Chemosphere 148 (2016) 171–177

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

The effects of mariculture on heavy metal distribution in sediments and cultured fish around the Pearl River Delta region, south China

Peng Liang [a], Sheng-Chun Wu [a], Jin Zhang [a], Yucheng Cao [a], Shen Yu [b, **], Ming-Hung Wong [a, c, d, *]

[a] School of Environmental and Resource Sciences, Zhejiang Agricultural and Forest University, Lin'an, Zhejiang Province, China
[b] Key Laboratory of Urban Environment and Health, Institute of Urban Environment, Chinese Academy of Sciences, Xiamen, China
[c] School of Environment, Jinan University, Guangzhou, China
[d] Consortium on Health, Environment, Education and Research (CHEER), Hong Kong Institute of Education, Hong Kong, China

HIGHLIGHTS

- PCA analyses show C, N, Cu, Zn and Pb are classified in one subgroup.
- Result implies that Cu, Zn and Pb were derived from the input of fish feed.
- Heavy metal detected in fish were not correlated with concentrations in sediment.

ARTICLE INFO

Article history:
Received 3 June 2015
Received in revised form
19 October 2015
Accepted 26 October 2015
Available online 22 January 2016

Handling Editor: Martine Leermakers

Keywords:
Aquaculture
Heavy metals
Pearl River Delta
Sediment
Fish

ABSTRACT

Thirty-six sediment samples were collected from six mariculture sites and corresponding reference sites (approximately 200–300 m away from each mariculture site) to study the effects of mariculture on heavy metal: copper (Cu), zinc (Zn), chromium (Cr) and lead (Pb) distribution in sediments and cultured fish around the Pearl River Delta region, south China. The mean concentrations of Cu, Zn, Cr and Pb in all mariculture sediment samples were 109, 273, 99 and 33 mg/kg, compared with 63, 209, 56 and 23 mg/kg for reference sediment samples, respectively. The Pollution Load Index of sediment for each site was over 1, implying substantial heavy metal pollution. The results of principal component analysis and hierarchical clustering analysis indicate that marine aquaculture activities have enriched the surface sediments underneath mariculture rafts with Cu, Zn and Pb, possibly due to the accumulation of unconsumed fish feeds, fish excreta and antifouling paints (except Cr). Two kinds of fish feeds, feed pellets and trash fish commonly used in Hong Kong were analyzed. The concentrations of Zn and Cu contained in feed pellets were significantly higher ($p < 0.05$) than in trash fish due to addition of these elements as growth promoters during pellet production. However, the Pb content in trash fish (due to contamination) was significantly higher ($p < 0.05$) than in feed pellets. Three cultured fish species, namely red snapper (*Lutjanus campechanus*), orange-spotted grouper (*Epinephelus coioides*) and snubnose pompano (*Trachinotus blochii*), were collected from each mariculture site for metal analysis. Lead concentrations in 21 fish samples exceeded the Chinese safety guideline (0.5 mg/kg, GB18406.4–2001), indicating that Pb contamination in cultured fish would be a public health concern.

1. Introduction

Heavy metals are important pollutants in the environment due to their potential toxicity, persistence and tendency to bioaccumulate (Ye et al., 2012). Zinc (Zn), chromium (Cr) and copper (Cu) are biologically essential, but the identification of contamination sources of these metals and their behavior in the natural system are also necessary, since they have the potential to be toxic to biota above certain threshold concentrations (Vallee and Auld, 1990). Lead (Pb) is classified as a priority pollutant because it is not required for metabolic activity and can be toxic even at quite

* Corresponding author. School of Environmental and Resource Sciences, Zhejiang Agricultural and Forest University, Lin'an, Zhejiang Province, China.
** Corresponding author.
E-mail addresses: syu@iue.ac.cn (S. Yu), mhwong@hkbu.edu.hk (M.-H. Wong).

http://dx.doi.org/10.1016/j.chemosphere.2015.10.110
0045-6535/© 2015 Published by Elsevier Ltd.

low concentrations. Moderate exposure to Pb can significantly reduce human semen quality and is related to many diseases in adults and children alike (e.g. damage to DNA or impairment of the reproductive function) (Telišman et al., 2007).

Aquaculture activities generate considerable amounts of effluent, exerting undesirable impacts on the marine environment (Wu, 1995). Direct pollution discharges commonly occur at two stages of aquaculture, i.e. during aquaculture production and farm activities (Fernandes et al., 2001). During the first stage, effluents containing organic and inorganic nutrients are released to the vicinity of the water body (Wu, 1995). Farm activities in the second stage such as dumping of fish processing waste and dead fish contribute additional pollutant loading to the aquatic environment. Moreover, cage aquaculture practices may also impose adverse effects by releasing substantial amounts of various heavy metals to the ambient marine environment (Tal et al., 2009).

Recently, there has been increasing public concern about the safety of mariculture products due to potential metal transfer from contaminated sediments to aquatic biota at mariculture zones (Feldlite et al., 2008). Accumulation of heavy metals in aquatic products may subsequently pose a threat to human health via seafood consumption.

Previous studies have reported heavy metal concentrations in sediments and fish collected from Hong Kong coastal waters (Cheung et al., 2008) but no attempt has been made to trace the sources of heavy metals accumulated in the sediment and fish, either from natural sources or aquaculture activities. The major objectives of the present study were therefore to evaluate the level of metal enrichment in mariculture surface sediments, to identify the possible origins and sources of metal contamination in sediments by principal component analysis (PCA) and hierarchical cluster analysis (HCA), and to evaluate the potential health risks of heavy metals contained in major species of cultured fish for human consumption.

2. Materials and methods

2.1. Study areas

Six marine aquaculture sites around Hong Kong and adjacent mainland China coastlines comprising Tung Lung Chau (TLC), Sai Kung (SK), Sam Mum Tsai (SMT), Tsing Yi (TY), Mirs Bay (MB) and Xi Xiang (XX) were selected as sampling sites (Fig. 1). Detailed information on these sampling sites is listed in Table S1.

2.2. Sampling procedures

Surface sediments were collected by a grab sampler from mariculture sites and the corresponding reference sites (about 200–300 m away from the fish rafts of each site). Triplicate samples underneath the floating rafts of each site and the corresponding reference sites (without floating rafts) were collected.

Three major species of cultured fish, namely red snapper (*Lutjanus campechanus*), orange-spotted grouper (*Epinephelus coioides*) and snubnose pompano (*Trachinotus blochii*), were collected from each site (except XX), with 4–6 replicates for each species. Fish samples from XX were purchased from wet markets owing to the unavailability of live fish at the site. The site has been designated as a non-mariculture zone since 2004 (Shenzhen Government, 2009) because of poor water quality. Fish feed samples, originally bought from markets, were obtained from a fish farmer in TLC only as fish farmers from other sampling sites confirmed that their feeds were also purchased from local markets.

All the sediment and fish samples were transported to the laboratory after collection and freeze-dried, ground and homogenized.

2.3. Sample analysis

Fish (flesh) and sediment samples were subjected to acid digestion. 0.5 g of fish and sediment samples was digested using concentrated nitric acid and aqua fortis at 95 °C, until a clear solution was obtained. The metal concentrations were determined by inductively coupled plasma mass spectrometry (ICP-MS) (Agilent 7500 CX, Santa Clara, CA).

2.4. Quality assurance of sample analysis

Two method blanks, two certified reference materials (CRMs), NIST 2702 (National Institute of Standards and Technology, US, soil) and TORT-2 (National Research Council Canada, oyster), and 10% replicate samples accompanied each sample batch (up to 30 samples). Recovery rates across all samples ranged from 77.3 to 112.6% and relative percentages of sample duplicates ranged from 0.37 to 24.4%.

2.5. Data analysis

The differences in heavy metal concentrations in sediments and fish among all the sites were evaluated by conducting analysis of variance using the SPSS 16.0 for Windows software package.

The Contamination Factor (CF) was calculated as the ratio obtained by dividing the concentration of each metal in the sediment using the baseline value or background value. CF values were based on Hakanson (2011) and cite reference where: CF < 1 indicates low contamination; 1 < CF < 3 moderate contamination; 3 < CF < 6 considerable contamination; and CF > 6 very high contamination.

Pollution Load Index (PLI) was used to determine the integrated pollution status of combined toxicant groups at each sampling station by calculating the nth root of the product of the n CF for the tested metals using the following formula:

$$PLI = (CF_1 \times CF_2 \times CF_3 \times CF_n)^{1/n}$$

This index provides a simple, comparative means for assessing the level of heavy metal pollution, where PLI>1 indicates that pollution exists and PLI<1 denotes no metal pollution.

Enrichment factor (EF) is a useful tool in determining the degree of anthropogenic heavy metal pollution (Sakan et al., 2009). The EF is calculated using the relationship below:

$$EF = (Metal/Fe)\ Sample/(Metal/Fe)\ Background$$

Fe was used as the reference element for geochemical normalization in this study since Fe is associated with fine solid surfaces. In addition, Fe geochemistry is similar to that of many trace metals and the natural concentration of Fe tends to be uniform. EF values were calculated (Sakan et al., 2009), where: EF < 1 indicates no enrichment; <3 minor enrichment; 3–5 moderate enrichment; 5–10 moderately severe enrichment; 10–25 severe enrichment; 25–50 very severe enrichment; and >50 extremely severe enrichment.

The enrichment ratio (ER) of sediment was calculated as: $ER = (X_m - X_r) \times 100/X_r$, where X_m is the average concentration of metals at marine aquaculture sites, and X_r the average concentration of metals at reference sites.

3. Results

3.1. Heavy metal concentrations in sediments

Table 1 shows the concentrations of heavy metals in surface