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Lithium Levels in Peninsular Malaysian Coastal Areas: An Assessment Based on Mangrove Snail *Nerita lineata* and Surface Sediments

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ABSTRACT

Sampling for the mangrove snails, *Nerita lineata*, and surface sediments was carried out from nine geographical sites of Peninsular Malaysia in April 2011. The concentrations of Lithium (Li) were determined in the shells, opercula and soft tissues of *N. lineata* and in the surface sediments by using ICP-MS. The ranges of Li concentrations (µg/g dry weight) were found to be 0.107-0.283 for shells, 0.021-0.177 for opercula and 0.011-0.634 for total soft tissues of *N. lineata*. For sediments, Li ranges were found to be between 21.84-146.22 µg/g dry weight). The distribution of Li was found to be: sediment> ST> shell> opercula. The Li sediment data in the present study were comparable with the results of Li contaminated sediments which had been previously reported in the literature and higher than those of continental crust materials and igneous rocks. There was no significant correlation (P>0.05) for the Li levels between the sediments. The snails (shells, opercula and soft tissues) and this indicated that Li is an essential metal for metabolism and thus is being regulated in the body of the snail.

Keywords: Li, biomonitoring, snails

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INTRODUCTION

Lithium (Li) is the lightest of the alkali metals belonging to group 1 of the Periodic Table, along with Na, K, Rb and Cs. Li has been extensively utilized in a wide number of industrial, scientific, and clinical

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applications (Hu, 2013). In particular, the commonly known usage of Li in our daily life involves Li batteries. Lithium batteries, both rechargeable and nonrechargeable, are now used in a very wide range of devices. This is because of the fact that Li has the greatest electrochemical potential of any metal (~3 V) (Hu 2012). However, its anthropogenic use result in Li waste in our environment.

All trace metals occur naturally in the environment but they may exhibit high levels due to human activities, which could cause harmful effects on the marine coastal ecosystem (Bodin *et al.,* 2013). In this study, surface sediments are focused upon because they are the ultimate sink for trace metals and can reflect the degree of pollution of an environment (Yap *et al.,* 2009). In the study of sedimentary geochemistry, Li is generally considered a conservative element in marine sediments and in metal enrichment studies. Therefore, it is often used to normalize metal concentrations to compensate for the natural textural and mineralogical variability (Aloupi & Angelidis, 2001).

Seafood such as mollusks, collected from mangrove areas, have been of worldwide concern nowadays due to trace element bioaccumulation in the tissues (Guerin *et al.,* 2011; Palpandi & Kesavan 2012; Bodin *et al.,* 2013; Fung *et al.,* 2013; Yap & Cheng, 2013). Most gastropod species have been employed as biomonitors of trace metal pollution in the coastal environment (Pearce & Mann, 2006) due to their abilities to accumulate trace metals in their tissues (Rainbow, 2002) on exposure to various sources of anthropogenic pollution (Wang *et al.,* 2005) and ubiquitous distribution (Oehlmann & Schulte-Oehlmann 2003) in marine coastal ecosystems and easy sampling.

Since there has been no report on Li in snails and sediments from Malaysia, the objective of this study was to provide the baseline levels of Li in the mangrove snail *Nerita lineata* and surface sediments from Peninsular Malaysia.

MATERIALS AND METHODS

The snails, *N. lineata*, and sediments were randomly collected from nine sampling sites in the mangrove areas of Peninsular Malaysia in April 2011 (see Fig.1). Sampling information is given in Table 1. About 20 individuals of the snails of similar size were selected from each sampling site, dissected and pooled into soft tissues, opercula and shells. The snails' gut contents were not depurated in this study, as suggested by Yap *et al.* (2010).

The dissected tissue parts were then dried at 60°C until constant dry weights. Three aliquots of each tissue part were measured, with an approximate amount of 0.5 g each, and placed in the TFM vessels. A mixture of acids (7 ml of HNO_3 65% + 1 ml H_2O_2 30%) was added into the dried samples before inserting them into the microwave cavity. For the sediment samples, they were dried at 60°C until constant dry weights and sifted using a stainless steel sift of 63µm in mesh. Triplicates of 0.5 g each were obtained from the sampling sites and placed in TFM vessels. A mixture of acids (9 ml of HCl + 3 ml of HNO_3 65%) was added into the dried sediment samples.

For digestion of snail and sediment samples, the microwave digester used was the Milestone ETHOS labstation with easyWAVE or easyCONTROL software HPR1000/10S high pressure segmented rotor. To digest the snail samples, the microwave digester was set to increase the temperature to 200°C for the first 10 minutes and maintained at 200°C for the following 20 minutes, with the application of 1000 W of microwave power. Similar procedures from the snail tissue preparation were also applied to the sediments but with the temperature raised to 200°C for the first 10 minutes and maintained at 200°C for the following 15 minutes. The samples were left in the microwave digester to cool down to room temperature for 10 minutes after the digestion had been completed. The digested samples were then diluted to 100 ml with double distilled water (DDW) and filtered with Whatman No. 1 filter paper before they were stored for metal analysis. All the

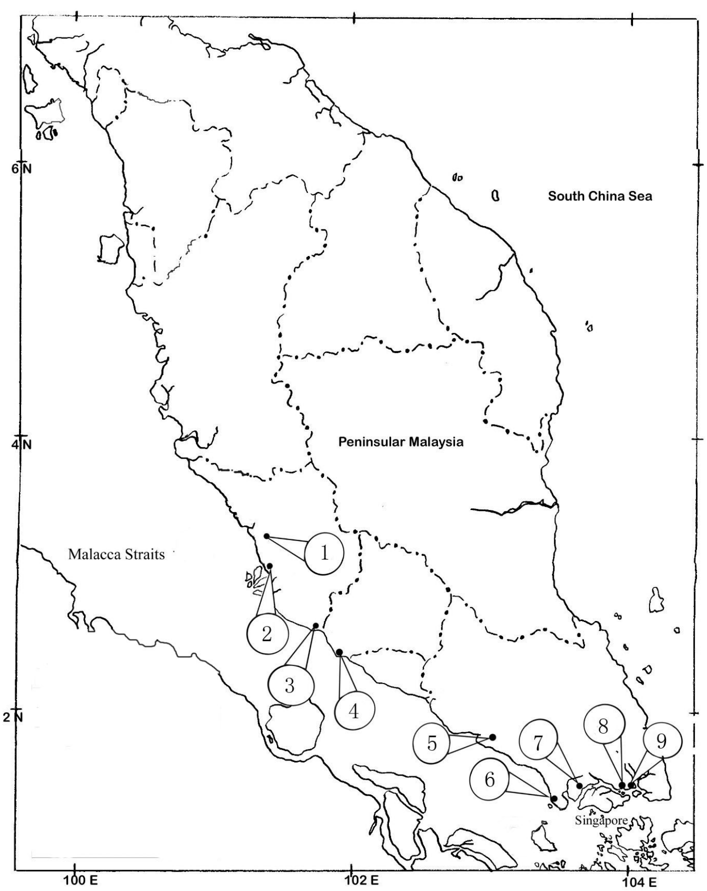

Fig.1: Sampling map. (Sites: 1=SJanggut; 2=JKetam; 3=Sepang; 4=Lukut; 5=SAyam; 6=Kukup; 7=KSMelayu; 8=KPPuteh; 9=TLangsat)

TABLE 1
Descriptions of the sampling sites

No	Sampling Sites	Date	GPS	Shell length (mm)	Site activities
1	Sungai Janggut (SJanggut)	8-Apr-11	N 03° 10.307’ E 101° 18.524'	25.19 - 33.82	It is a jetty in mangrove area with fishing village nearby. Water irrigation was also observed here.
2	Pulau Ketam (JKetam)	8-Apr-11	N 03° 00.567' E 101° 21.649'	25.28 - 28.34	A jetty to Pulau Ketam, fishing and shipping activities were observed here. Construction work (building) were being carried out during sampling.
3	Sepang	8-Apr-11	N 02° 36.076' E 101° 42.565'	19.56 - 27.87	Nearby housing and mangrove area, prawn and mussel aquaculture, fishing and shipping activities were observed here.
4	Lukut	8-Apr-11	N 02° 34.511' E 101°47.529"	21.58 - 28.64	Industrial and urban area. It is a fishing village with mooring activities.
5	Sungai Ayam (SAyam)	15-Apr-11	02° 34.423’ E 102° 02.346'	23.78 - 30.4	Fishing village with water irrigation. Massive amount of domestic waste were observed in this area. Mussel aquaculture, fishing and shipping are the main activities here.
6	Kukup	15-Apr-11	N 01° 19.471’ E 103° 26.521'	23.08 - 30.82	It is a port and tourist attraction site with restaurants and resorts. The main activity here are fishing and shipping.
7	Kampung Sungai Melayu (KSMelayu)	16-Apr-11	N 01° 27.043’ E 103° 41.699'	21.05 - 26.44	A jetty where the main activities here are fishing and shipping. Mussel aquaculture was observed here.
8	Kampung Pasir Puteh (KPPuteh)	16-Apr-11	N 01° 26.082' E 103° 56.094'	22.54 - 30.91	It’s a jetty with restaurants and a fishing village. Shipping and industrial activities were observed here.
9	Tanjung Langsat (TLangsat)	16-Apr-11	N 01° 28.190’ E 104° 0.041'	24.46 - 31.46	It’s a jetty with restaurants and a fishing village. Shipping and industrial activities were observed here.

prepared samples were determined for Li by using the Perkin Elmer SCIEX ELAN DRC-e ICP-MS with the limit of detection of < 0.1 ppt.

For quality control, the precision and quality of the method were checked with Certified Reference Material for Marine Sediment (MESS-3) (Li certified value: 73.6 ± 5.2 µg/g dry weight; Li measured value: 66.5 ± 5.0 µg/g dry weight) with an obtained percentage of recovery and standard deviation of 90.3 ± 6.8 %.

For statistical analysis, One-way ANOVA Student-Newman-Keuls test was carried out to determine the differences between the metal levels in the different tissues of the *N. lineata* and the sediments. Pearson's correlation coefficient (IBM SPSS Statistics version 19) and Single Linkage Euclidean distances (STATISTICA version 8.0) were used to determine the relationships of Li between the snails and sediments.

In this study, the distributions of Li levels between the snails (three parts) and in the associated surface sediments, estimated based on the biota-sediment accumulation factors (BSAF), were calculated (Szefer *et al.,* 1999). The formula used is given as follows:

$$\text{BSAF}= \frac{Cx}{Cs}$$

where C*x* and C*s* are the mean Li concentrations in the different parts of the snails and in the surface sediment, respectively.

RESULTS AND DISCUSSION

The Li concentrations of the snails and sediments are shown in Fig.2. The ranges of the Li concentrations (µg/g dry weight) were 0.107-0.283 (mean: 0.195 ± 0.020) for shells, 0.021-0.177 (mean: 0.099 ± 0.018) for opercula and 0.011-0.634 (mean: 0.310 ± 0.062) for total soft tissues. Interestingly, Sepang recorded the highest Li levels in both the shells and opercula, while KSMelayu recorded the highest levels of Li in soft tissues. The present ranges (0.107-0.283 µg/g dry weight) of Li in the snail shells were below those of the *Mytilus edulis* shells sampled from the Polish coast of the Baltic Sea, which were reported as being 0.275-3.484 µg/g dry weight (Protasowicki *et al.,* 2008). However, comparison of Li in the soft tissues and opercula of mollusks is difficult to be made due to the lack of similar Li data in the literature.

As for sediments, the ranges of Li were between 21.84-146.22 (mean: 50.44) with SAyam recording the highest and Lukut the lowest. The data of SAyam were reported by Yap *et al.* (2009) with elevated levels of Pb due to hidden industrial activities in the nearby area, which could also be the source of the high level of Li in this site. As for KPPuteh, with the second highest level of Li, it is known to be surrounded by heavy anthropogenic activities that could contribute to the high level of Li in this area (Yap *et al.,* 2004). The other sites were basically fishing villages or aquaculture sites that had lower levels of Li pollution (see Table 1).

Therefore, Li concentrations generally follow sediment> ST> shell> opercula. In order to estimate the status of Li, the present ranges of Li in the sediment were compared

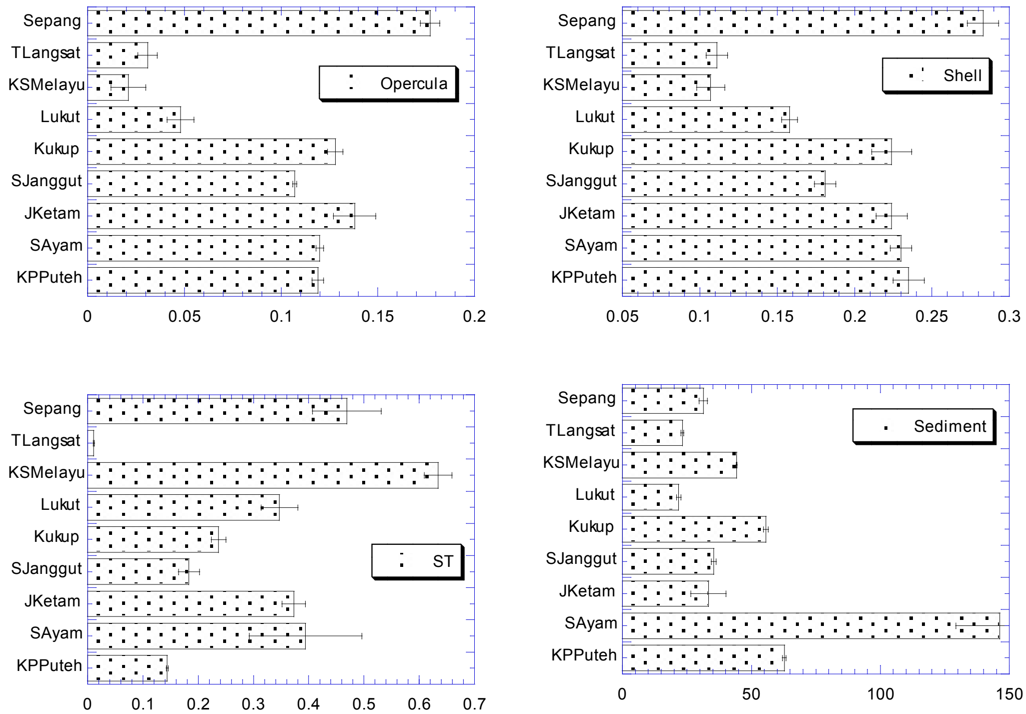

Fig.2: Lithium levels (mean ± SE, µg/g dry weight) in the shells, opercula and soft tissues of *Nerita lineata*, and surface sediments collected from nine sampling sites in Peninsular Malaysian mangrove areas.

to the background Li value, upper and lower continental crust materials (CCM) for Li which were reported as being 22 and 13 µg/g dry weight, respectively (Wedepohl, 1995), and the Li level in the igneous rock (32.0 µg/g dry weight) (Vinogradov, 1962). The ranges of Li in the current work appeared to be higher than CCM and the igneous rocks, and thus, posed an environmental concern whether our Malaysian mangrove area is facing Li pollution.

The present Li ranges were further compared to the Li data in the sediments reported in the literature. Miranda-Avilés *et al.* (2012) reported the Li levels as being 125.10 µg/g dry weight (0-20 cm) and 58.03 µg/g dry weight (0-70 cm) for metal-contaminated overbank-sediment and pristine overbank-sediment, respectively, from Guanajuato City, Mexico. Christiansen *et al.* (2009) reported the Li content as being high (>50 µg/g dry weight) in Skagerrak and northern Kattegat, whereas the sediments in the Belt area and the western part of the Baltic Sea have low concentrations at 20-40 µg/g dry weight. The present ranges (21.84-146.22 µg/g dry weight) of Li in the surface sediments were higher than those reported for Medway and Swale (6.00 - 39.00 µg/g dry weight) (Spencer *et al.,* 2006), South China Sea (27.6 - 51.6 µg/g dry weight) (Rezaee *et al.,* 2011) and Senegal (3.50 -

8.40 µg/g dry weight) (Bodin *et al.,* 2013). All the above comparisons showed that the ranges of Li in the current work were close to the known Li contaminated range. Knutzen and Skei (1990) defined "high background levels" for heavy metals in sediments and biota to indicate areas affected by heavy metal pollution.

It appeared that the positive relationships are weak for shell-sediment (R= 0.322), opercula-sediment (R= 0.240) and ST-sediment (R= 0.148), but these three pair-wise values were not significantly (P> 0.05) correlated. These insignificant correlation (P>0.05) for the Li levels between the sediments and the snails (shells, opercula and soft tissues) indicated Li regulation in the tissues of *N. lineate*, Therefore, it could poorly reflect the ambient Li level. Our finding was supported by that reported by Templeman *et al.* (2010) in which Li appeared to be actively regulated within the tissues of the benthic jellyfish, *Cassiopea* sp. collected from the northern and eastern coast of Australia. The available experimental evidence now appears to be sufficient to accept Li as being essentially important for humans and the metal is normally present in all organs and tissues (Schrauzer, 2002). Previous knowledge indicated that essential metals are generally regulated in the tissues of mollusks (Rainbow, 1995) and the present insignificant relationship of Li between snails and sediments is no exception.

The BSAF values are presented in Table 2. It was found that all the BSAF values were below 1.00 (0.05-1.50 × 10^{-2} for soft tissue/sediment, 0.05-0.56 × 10^{-2} for opercula/sediment and 0.16-0.90 × 10^{-2} for shell/sediment). Therefore, according to Dallinger (1993), all the values of BCF are categorized as deconcentrators. Moreover, the differences of the Li levels between the three parts of the snails and the surface sediment are significant (P< 0.001, T-test). Therefore, the snails are generally not good bioaccumulators of Li and the present BSAF values showed that Li was not significantly bioaccumulated in the *Nerita* from their environmental habitats.

TABLE 2
Biota-sediment accumulation factor (BSAF) values (× 10^{-2}); Li concentrations between the snail parts (soft tissues (ST), opercula (Oper), and shell) and surface sediment (SED)

Sites	ST/SED	Oper/SED	Shell/SED
KPPuteh	0.23	0.19	0.37
SAyam	0.27	0.08	0.16
JKetam	1.12	0.41	0.67
SJanggut	0.52	0.30	0.51
Kukup	0.43	0.23	0.40
Lukut	1.50	0.21	0.68
KSMelayu	1.43	0.05	0.24
TLangsat	0.05	0.14	0.51
Sepang	1.49	0.56	0.90

CONCLUSION

This study serves to provide the baseline data of Li levels in the shells, opercula and total soft tissues of *N. lineata* and the surface sediment from Peninsular Malaysia. The present insignificant relationships of Li between the snails and sediment showed that Li is an essential metal for metabolism and thus it is being regulated in the body of the snails. The fact that the snails are categorized as deconcentrators indicated that further studies are needed to justify them as being reliably employed as good biomonitors of Li pollution.

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