Spatial Distribution and Contamination Status of Arsenic, Cadmium and Lead in some Coastal Shrimp (*Macrobrachium rosenbergii*) Farming Ponds of Viet Nam.

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ABSTRACT

The present investigation was executed to determine the distribution pattern and contamination status of arsenic (As), cadmium (Cd), and lead (Pb) in the coastal shrimp (*Macrobrachium rosenbergii*) industry of Viet Nam. Mud samples were collected from twenty randomly selected ponds of four shrimp farming coastal zones and analyzed using an atomic absorption spectrophotometer. Estimated results of three metals clearly revealed As (135.55–1868.5 μg kg⁻¹) and Cd (0–2708 μg kg⁻¹) content of the mud samples were remarkably higher than that of the Pb (0–59.57 μg kg⁻¹) in all ponds. Considering the mud-water dynamics and permissible limit of metals in drinking water, surface water quality, and livestock water quality, it may be concluded that 55, 65, and 5% investigated ponds are contaminated by As, Cd, and Pb, respectively. In contrast, it may also be suggested that As and Pb concentration of all investigated ponds are within permissible limits whereas 60% ponds are contaminated with Cd if the values are compared with the common range of these metals as considered for soil. From these results it may be concluded that the estimated high metal concentrations of these ponds facilitate progressive bioaccumulation within the shrimp tissue by means of various environmental factors.

(Keywords: coastal shrimp pond, mud, toxic metals, heavy metals, distribution patterns, contamination)

INTRODUCTION

Recently, degradation of coastal environments through aquatic contamination has been recognized as a growing concern worldwide due to persistent and accumulative threat from heavy metal posing which poses serious human health implications through the ingestion of marine organisms. Coastal environments are often extensively contaminated by receiving various pollutants such as toxic metals, nutrients, and pesticides (Readman et al., 1992; Green-Ruiz and Paez-Osuna, 2001; Ruiz-Fernandez et al., 2001a, 2001b, 2002, 2003; Soto-Jimenez et al., 2003; Takarina et al., 2004; Bhakta et al., 2009). The world once witnessed an extreme situation of heavy metal contamination of coastal waters in Minamata Bay, Japan where hundreds of individuals suffered severe and tragic cases of mercury toxicity (Minamata Disease) (Zhang et al., 2005).

Mining and smelting; industrial sources (coal, oil, chemical, fertilizer, pesticides, etc.); urban waste, wastewater discharges; and shipping activities are the major anthropogenic sources which contribute significantly to the natural background of toxic metals in soils and sediments of coastal regions. Despite various anthropogenic sources, toxic metal contamination and enrichment in water, sediment, and soils may also be affected by a variety of geochemical (weathering of rocks), biogeochemical, and biological factors. According to Filipek and Owen (1979) distribution and pollution of heavy metals in sediments is regulated by several factors including source materials, weathering processes, sediment transport, dissolution, and reaction kinetics. Robertson (1989) proposed that arsenic bearing rocks and minerals are the priority natural sources of arsenic; whereas Ni enriched basic rocks (Forstner, 1977) and Pb enriched igneous rocks (Hume, 1934; Maynard, 1983) are also important sources of Ni and Pb, respectively, in natural background. The breakdown of organic matter and the subsequent release of metals, due to falling of the Eh-pH environment, are known to
influence vertical and horizontal distribution of metals in sediment (Lasheen, 1974).

Among the most toxic and heavy metals, arsenic (As), cadmium (Cd), lead (Pb), and mercury (Hg) are the major hazardous priority substances in the list of pollutants which are responsible for causing a serious health hazards even at low concentrations (Choudhury and Modipalli, 2008; Roberts, 1999). In general, these metals are biologically nonessential, non-biodegradable, persistent type of toxic metals, and can easily accumulated in the sediments and in the aquatic flora and fauna (by bioaccumulation and biomagnification), thus causing a gross biological impact.

Arsenic is ubiquitous in the environment having the property of causing acute and severe toxic metal poisoning of water (surface and ground) which is already identified in different countries reporting its health risk by many scientists (Hemond and Solo-Gabriele, 2004; Murphy et al., 1989). Cadmium is a ‘priority pollutant’, not only from the human health perspective, but also from a broader ecosystem viewpoint (Campbell, 2006). Heavy metals have been highlighted in the discovery of excessive levels of toxic substances (particularly cadmium) in seafood and other living organisms (Talbot, 1983; McConchie et al., 1988). Pb is also known as a toxic element at even low concentrations may progressively accumulate in water, sediments, and biological tissues to levels above natural background. Elevated metal concentrations in aquatic systems may pose health risks to marine life (e.g. primary producers, fish, and filter feeders) and consumers of these organisms, including humans (Bryan and Langston, 1992; Green-Ruiz and Paez-Osuna, 2001; Paez-Osuna et al., 2002; Soto Jimenez et al., 2001, 2003; Frías-Esparciueta et al., 2008).

In aquatic ecosystems, different trophic organisms take up heavy metals, including Cd, (Bhakta and Munekage, 2008) and concentrate them in amounts considerably higher than those found in the environment (Ferard et al., 1983). Bottom dwellers such as shrimp, crabs, lobsters, and filter-feeders accumulate more heavy metals than fish due to their frequent contact with bottom sediments (Attar et al., 1992; Bou-Olayan et al., 1995; Campbell, 1988; Forstner, 1990; Kaviraj and Guhathakurta, 2004).

On account of the above impacts, the present study includes the coastal shrimp farming area of Viet Nam, a South East Asian developing country rapidly developed industries leading to immense problem in their coastal environment. In addition, the coastal aquaculture industry of Viet Nam is recognized as an important livelihood and developed rapidly due to heavy demand and attractive returns during last decades. In Viet Nam, Cd pollution has been reported by the Center for Experimental Analysis in 1996 in the mud of the Nhieu Loc Thi Nghe canal (fluctuated between 28 – 35 mg kg\(^{-1}\)) and the Institute for Environment and Resources in 1998 in the Sai Gon Dong Nai River system (9.7 – 25 mg kg\(^{-1}\)). It has been reported that Red River delta is also contaminated by arsenic and other metals (Michael et al., 2007; Shinkai, 2007; Bhakta et al., 2009). Though various researchers have examined metals pollution, information regarding the contamination status in the mud of coastal aquaculture industry in Viet Nam is very scant. Therefore, the present study has been undertaken to determine the spatial distribution and contamination status of arsenic, cadmium and lead in mud of coastal shrimp firming ponds of Viet Nam.

**MATERIALS AND METHODS**

**Description of Study Sites**

The present investigation considered four coastal shrimp farming zones; Vunh Tau, Nha Trang, Da Nang, and Hue located in Viet Nam. Vunh Tau (VT) (10°21'0" N and 107°4'0" E) and Nha Trang (NT) (12°13'40" N and 109°11'38" E) are situated on the coast of the southeastern region whereas Da Nang (DN) (16°23'38" N and 108°11'38" E) and Hue (HU) (16°28’0" N and 107°36’0" E) are also located on the coast of the South China Sea (Figure 1). Vunh Tau is one of the most industrially developed provinces of Viet Nam with industries including petroleum (the most important), electricity, power plants, petrochemicals (urea plant, polyethylene, steel and cement industries), fisheries and shrimp, as well as tourism. Coastal aquaculture, tourism, and a number of light industries are recognized as the major industries in the remaining three coastal zones. Mountains are the specific geographical criteria of these four zones of Viet Nam.
Bottom mud of shrimp ponds are clay and silty in nature with sand (VT 26 – 34%, NT 24 – 30%, DN 31 – 40% and HU 19 – 27%). All investigated ponds are engaged for shrimp, *Macrobrachium rosenbergii* farming during last 7 to 8 years and various chemical fertilizers as well as commercial feeds are applied during the course of culture practice (information was obtained from each farmer while sampling). Though shrimp farming is continuing in all investigated ponds presently, the growth rate and production of shrimp at these sites have shown a surprisingly downward trend year after year due to the prevalence of disease and water pollution.

**Sampling**

Three mud samples (0 – 5 cm) of three different places of each pond were collected from five randomly selected shrimp ponds of each predetermined four sampling stations (VT1–5, NT6–10, DN11–15 and HU16–20) during the period of September 17 to 30, 2008. Procured mud samples were preserved with 0.5 N HCl in clean plastic bottles (250 ml) for analysis.

**Processing and Analysis of Mud**

Collected samples were evenly mixed and allowed to air dry for a one week period in the laboratory. Air dried mud samples were ground with ceramic mortar and pestle to a homogeneous powder and sieved through a 0.42 mm mesh to get an equal size of particles for analysis (Bhakta and Munekage, 2008).

One gram of powdered mud was digested by the mixture of 3 ml of concentrated nitric acid and 9 ml of concentrated hydrochloric acid using the multiwave reaction system (Multiwave 3000, Anton Paar, Perkin Elmer) following the guidelines mentioned in the manual of this equipment for mud digestion. After digestion the resulting solutions were allowed to cool, filtered through Whatman No. 42 filter paper, and the residue was diluted with double-distilled water to 50 ml and finally measured by using flame atomic absorption spectrophotometer (Perkin Elmer, AAnalyst 200). The concentration of each sample was calculated in terms of microgram for per kilogram dry weight of mud.

**Statistical Analysis**

Three mud samples for each pond were used as three replicates in the analysis process and the mean value of three replicates was considered for statistical analysis. All data were subjected to one way analysis of variance (ANOVA) in the general linear model and correlation study using the SPSS 10.05 statistical package. The statistical package (EASE, M–STAT) was used in the computer to perform the analysis of least significance difference (LSD). Statically significant differences were expressed as p < 0.05.

**RESULTS AND DISCUSSION**

**Arsenic (As)**

A significant difference was observed in the estimated As content (135.55 – 1868.5 μg kg⁻¹) of all twenty investigated ponds (ANOVA, P<0.05). The concentration of As was varied from 1330.5 to 1868.5 μg kg⁻¹, 388.7 to 1810 μg kg⁻¹, 135.55 to 679.5 μg kg⁻¹, and 204.35 to 632 μg kg⁻¹ in the five ponds of VT, NT, DN and HU, respectively (Figure 2a). The maximum value 1868.5 μg kg⁻¹ of VT₃ exhibited 1.9 to 1278 % higher than that of the remaining ponds.
As Concentrations of Different Ponds (a) and Distribution Pattern of As (b) Obtained from Twenty Investigated Coastal Shrimp Ponds of Viet Nam. (Ponds corresponding to same script (a – f) revealed no significant difference. Inset of b shows percentage occurrence of three frequency groups of As).

Frequency distribution of As exhibited that higher percentage classes (>1000 – ≤2000 \(\mu g\) kg\(^{-1}\)) were exclusively occurred in VT (25%) and NT (15%), whereas, 15% moderate classes (>500 – ≤1000 \(\mu g\) kg\(^{-1}\)) and 45% lower percentage classes (0 – ≤500 \(\mu g\) kg\(^{-1}\)) were pronounced in NT, DN and HU (Figure 2b).

**Cadmium (Cd)**

Content of Cd in the mud of twenty shrimp ponds also showed a remarkably significant (ANOVA, \(P<0.05\)) range (0 – 2708 \(\mu g\) kg\(^{-1}\)) of variation. Concentration of Cd ranged from 2128.25 to 2708 \(\mu g\) kg\(^{-1}\), 492.05 to 1291.75 \(\mu g\) kg\(^{-1}\), 0 to 1254.75 \(\mu g\) kg\(^{-1}\) and 0 to 1520 \(\mu g\) kg\(^{-1}\) in each five ponds of investigated regions, VT, NT, DN, and HU, respectively (Figure 3a). Though value was maximum in VT\(\text{5}\) (2708 \(\mu g\) kg\(^{-1}\)) but DN\(\text{13,14}\) and HU\(\text{4}\) showed nil (0 \(\mu g\) kg\(^{-1}\)) Cd content in mud. Frequency distribution of Cd showed that higher percentage classes (>2000 – ≤4000 \(\mu g\) kg\(^{-1}\)) were exclusively occurred in VT (25%), whereas, 40% lower percentage classes (0 – ≤1000 \(\mu g\) kg\(^{-1}\)) and 35% moderate frequency classes (>1000 – ≤2000 \(\mu g\) kg\(^{-1}\)) occurred exclusively in the ponds of NT, DN and HU (Figure 3b).
**Lead (Pb)**

As with As and Cd, the concentration of Pb also registered a significant difference in all ponds ranging from 0 to 59.57 μg kg⁻¹ in all ponds (ANOVA, P<0.05). In each sampling station, the concentrations of Pb also varied greatly and inconsistently from 20.45 to 59.57 μg kg⁻¹, 6.17 to 35.6 μg kg⁻¹, 0 to 17.22 μg kg⁻¹, and 0 to 15.6 μg kg⁻¹ among the five ponds of VT, NT, DN, and HU, respectively (Figure 4a). Estimated Pb content of mud was highest in VT (59.57 μg kg⁻¹) and no Pb was found in DN, NT, and HU (0 μg kg⁻¹). Frequency distribution of Pb revealed that 45% lowest concentrations classes (0 – ≤15 μg kg⁻¹) occurred in NT, DN and HU followed by 35% moderate concentration classes (>15 – ≤30 μg kg⁻¹) which showed nearly even distribution in VT, DN, NT and HU and 20% higher concentration classes (>30 – ≤60 μg kg⁻¹) were exclusively prevalent in VT and NT (Figure 4b).

Spatial distribution pattern of three metals, arsenic, cadmium and lead varied significantly from one pond to another within same station as well as among the different stations (Figure 2b, 3b and 4b). A distinct sampling zone wise variation was clearly apprehended among four investigated sites from the critical appraisal of frequency distribution of different concentration level of three toxic metals. Moreover, all data also clearly revealed that As (135.55 – 1868.5 μg kg⁻¹) and Cd (0 – 2708 μg kg⁻¹) content were significantly higher than that of the Pb (0 – 59.57 μg kg⁻¹) in all ponds excepting pond 19 where no Cd recorded.

Frequency distribution of three metals demonstrated that (1) higher concentration classes of Cd (>2000 – ≤4000 μg kg⁻¹) were exclusively occurred in VT (25%) whereas, As (>1000 – ≤2000 μg kg⁻¹) and Pb (>30 – ≤60 μg kg⁻¹) were also found only in VT (As 25%, Pb 15%) and NT (As 15% and Pb 5%); (2) moderate concentration classes of As (>500 – ≤1000 μg kg⁻¹), Cd (>1000 – ≤2000 μg kg⁻¹) and Pb (>15 – ≤30 μg kg⁻¹) were found in VT (As and Cd 0%, Pb 10%), NT (As 5%, Cd 15% and Pb 10%) DN (As 5%, Cd 10% and Pb 10%) and HU (As 5%, Cd 10% and Pb 5%); and (3) lower concentration classes of As (0 – ≤500 μg kg⁻¹), Cd (0 – ≤1000 μg kg⁻¹) and Pb (0 – ≤15 μg kg⁻¹) were only concentrated in NT (As 5%, Cd 10% and Pb 10%), DN (As 20%, Cd 15% and Pb 15%), and HU (As 20%, Cd 15% and Pb 20%) with higher percentage.

From this critical interpretation of distribution pattern of three metals, all ponds can be grouped into three categories – lower metal concentration group which comprises the ponds of NT (25%), DN (50%), and HU (55%); moderate metal concentration group were found in VT (10%), NT (30%), DN (25%), and HU (20%) – and higher metal concentration group exclusively occurred in VT (65%) and NT (20%) (Figures 2b, 3b, and 4b). This proposition may also be concluded that VT is the zone of higher metal concentration due to exclusively possessing higher metal content ponds whereas NT, DN and HU are belong to the mixed types of metal content zones because of
having ponds with moderate and lower metal concentration according to the results of three metals obtained from our present study.

From the available literature, As, Cd, and Pb content in the mud of twenty investigated shrimp ponds are lower (Figure 2, 3 and 4) than that of the common range of soil mentioned in Table 1; but it is important to note that bottom mud is directly associated with short overlaying water column of shrimp ponds which may poses a serious adverse impact to benthic as well as aquatic organisms by increasing the concentration in this short water column having recommended permissible limit of these metals are narrow. In this respect, it should be strongly mentioned that any substances of mud and water compartment of an ecosystem always has a tendency to reach an equilibrium state by moving from higher to lower concentration zone by means of absorption, adsorption, desorption, bioturbation and microbial metabolic process. Mud involves inputs and losses of substances, movement of substances within pond water and mud, transfer of substances across the soil-water interface, and uptake or release of substances by the soil through ion exchange, dissolution and decomposition (Tchobanoglous and Schroeder, 1987; Bhakta et al., 2007). On the basis of these factors, it is obvious that mud-water interfaces interaction is an important governing factor to increase the metal concentration in the overlaying water which bottom mud is with high metal content. Therefore, though mud content a lower concentration of metal than the common range of soil (Table 1) but it is higher than that of the live stock water quality limit in some ponds which may be responsible to increase the metal concentration in their overlaying water through the function of above mentioned conditions that is not save for the aquatic organism inhabited or cultured. Conditions having a measurably higher magnitude of metal content in bottom mud of aquaculture system equally effect the growth and health condition of aquatic organism by accumulating in the tissue level as directly governed by the high metal content water.

Table 1: Permissible Limit of Metals (As, Cd and Pb) in Drinking Water, Surface Water Quality, Livestock Water Quality, and Common Range in Soil.

<table>
<thead>
<tr>
<th>Metal</th>
<th>Primary drinking water standard (MCL)¹ (µg l⁻¹)</th>
<th>Recommended permissible limit of drinking water (WHO)² (µg l⁻¹)</th>
<th>Surface water Quality³ (µg l⁻¹)</th>
<th>Livestock water quality⁴ (µg l⁻¹)</th>
<th>Common range in soils⁵ (µg kg⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Arsenic (As)</td>
<td>50</td>
<td>10</td>
<td>40</td>
<td>500</td>
<td>1000-50,000; 1000-40,000⁷</td>
</tr>
<tr>
<td>Cadmium (Cd)</td>
<td>5</td>
<td>3</td>
<td>20</td>
<td>500</td>
<td>10-700</td>
</tr>
<tr>
<td>Lead (Pb)</td>
<td>15⁶</td>
<td>10</td>
<td>100</td>
<td>50</td>
<td>2000-200,000</td>
</tr>
</tbody>
</table>

Note: Table modified according to required metals and incorporated WHO recommended limits in second column.

1MCL-Maximum Contaminant Level for drinking water from a public water supply system. From “Current Drinking Water Standards”, Environmental Protection Agency, Office of Water.
2WHO-World Health Organization drinking water guidelines.
4Levels of metals in water suitable for cattle or other livestock. Summarized from Environmental Protection Agency, Council on Agriculture, Science and Technology (CAST), and National Academy of Sciences (NAS).
5Naturally occurring in the soil. Limits are taken from Lindsay, 1979.
6Lead has action levels in drinking water standards instead of MCL’s. From “Current Drinking Water Standards”, Environmental Protection Agency, Office of Water.

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Studies suggested that higher concentrations of Cd, Pb, and Fe in water often exceed the WHO guideline values of these metals for drinking purpose which are capable to bio-concentrate in fish and shrimps cultured in these waters and humans consuming such fish or shrimp may exceed the provisional maximum tolerable daily intake levels for human of these metals (Kaviraj and Guhathakurta, 2004). Deb and Santra (1997) observed, in a sewage fed fish pond ecosystem, that from a relatively innocuous concentration of Pb (0.09 μg kg⁻¹) and Zn (0.23 μg kg⁻¹) in water, fish could concentrate 78 to 80% higher amounts of Pb and 77% higher amounts of Zn in their body. In brackishwater ponds of Sunderban, shrimp have been found to accumulate Zn at much higher levels than fish (Ghosal et al., 1997).

On account of the above discussion and considering the permissible limit of metals (As, Cd, and Pb) in drinking water, surface water quality, livestock water quality, and common range in soil mentioned in Table 1, the investigated twenty shrimp ponds of four coastal region of Viet Nam may also be divided into two categories in respect to contamination level of aquaculture environment – contaminated ponds which having >500 μg kg⁻¹ concentration of As and Cd as well as > 50 μg kg⁻¹ concentration of Pb and non contaminated ponds which having As and Cd ≤ 500 μg kg⁻¹ and Pb ≤ 50 μg kg⁻¹. Furthermore it might be mentioned that ponds VT₁–₅, NT₁–₅ and DN₂ are contaminated with both As and Cd; only Cd contaminated ponds are DN₃, HU₃ and 5 whereas VT₁ is contaminated with only Pb (Figures 2b, 3b, 4b).

The statistical analysis of all data showed positive and significant correlation between As and Cd (r = 0.8064), As and Pb (r = 0.7912), and Cd and Pb (r = 0.7400) in the mud of twenty examined ponds. From the statistical results it might be suggested that the types of source of the three metals accumulated in the mud of the investigated shrimp ponds are very closely related that is may be for introducing the highly metal polluted water from the industrial sources during culture period are responsible for this situation of higher metal concentration in one hand and natural metals enriched soil that releases toxic and heavy metals by different natural factors on the other hand. The distribution of metals in sediments depends upon several factors as follows: 1) source materials; 2) weathering processes; 3) sediment transport; and 4) dissolution and reaction kinetics (Filipek and Owen, 1979). Awadallah et al. (1994) stated that the mechanisms appear to be responsible for progressive accumulation of trace metals in bottom sediments as following: 1) adsorption or coprecipitation on organic and detrital organic particles; 2) the preferential accumulation of trace metals by benthic organisms. Awadallah et al. (1993) also proposed that sediment pollution with metals is related to the influence of seasonal water discharge, Nile transport particulate matter, and water quality conditions.

**CONCLUSION**

Summarily, it can be concluded that 55, 65, and 5% ponds of four investigated zones are contaminated by As, Cd, and Pb, respectively, according to livestock water quality limits described in Table 1. The overall discussion also implied that Vunh Tau is more contaminated among the four studied coastal areas because as it shows a higher magnitude in the concentration of all metals estimated than that of the rest three locations which was clearly apparent from the frequency distribution pattern. This concentration may primarily be due to industrial pollution as this area is industrially more developed than the remaining three study sites. Likewise, it may also be suggested that all investigated ponds are with permissible limit of As and Pb concentration, whereas 60% ponds are affected with high level of Cd contamination, if the estimated metal content of the mud is compared with the common range of metals for soil as mentioned in Table 1.

The most significant point is that the presence of these toxic metal concentrations in the mud samples may contaminate the shrimp tissue through the process of progressive bioaccumulation. It may be concluded that metal concentration in the mud is an important concern to the coastal aquaculture of Viet Nam which may pose severe impact by bioaccumulation and biomagnification processes of the food chain leading to a negative impact on human health. In this regard, finally, a short period shrimp/fish culture as well as regular monitoring of toxic and heavy metal content in mud as well as introducing water should be undertaken to overcome the risks of bioaccumulation related health hazards from these metals in coastal aquaculture.
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