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The use of intertidal molluscs in the monitoring of heavy metals and organotin compounds in the west coast of Peninsular Malaysia

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Abstract—Over 60% of human activities in the west coast of Peninsular Malaysia are concentrated in the coastal areas. High human activities in the coastal areas may cause hazardous chemical pollution. In this report, intertidal molluscs were used to assess heavy metals pollution in the coastal areas of the west coast of Peninsular Malaysia. Intertidal gastropods such as *Thais*, *Cherithedia* and *Nerita*, and bivalves such as *Anadara*, *Isognomon alatus* and *Perna viridis* have been studied and analysed for their suitability as biomonitoring agents. These species are abundant in the intertidal areas of the west coast of Peninsular Malaysia and represent different groups of habitat, behaviour, physiology and feeding habit. *Perna viridis* and *Isognomon alatus* are good indicators for heavy metals pollution, and *Perna viridis* and *Thais* are good ones for organotins pollution, because they are easily available and analysed, and also able to reflect the quality of the environment. These molluscs can help in the monitoring and management of heavy metals in the coastal ecosystem.

Key words: intertidal mollusks, heavymetals, organotion compounds, peninsulan Malaysia

Introduction

Malaysia is rapidly developing towards being an industrial country. Many heavy and light industries, small and medium industries, and backyard industries have been growing in the last ten years. Agricultural activities involving aquaculture, orchards, oil palm and rubber estates, poultry, cattle and pig farming are also increasing. The increase in industrial and agricultural activities has created new demands in housing, urbanisation, transportation and medication. All these will contribute to the accumulation of hazardous chemicals such as heavy metals, organotin compounds and organochlorine compounds in the environment especially in the aquatic ecosystem.

The Straits of Malacca is among the oldest of the world's most important international navigation waterways. Trade between Europe and the Middle East, and East Asian countries like Japan, South Korea, Hong Kong and China flows through the straits. Over 50,000 cargos pass through the straits every year, including about 20,000 supertankers, about 6,000 ships a month, and at least 300 ships daily. More than 50% of those ships are oil tankers and 25% are cargos. In the year 2005, over 70,000 ships are expected to navigate through this 900 km narrow passage. The Straits is also important as a source of food and livelihood to Malaysia and Indonesia. It is the most important fishing ground in Malaysia, accounting for approximately 70% of total landings of the country (Eng et al. 1989). Human activities such as shipping, industries, agriculture, and the discharging of sediment and pollutant through rivers have posed harmful threats to the coastal environment including mangroves, coral reefs and sea grass which support fishing industries.

Heavy metals and organotins compounds are known as main pollutants in modern life. Especially organotin compounds have been utilised as an active biocide in antifouling paints since the early 1960s. It is well known that organotin compounds released into the water column from coatings applied to boat hulls caused deleterious effects in non-target marine organisms (Laughlin and Linden 1985, Bryan and Gibbs 1991). Among the organotin compounds, tributyltin (TBT) can be concentrated readily in marine organisms from a number of sources involving water, sediment and diet (Bryan and Langston 1992, Langston and Pope 1995, Harino et al. 2000). In most cases, little is known of uptake pathways and processes, or the subtle phylogenetic variation that may occur as a result of habitat and feeding style. Bivalve like clams and mussels are common in faunal suspension feeders, ingesting small detrital particles and phytoplankton, as well as bacteria and micro zooplankton which are suspended in the water column just above the bottom. Tributyltin is known to cause some toxicological effects which in turn affect the population of organisms in a specific habitat. Some specimens of marine fish collected in Malaysian coast show high level on heavy metals and organotin compounds and may be hazardous to Malaysian people (Agusa et al 2005). Continuous monitoring on these hazardous chemicals in the environment and specific ecotoxicological studies on organisms and habitats should be carried out in order to maintain a healthy and productive ecosystem.

The west coast of Peninsular Malaysia comprises about 100,000 hectares and this contributes significantly to coastal productivity. The area is rich in renewable resources due to its productive coastal ecosystems and has extensive cultured fisheries, aquaculture and intertidal molluscs. Among the intertidal molluscs, *Perna viridis*, *Cherithedia obtusa*, *Nerita lineata*, *Isognomon alatus*, *Anadara granosa*, *Telescopium telescopium* are easily available along the west coast of Peninsular Malaysia. The green mussels *Perna viridis* and blood cockles *Anadara granosa* are cultured for human consumption and in some places grow wild. In Malaysia, the green-lipped mussel is a commercial bivalves species although it is not as popular as the cockle *Anadara granosa* among the local people. Other molluscs such as *Telescopium telescopium*, *Cherithedia obtusa* and *Nerita lineata* are also sometimes collected by the locals for consumption.

Since these molluscs are available along the coastal areas of the west coast of Peninsular Malaysia and are easily collected, it can therefore be suggested to be good monitoring agents for chemicals pollution. The intertidal molluscs identified above represent different habitats and behaviours. These differences may reflect the behaviour of heavy metals and organotin compounds intake by the molluscs from their environment. *Perna viridis* and *Isognomon alatus* filter particulate matter from water environment, *Anadara granosa* like other clams may filter organic matter from sediment and *Cherithedia* grazes on the surface of intertidal mudflat while *Nerita* grazes on the rocky areas or roots of *Rhizophora* mangroves.

As for the green-lipped mussel *Perna viridis*, many researches have suggested that it can be used as a bioindicator or biomonitoring agent for chemicals pollution such as heavy metals and organochlorine compounds in this region. Within the Asia-Pacific region, the green-lipped mussel, *Perna viridis* has been established as a biomonitoring agent for these chemicals in Thailand, India, Indonesia, Philippines, and Malaysia (Monirith et al. 2003). In fact, the use of mussels as bioindicators has been developed for years since 1975 by Goldberg in USA and later in Asian region by Phillips (1985). This is due to their wide geographical distribution, sedentary lifestyle, stable population, easy sampling, bioaccumulative and correlative properties with the average pollutants of the environments, tolerance to salinity, resistance to stress of high accumulation of wide range of chemicals, and they provide an assessment of bioavailability. Studies on heavy metals in Malaysian green-lipped mussel were first started in 1980s and in 2000; mussels were used in the Mussel Watch programme to monitor marine pollution in Asian waters (Tanabe 2000). Many molluscs in the intertidal areas of Malaysian coasts fulfill the characteristics of bioindicators. This paper reviews studies of some intertidal mollusks from aspects of the biomonitoring of heavy metals and organotins in the coastal areas of the west coast of Peninsular Malaysia.

Perna viridis, *Isognomon alatus* and other gastropods as a biomonitoring agent for heavy metals

Bioavailability of heavy metals in mussel have been shown to have an ecotoxicological significance in their habitat and reflecting the metal originating from both natural and anthropogenic sources (Rainbow et al. 2004). *Perna viridis* filters out food from water column and has pumping and filtering mechanisms capable of bioaccumulating a wide range of contaminants including heavy metals. They are in general had shown to fulfill the criteria of a good biomonitoring agent. Studies on metals accumulation in *Perna viridis* collected along the west coast of Peninsular Malaysia, show a significant correlation ($P<0.05$) between Cd, Cu and Pb concentrations in total soft tissue of *Perna viridis* and some geochemical fractions of these metals in sediments were observed. This suggests that the total soft tissue of *Perna viridis* is a good biomonitoring agent for Cu, Cd and Pb. Only Zn show lack of relationship between Zn levels in *Perna viridis* and Zn levels in all the geochemical fractions and total Zn. This could possibly due to the ability of *Perna viridis* to regulate Zn from its soft tissue (Yap et al. 2002). There are many factors such as environmental, physiological and genetic factors may affect the accumulation and distribution of metals in the tissue.

Laboratory studies on the accumulation and depuration of heavy metals in *Perna viridis* also show that it can be a good biomonitoring agent (Yap et al. 2004a). Extended studies on the possibility of specific tissues of *Perna viridis* to be good biomonitoring materials were also carried out. The results found that byssus indicates to be more reflective to the heavy metals contaminations in the field environment compared to other soft tissues (Yap et al. 2005). The characteristic of byssus that serves as an organ where mussels attached to the hard substratum makes it easily collected without collecting the mussels. If it can be a better material as a biomonitoring agent for heavy metals, this will make sampling and monitoring easier. Since byssus consists of collagen fibres and maintains a continuous growth, heavy metals might be excreted through the byssus as a process of detoxification by mussels. Thus, the levels of heavy metals in the byssus reflected the heavy metals in the soft tissues and the environment.

Another potential biomonitoring material in *Perna* *viridis* is shell. Metal like Zn usually well regulated by the animals due to it is an essential element. Through the physiological processes Zn excreted through shell. Yap et al 2004b analysed the correlations of Zn levels between shells and the different geochemical fractions in sediments to assess their suitability as a biomonitoring material. As shown in byssus, higher coefficients between shell-geochemical fractions of sediments than between soft tissues-geochemical fractions of sediments indicated that the shell could be used as a better biomonitoring material for Zn. Even though *Perna viridis* in general can be a good biomonitoring agent for heavy metals, the above discussion suggest more studies needed to confirm the tissues that really suitable as a biomonitoring agent for a certain metal.

Flat-tree oysters *Isognomon alatus* inhabit the mangrove areas of the river estuaries. They are easily available as they are attached to the mangrove roots in the estuaries of the west coast of Peninsular Malaysia. Mangroves are well known as breeding places and nursery grounds for marine lives. Contaminants released to the sea pass through the estuaries. Flat-tree oysters living in the estuaries, attached to the mangroves roots are the most strategic biomonitoring agents for the contaminants such as heavy metals. One of the areas that were used to study bioaccumulation of heavy metals in this oyster is in Sepang River (Saed et al. 2004). There are two Sepang Rivers which are known as Sepang Besar and Sepang Kecil. Sepang Besar River is known to receive pig farm effluents which contain high level of Cu and Zn (Ismail and Ramli 1997).

A six-month field study on flat-tree oysters *Isognomon alatus* in contaminated (Sepang Besar) and uncontaminated rivers (Sepang Kecil) show that they are responding to the levels of heavy metals in the environment. Positive correlations were observed between heavy metals concentration in the soft tissues of oysters and their environment. The accumulation rates of heavy metals Cu, Pb, Zn and Cd, might reflect the toxicity behaviours of metals in oyster (Saed et al. 2001). Studies on the accumulation and depuration of heavy metals in flat-tree oyster also show significant correlation with the environment although the depuration rates of metals studied in the laboratory are significantly faster than in the field (Saed et al. 2004). Accumulation rates of metals by oysters under the field conditions were found to be 42.7, 1.7, 0.9 and 0.8 μgg^{-1} $month^{-1}$ for Zn, Cu, Pb and Cd respectively.

Perna viridis and *Isognomon alatus* are filter feeders. They may have a similar habit of food filtration processes. They filter organic and bacterial food sources adhering to particles and resuspended benthic algae. Other organic matters may also be important as food sources. Their ability to regulate filtration rates and ingestion rates may depend upon the quantity and quality of planktons and other particulate matters in the water. This might be influenced by their habitat and nature. *Perna viridis* are usually cultured in an intertidal area which is about 50 to 1000 meters from the shore whilst *Isognomon alatus* are naturally attached to the roots of mangroves near shore lines. These two areas are different in term of water quality, turbidity and current. Therefore, their filtration rates are probably different and the behaviour of metal uptake and toxicity may also be different. The use of the two species of bivalves as biomonitoring agents may be specific to the local habitat.

Cherithedia and *Nerita* are two gastropods that live in different habitats.*Cherithedia* have been observed to graze on the surface of intertidal mud flats, and so do *Telescopium telescopium* whereas *Nerita* can be found to graze on the surface of rocky areas and the surface of mangrove stems or roots. Their habits may influence the behaviour of heavy metals taken into the body system of the snails. Studies on *Cherithedia obtusa* and *Nerita lineata* in Sepang River, also show a positive correlation ($P<0.05$) between heavy metals in the soft tissues of snails compared to the surface sediments. Sepang River at one time was a polluted river due to pig farming activities. Now all pig farming activities in the area were stopped after the outbreak of Nipah Virus. The range of heavy metals in sediment analysed were 10–250 μgg^{-1} for Zn and 3–260 μgg^{-1} for Cu whereas for gastropods, Cu ranged from 20–40 μgg^{-1} and Zn ranged from 15–90 μgg^{-1} in soft tissues of *C. obtusa* and 4–7 μgg^{-1} for Cu and 14–20 μgg^{-1} for Zn in soft tissues of *N. lineata*. *Cherithedia obtusa* accumulates higher level of heavy metals than *N. lineata* (Ismail and Ramli, 1997). Heavy metals accumulate higher in the surface sediments compared to the algae collected from the surfaces of mangrove stems.

Telescopium telescopium may show a similar habit to *Cherithedia*. They graze on the mud flat. *Telescopium telescopium* is bigger in size and uses a larger area compared to *Cherithedia*. Studies on Cu and Zn concentrations in *Telescopium telescopium* collected from Lukut River show high levels of those metals in the soft tissues. Lukut River is also one of the polluted rivers due to human activities. The concentrations of Cu and Zn in surface sediments are within the range of 37 to 100 μgg^{-1} and 100 to 210 μgg^{-1} respectively. Twenty to sixty percent of copper and 40 to 63% of zinc are anthropogenic input and these values are considered high in coastal areas of Peninsular Malaysia. Copper and zinc levels in *Telescopium telescopium* are between 50–60 μgg^{-1} and 35–60 μgg^{-1} respectively. There are some inconsistencies in the correlation of Cu and Zn levels in surface sediments and *Telescopium telescopium* from Lukut River (Ismail and Shafahieh 2005). More studies on the behaviour of Cu and Zn in sediments and *Telescopium telescopium* are needed before suggesting *Telescopium telescopium* as a good monitoring agent.

***Perna viridis* and *Thais* sp. as a biomonitoring agent for butyltin contamination**

High shipping activities in the Straits of Malacca, ports and marinas have been reported to contribute to the contamination of butyltin compounds in the coastal environment. Tributyltin, for example, is one of the hazardous chemicals that caused hazard to marine ecosystems. Tributyltin (TBT) has been used for almost four decades as an effective antifouling agent added in marine paint formulations used on pleasure boats, ships, vessels, docks, fish nets, and also as lumber preservatives and slimicides in cooling system. It is already well-established that shipping activities contribute to the level of TBT in the sea and the coastal environment. Besides ships, many other places such as marinas, harbor structures, and activities such aquacultures and mariculture are using TBT-contained antifouling agents. Tributyltin was developed and first applied in the 1960s as a biocide to prevent the settlement of organisms at the ships' hull.

These organotin compounds also have been used commercially for many years in a variety of applications such as polyvinyl chloride (PVC) stabilizers, industrial catalysts, wood preservatives, and biocides. Industrial activities also contribute to elevated levels of TBT (Takahashi et al. 2000). Contamination by BTs is widely distributed along coastal waters of Malaysia and has been accumulating in various environmental media and aquatic biota.

Since Malaysia is continuously developing and the Straits of Malacca is expected to increase its shipping activities, ports and marinas, BTs contamination along the coastal area of straits is also expected to increase. High levels of TBT may affect the ecosystems and cause hazards to organisms such as imposex, reproduction, deformities, etcetera. Close monitoring on the levels of TBT contamination and their ecotoxicological effects are necessary to be carried out. Monitoring can be done effectively by using bio-monitoring agents or sediments.

Through the Mussel Watch programme in the Asia Pacific region, green-lipped mussels (*Perna viridis*) have been used as an indicator for monitoring TBT contamination in the coastal areas. The study shows that relatively high concentrations of BTs were observed in mussels from the aquaculture areas in the west coast of Peninsular Malaysia. Beside high accumulation of TBT in mussels and fishes, 100% imposex cases were also observed in snails such as *Thais* sp. Imposex is known to be caused by TBT contamination. Contamination of TBT is widespread along the coastal areas. Major sources of BTs are anthropogenic. Indicator species such as bivalves (*Perna viridis*), gastropods (*Thais* sp.) and fish (Java medaka) can be used as good bio-monitoring agents for tributyl tin contamination.

Tong et al. (1996) reported that TBT levels in tissues of random cockle and soft-shell clam samples from local markets were found in the range of$<$0.5 to 3.7 ngg^{-1} wet wt. and the level of TBT in green mussel samples both from market (23.5 ngg^{-1} wet wt) and from mussel farm (14.2 ngg^{-1} wet wt) indicate slight accumulation whereas the level of TBT in sediment samples of Straits of Malacca were found to be in the range of$<$0.7 ngg^{-1} dry wt in unexposed coastal sites to as high as 216.5 ngg^{-1} dry wt for a site within a port area. A few years later, another study on concentrations of TBT in the sediments of the straits has been conducted. They found the level of TBT ranged from 2.8–1100 ngg^{-1} dry wt (Sudaryanto et al. 2004a). Sudaryanto et al. (2004b) also reported TBT levels in fish muscles collected along Straits of Malacca were ranged from 2.8 to 190 ngg^{-1} wet wt.

From the above discussion, it is clear that Malaysian coastal waters are contaminated with organotin compounds and will continue in the future due to its strategic location for sea transportation which link east and west trades. Based on this scenario, even though the use of TBT in paint was banned in Malaysia, continuous monitoring is needed in order to assess sensitive and important localities such as aquaculture farms or nursery grounds. Two molluscs, *Perna viridis* and *Thais* sp. were collected along the west coast of Peninsular Malaysia (Fig. 1) and investigated for monitoring the contamination of organotin compounds.

Sudaryanto et al. (2002) reported TBT concentrations in green mussels (*Perna viridis*) and sediment from some localities along west coast of Peninsular Malaysia to be high. Organotin compounds are the major component of total tin measured in both *Perna viridis* and sediment. The concentration of TBT varied widely depending on the locations. High organotin contamination mainly occurred close to pleasure boating activities especially in or near large harbours and major shipping traffic areas (Penang and Johor). The narrowest area of the straits (Johor) has the highest concentration of TBT. Lower TBT concentrations were reported in samples taken from other stations with low maritime activities such as recreational beach, fisheries with small boats and small scale aquaculture. The levels of TBT in *Perna viridis* seem to reflect the TBT concentration in the sediment. According to Sudaryanto et al. (2002) levels of TBT in some polluted areas are comparable to other polluted areas in Hong Kong, India, Philippines and Thailand.

As an evidence of ecotoxicological impact of TBT, imposex phenomenon has been recorded in gastropods in at least 72 species of gastropods, belonging to 49 genera (Oehlman et al. 1991). Among them is *Thais* sp. that can be found in Malaysia. Imposex cases in *Thais kieneri* and *Thais savignyi* are observed in coastal areas of Ambon Island, Indonesia (Evans et al. 1995), in three species of *Thais* from Singapore (Tan, 1997), and in *Thais clavigera* from oyster marine culture areas in Taiwan (Hung et al. 2001). Imposex have been recommended as suitable bioindicators of TBT pollution.

Ismail et al. (2004) reported that nearly 100% of female

Thais collected along the Straits of Malacca (Fig. 1) display imposex. The analyses of snails show that 100% of *Thais tuberrosa*, *Thais bitubercularis* and *Thais hippocastanum* and over 96% of *Thais gradat*a collected from the intertidal areas of the Straits of Malacca show imposex. Even though there were no detailed data on TBT levels in sediment to compare with the degree of imposex in the Straits of Malacca, the imposex trend was similar with the results of butyltin, specifically TBT monitoring survey in this country as reported by other studies (Tong et al. 1996, Sudaryanto et al. 2002 and Sudaryanto et al. 2004a). The locations where high incidence of imposex was observed are identical with those found to be highly contaminated by TBT.

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