish with known life style and life cycle. This would show the accumulation and distribution pattern of pollutants. Furthermore, since fish are mobile and diverse, the overall data which would help us to explain the heavy metal contamination should at least consist of data on heavy metals in water, sediment, fish, plankton, shells and waterweeds (Yap et al., 2002, Mansour and Sidky, 2003). However, in attempts to explain the results by using heavy metal analyses of water from the same site at the same sampling date performed by Mungkarndee and Wirojanagud (2003), there was only one possible heavy metal, Cr, for comparison. While the water showed high Cr at site B only, the adipose tissues and muscles of the fish accumulated Cr differently. It is, therefore, indicated that the results should not be discussed in relation to the industrial site because the most decisive factor affecting the presence of heavy metals in any fish is the absorption and distribution of the ions in the body of the fish.

Conclusions

The study reveals that certain heavy metals accumulated in the muscle and adipose tissue of herbivorous and carnivorous fish at some sampling sites along the Pong and Chi rivers might have been involved in the food chain of its habitat. Although the selected sites of study were the most likely to be contaminated ones and during the most vulnerable period of the year, the sources and causes were not able to be identified. Pb was the hazardous heavy metal found in the samples of the fish. Pollutant stress in fish should be emphasized with an establishment of future biomonitoring at potential contamination sites.

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Figure 1. Sampling Stations along the Pong River.



Trichogastertrichopterus Pallas Family Osphronemides



Henicorhynchus siamensis Sauvage Family Cyprinidae



Osteochilus hasselti Valenciennes Family Cyprinidae



Figure 2. Illustration of fish sampled from sites A – H categorized into herbivorous fish (top row) and carnivorous fish (bottom row).



Figure 3. Heavy metals found in herbivorous fish sampled from sites A – H along the Pong and Chi rivers: white columns represent μg of heavy metal per g of wet weight of tissues; gray columns μg of heavy metal per g of dry weight of tissues (n = 3).



Figure 4. Heavy metals found in carnivorous fish sampled from sites A – H along the Pong and Chi rivers: white column represents μg of heavy metal per g of wet weight of tissues; gray column μg of heavy metal per g of dry weight of tissues (n = 3).