



Land use Change Impacts on Heavy Metal Sedimentation in Mangrove Wetlands—A Case Study in Dongzhai Harbor of Hainan, China

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Abstract When human activities change land use patterns, many environment elements are being changed accordingly. Well understanding the links between various changes could help to achieve ecological protection through adjusting land use pattern. Mangrove is one of the most valuable ecosystems that have been changed dramatically during the recent decades. Based on Remote Sensing and Geography Information System, along with soil sample determination, we present a quantitative study about the correlations between land use changes and the variations in heavy metal contents in mangrove soil sediments of Dongzhai Harbor in China. The results showed that the indices depicting the landscape pattern characteristics had been changed remarkably during 1988 to 2007. Meanwhile, the heavy metal contents had been increasing significantly in mangrove soil. The correlations between landscape indices and metal contents were analyzed statistically. It was found that the metal contents were proportional to the values of main indices, revealing that both the landscape fragmentation and patch shape alternation were closely related to the metal accumulation. Further analyses showed that

increasing of aquaculture and residential regions could be responsible for the changes in land use patterns, which subsequently altered the distribution patterns of heavy metal sedimentation in mangrove wetlands.

Keywords Mangroves · Heavy metal sedimentation · Land use changes · Landscape fragmentation

Introduction

Mangroves thrive along the coastlines throughout most of the tropical and subtropical areas (Lin 1997). These inter-tidal forests play important ecological roles in the purification, coastline protection, carbon sinks and biodiversity nursery (Costanza et al. 1997; Barbier et al. 2010). Despite their enormous ecosystem functions, mangroves have been threatened by the rapid growing of social economics during recent decades all over the world (Stone 2006). From 1980 to 2000, over a third of total mangrove area (IUCN) were lost in India, Indonesia, Sri Lanka, Thailand and China, the clearance for settlements and conversion to farmlands were considered to be the top two reasons for the declines (Polasky et al. 2011), meanwhile ecological factors in these mangrove wetlands, such as water quality, community structure, physical and chemical properties of soil, had also been changed dramatically (Barnes et al. 2011, and Zang et al. 2011).

The natural mangroves have long been used as an effective sink for wastewater (Dunbabin and Bowmer 1992). Under both aerobic and anaerobic conditions, the mangrove wetlands contribute considerably to the heavy metals removal from wastewater (Weis and Weis 2004), in the meantime, an increasing content of heavy metals sedimentation in the soil were observed in many mangrove wetlands (Tam and Wong 1995; Xin 2005; Wang et al. 2011). The changes of land use/

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land cover(LU/LC) around mangrove areas were considered to be responsible for the heavy metals increasing (Ivezić et al. 2011). Among various human activities, urban expansion and aquaculture that destroyed mangrove forest and discharged sewage into mangrove, were proved to be the main responsible factors for the increasing (Ren et al. 2011).

Relationships between land use and ecological factor changes were noticed in many fields. Dong et al. (2008) found land use change led to the increasing of greenhouse gases in U.S. croplands; McKinley et al. (2011) studied the influence of land-use change on fish distribution in estuarine; Northington et al. (2011) estimated the eco-function of restored mangrove in Virginia coalfield streams; Billings (2006) discussed the links between land-use change and soil organic matter dynamics in forest/grass ecotone. Sowana studied the influences on soil contamination imposed by the coastal land use changes in Thailand (Sowana et al. 2010). There are also many studies concerning the changes happened in heavy metal sedimentation in mangrove. Zhou (Zhou et al. 2010) contrasted the changes of Pb, Cu, Zn and Cr before and after the mangrove reforestation in Yifeng Estuary of China; Essien et al. (2009) studied the heavy metal sedimentation changes in Qua Iboe Estuary mangrove since sewage discharged into mangrove wetlands. Many studies showed that heavy metal sedimentation in mangrove increased in recent decades. While, the quantitative study of links between land use change and heavy metal sedimentation has not been considered.

In this study, we took Dongzhai Harbor, the first national reserve of mangrove wetlands in China, as example to discuss the influence of land use changes caused by human activities on heavy metal sedimentation in mangrove wetlands through

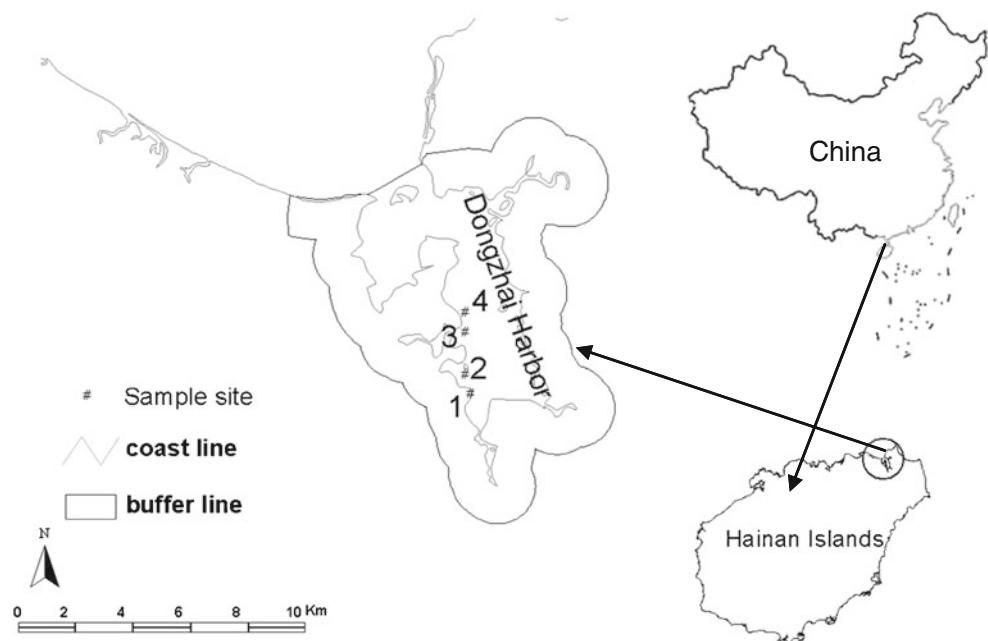
analyzing correlations of changes between landscape indices and heavy metals sedimentation during recent two decades.

Methods

Study Area

The study was conducted in Dongzhai Harbor mangrove wetland, Hainan Island, China (N 19°51'~20°01', E 110°30'~110°37'). The mangrove in the study area has experienced the decreasing, increasing and centroid moving during the past 20 years; therefore it is necessary to delimit the study scope (Xin 2009). In order to make clear the relations between changes of land use and heavy metals sedimentation in mangrove soil, the areas with high human activity intensity should be included in the study. Based on Remote Sensing interpretation and field investigation, we found all natural and artificial mangroves, most of new aquaculture ponds and farmlands, and main residential areas located within a 2 km width buffer zone along the coast line (the average monthly low tide level of Dongzhai Harbor is 0.99 m–1.32 m, so we took 1 m contours as datum coast line), which we delimited as our study area (Fig. 1). It includes the area of Dongzhai Harbor Mangrove Nature Reserve and main human activity areas nearby. The total area of selected buffer belt is 232.862 km². There are 27 rivers of different sizes joining into the sea within this gulf. 19 plant families and 35 mangrove species are found in this area, among them, *Bruguiera gymnorhiza*, *Bruguiera sexangula*, *Kandelia obovata*, *Ceriops taga* and *Aegiceras corniculatum* are main making up species.

Fig. 1 The study area of Dongzhai Harbor mangrove wetland of Hainan, China



Land use Mapping Method

Combined with the ground survey, 10-m resolution SPOT-5 images of the year 2007 and 30-m resolution Landsat-TM images of 1987 and 1999 were used to interpret landscape maps of the study area. ArcGIS9.2 were used in classification and mapping. According to the level II of the national land use 21 standard (GB/T 21010-2007 (Current Land Use Classification of National Standard, 22 PRC)), the land use patterns in the study area were classified into 6 types, i.e., mangroves, farmland, construction, nature water, ponds and other forest.

Land use Changing Analysis Method

The spatial pattern analysis software Fragstats3.3 was used to calculate the landscape pattern index. There were two levels of index, i.e., landscape-level and class-level (Wu 2002). Totally 8 indices of 2 levels were used here (see Table 1)

Sample Collection Method

Four soil sample sites were set in the study area (Fig. 1); six soil samples from the different mangrove communities at four

sites were collected in three replicates using Peterson Bottom Grab in April of 2002 and 2010. The collected soil samples were sealed separately in labeled polythene bags and transported immediately to the lab for analyses. In laboratory, the soil samples were dried at 80 °C for 12 h, then grounded and sieved by 100 mesh sieves. The powdered samples were stored in bottles and preserved for further analysis (Essien et al. 2009).

Heavy Metal Determination Method

0.5 g soil sample (a bit distilled water was required for wetting the soil sample) was mixed with 1 mL HCL in a 30 mL PTFE crucible. After 1 h, 4 mL HNO₃, 1 mL HF and 1 mL HClO₄ was added subsequently to the soil sample. After 15 min standing, the crucible was placed onto an electrical heating plate until a large amount of white dense smoke coming out. Then, the crucible lid was removed and the crucible was shaken to volatilize the silicide. The crucible was allowed to cool at the room temperature until the content was dry. After dissolved with 1 mL 50 % HNO₃, the leading solution was put into a 50 mL flask. 5 mL HNO₃ was added and the content was metered by using ionized water. Each sample was repeated twice and the blank digestion (the control) was also prepared (Jara-Marini et al. 2008).

Table 1 Landscape indices used in landscape pattern analysis in study area

Index(ABBR)	Full name	Computational formula	Ecological meaning
NP(n)	number of patches	$NP = n_i$	Number of patches in a landscape; $NP \geq 1$
PD(n/100 km ²)	patch density	$PD = \frac{N}{A} \times 100$	Density of patches for all types within the landscape; $PD > 0$
LPI (%)	largest patch index	$LPI = \frac{Max_{j=1}^n(a_{ij})}{A} \times 100$	Percentage of the total landscape area comprised of the largest patch; $0 < LPI \leq 100$
CONTAG	contagion index	$CONT = \left[1 + \sum_{i=1}^m \sum_{j=1}^n \frac{P_j \ln(P_j)}{2 \ln(m)} \right] (100)$	Coherent and dispersal between patches; $0 < CONT \leq 100$
SHDI	Shannon diversity index	$SHDI = -\sum_{i=1}^m P_i \times \ln P_i$	Complex degree of landscape; $SHDI \geq 0$
LSI	landscape shape index	$LSI = \frac{0.25E}{\sqrt{A}}$	Complexity of patch shapes; $LSI \geq 1$
DLFD	Double log fractal dimension	$DLFD = \frac{\left\{ \sum_{i=1}^m \sum_{j=1}^n [\ln(P_{ij}) \ln(a_{ij})] \right\} - \left[\sum_{i=1}^m \sum_{j=1}^n [\ln(a_{ij})] \right]}{\left\{ \sum_{i=1}^m \sum_{j=1}^n [\ln(P_{ij}^2)] \right\} - \left[\sum_{i=1}^m \sum_{j=1}^n [\ln(P_{ij})]^2 \right]}$	Patches border complexity of a land use type; $1 \leq DLFD \leq 2$
MNN	Mean nearest neighbor distance	$MNN = \frac{\sum_{i=1}^m \sum_{j=1}^n h_{ij}}{N'}$	Fragmentation degree of a single land use type; $MNN > 0$

Data Collection and Statistical Method

Earlier soil data were cited from the literature which the soil samples were collected in 1992 in Dongzhai Harbor of Hainan, China (Zhang 1997).

Data statistics and analysis were conducted using SPSS16.0 analysis software and Excel2007.

Results

Land use Changes

During the past 20 years, the land use patterns in Dongzhai Harbor of Hainan, China had experienced the significant changes (Fig. 2). The aquaculture pond area increased drastically from 0.2 km² in 1987 to 26.716 km² in 2007, and the similar increase was observed in the construction area too. In contrast, the mangrove area decreased from 23.318 km² in 1987 to 18.473 km² in 1997, followed by a slight increase to 18.578 km² in 2007. Meanwhile, the nature waters, arable lands and other forest decreased by 14.4 %, 10.4 % and 39.8 % respectively (Table 2).

Landscape Spatial Pattern Changes

Spatial pattern changes could be reflected by landscape indices on both landscape-level and class-level.

On the landscape-level (see Table 3), the NP, PD and LSI that depict the patch number and shape complexity increased remarkably, whereas shape indices, including the LPI and CONTAG decreased. The SHDI characterizing the landscape diversity increased slightly.

On the class-level (see Table 4), NP and LSI, the indices characterizing the mangrove fragmentation, increased dramatically over the past 20 years, while the shape indices, namely DFLD, MNN and CONT, decreased obviously.

Heavy Metal Sedimentation in Soil

The heavy metals Cu, Pb, Zn, Cr, Cd, As, Hg and Ni in the soil samples of different years were detected. Only three metals, i.e. Cu, Zn and Pb, which could be found varied in every soil sample, were recorded. An increase in Cu, Zn and Pb contents at each sample site was clearly evident from the year 1992 to 2010. According to South coastal saline benchmark of China, the background value of Cu, Zn and Pb in the soil of study area were 10.50±5.18 ug/g, 51.90±21.60 ug/g and 26.50±12.40 ug/g separately. The average Cu content was 9.29±0.51 ug/g in 1992, 14.31±5.02 ug/g in 2002, and 30.74±5.57 ug/g in 2010. The average Pb content was 13.03±2.52 ug/g in 1992, 31.66±5.36 ug/g in 2002, and 45.92±5.03 ug/g in 2010; the average Zn content increased much more rapidly than Cu and Pb: it was 43.38±5.22 ug/g in 1992, 65.28±5.00 ug/g in 2002, and 269.49±25.20 ug/g in 2010. (Figure 3).

Correlation Analysis of Landscape Indices and Heavy Metal Sedimentation

Correlations between landscape indices and the heavy metal sedimentation data were analyzed via Pearson correlation using SPSS 16.0. Significant correlations appeared between heavy metal and landscape indices, including NP, PD, SHDI, AREA, MNN and CONT (see Tables 5 and 6).

Fig. 2 Land use patterns of the year 1987, 1999 and 2007 in Dongzhai Harbor mangrove wetland of Hainan, China (MANG: mangrove forest; ARAL: arable land; NATW: natural water area; OTHF: other forest; CONA: construction area; ARTP: aquaculture ponds)

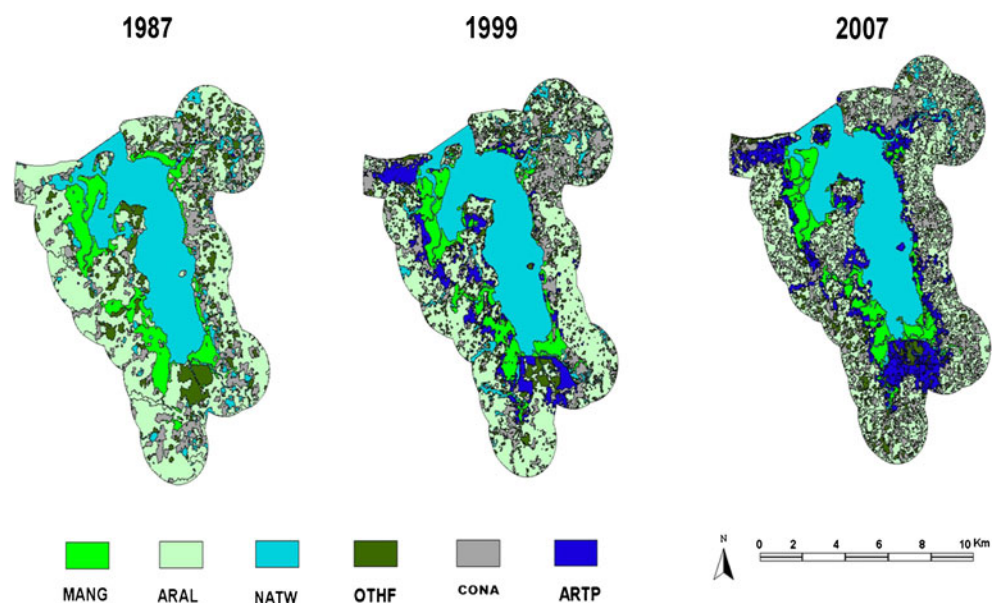


Table 2 Area changes for different land use in Dongzhai Harbor mangrove wetland of Hainan, China (km^2)

Year	ARAL	NATW	OTHF	RESA	ARTP	MANG
1987	97.613	60.037	20.315	27.376	0.200	23.318
1999	86.707	57.872	17.512	31.058	17.237	18.473
2007	87.458	51.366	12.239	32.502	26.716	18.578

MANG mangrove forest; ARAL arable land; NATW natural water area; OTHF other forest; RESA residential area; ARTP aquaculture ponds

Discussion

Land use Change Impacts on Landscape Pattern in Study Area

On landscape-level, the increase in total patch number (NP) and patch density (PD) as well as the decrease in the largest patch index(LPI) and Contagion index(CONT) indicated that a process of landscape fragmentation had occurred in the entire study area over the past two decades. Data analyses showed that the Correlation Coefficients exceeded 0.95 ($df=4$; $P<0.01$) between the indices of NP and PD and the area augmentation of the aquaculture ponds and construction. That is to say, the increase in the aquaculture and construction in the study area contributed primarily to the rise in the NP and PD. The decrease in natural water area led to the LPI decrease ($r=0.96$; $df=4$; $P<0.01$), and mangrove and residential area were both correlated to CONT ($r=-0.99$ and $r=0.94$ separately; $df=4$; $P<0.01$), which can be ascribed to the large natural patches being split into small pieces and the continuity of same type patches being reduced by artificial ponds and constructions.

On the mangrove class-level, it is obviously that the mangrove had experienced a sharp cutting down and fragmentation from 1987 to 2007. Mangrove AREA decreased and NP increased, similarly, MNN and CONT decreased. Correlation analysis showed that the AREA changes were negative related to the changes of aquaculture ponds and constructions ($P<0.05$). Although the mangrove area slightly increased from 1999 to 2007 due to twice *Sonneratia apetala* planting in 1985 and 1999 respectively, the shape indices such as

Table 3 Landscape-level indices of different years in Dongzhai Harbor mangrove wetland of Hainan, China

Index	1987	1999	2007
NP(N)	970	1699	1934
PD(N/km^2)	423,841.8	579,328.8	845,061.8
LPI	22.080	22.108	21.605
LSI	20.870	30.970	29.671
CONTAG	50.022	41.616	42.324
SHDI	1.422	1.581	1.591

Table 4 Mangrove class-level indices of different years in Dongzhai Harbor mangrove wetland of Hainan, China

Index	1987	1999	2007
AREA(km^2)	23.318	18.473	18.578
NP(number)	54	88	84
LSI	11.497	13.871	15.440
DLPD	1.366	1.342	1.042
MNN	92.457	89.788	88.656
CONT	93.067	90.454	89.312

LSI and DLPD kept changing. The increase in LSI indicated that the patch shapes were approached to the square over the time, while the decrease in DLPD implied that the boundary of mangrove patch was gradually simplified. There were probably two reasons: firstly the narrow band of the mangrove was cut by the artificial land use types possessing smooth boundaries such as the ponds; secondly the plantation areas that increased since 1999 may change the narrow shape of mangrove patches.

Impacts of Land use Changes on Heavy Metal Sedimentation

According to correlation analysis between landscape index and heavy metal sedimentation, significant correlation could be seen on both landscape and mangrove class level.

On the landscape level, both NP and PD were closely related to the levels of Cu, Zn and Pb (at 0.01 and 0.05 significant difference level, respectively). It indicated that an increase in the patch density, which was the direct consequence of the aquaculture ponds and construction area

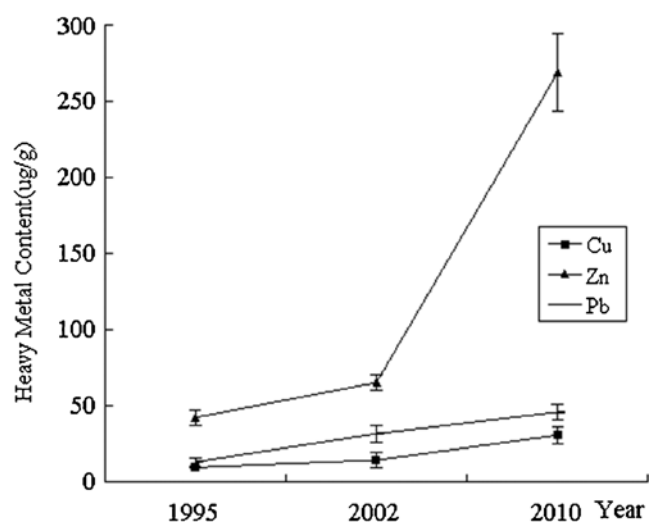
**Fig. 3** The soil contents of Cu, Zn and Pb in different years at Dongzhai Harbor Mangrove Wetlands of Hainan, China

Table 5 Pearson coefficient of correlation between the landscape-level indices and heavy metal contents at Dongzhai Harbor mangrove wetland of Hainan, China

	NP	PD(N/hm ²)	LPI	LSI	CONTAG	SHDI
Cu(ug/g)	0.8332 ^b	0.9890 ^a	-0.9624	0.5896	-0.6233 ^a	0.7186 ^a
Pb(ug/g)	0.9779 ^b	0.9744 ^a	-0.7961 ^a	0.8445	-0.8665	0.9234 ^b
Zn(ug/g)	0.7521 ^b	0.9605 ^a	-0.9900	0.4766	-0.5136	0.6195

^a and ^b indicate correlation is significant at 0.05 and 0.01 probability levels, respectively

increase, contributed significantly to the increase in Cu, Pb and Zn sedimentation contents. As Cu and Zn were widely used in the aquaculture feeds, it was reasonable to think that the surplus feed and feces might be an important source of Zn and Cu pollution in water. Snow et al. (2008) announced that traditional aquaculture was the main source of heavy metals in Nova Scotia wetlands; Cai et al. (2010) reported that feed additives was one of the most important source of heavy metals in aquaculture sewage in China. The aquaculture in the study area had been becoming flourishing since 1989, and the significant profits from aquaculture have attracted a large number of investors. During the first decade, enlarging aquaculture area was the main trend in Dongzhai Harbor area, however, with the increasing attention to the mangrove protection from the public and the government in the following years, ponds area had been limited; accordingly, the investors began to improve the productivity of per existing ponds by using more feeding additives. The survey showed that both Cu and Zn had been used as the feeding additives in local aquaculture since 2000, especially ZnSO₄·H₂O had been commonly used in shrimp breeding. That should be the reason of a sharp rising of Zn in sedimentation of mangrove wetlands from the year 2002 to 2010.

As to LPI, which quantifies the largest patch percentage in the area of total landscape, showed a negative correlation with Pb content while no significant correlations with the Cu and Zn contents. The decrease of LPI could be ascribed to the fact that the largest patches in the study area were broken by the tourism facility constructions and aquaculture ponds. Vehicles used in tourism and aquaculture discharged Pb into air and

Table 6 Pearson coefficient of correlation between the mangrove class-level indices and heavy metal contents at Dongzhai Harbor mangrove wetland of Hainan, China

	AREA (km ²)	NP(n)	LSI	DLFD	MNN	CONT
Cu(ug/g)	-0.6669 ^b	0.5981 ^b	0.9149	-0.9874 ^a	-0.8640 ^b	-0.8675 ^b
Pb(ug/g)	-0.8935 ^b	0.8501 ^a	0.9992 ^a	-0.8608 ^a	-0.9884 ^b	-0.9894 ^b
Zn(ug/g)	-0.5616 ^b	0.4860	0.8529	-0.9997	-0.7892	-0.7935

^a and ^b indicate correlation is significant at 0.05 and 0.01 probability levels, respectively

water, and finally deposited in the soil, therefore, Pb showed a better correlation with LPI.

SHDI (Shannon diversity index), which was used to measure the complex degree of landscape, showed a close correlation with Pb ($r=0.9234$, $P<0.01$) and Cu ($r=0.7186$, $P<0.05$) contents. According to the landscape index analysis, the decrease of mangrove area, which had negative relationship with SHDI, was the immediate cause of the increase of Pb and Cu. Many research showed that mangrove plants were heavy metal hyper-accumulator, while the intensity of absorbing different heavy metals were various (Tam and Wong 1995). According to Li's study (Li et al. 2012) carried on *Bruguiera gymnorhiza*, *Bruguiera sexangula*, *Kandelia obovata*, *Ceriops tagal* and *Aegiceras corniculatum*, the order of enrichment coefficient of three heavy metals was Pb>Cu>Zn. This probably could help to explain the results of correlation analysis.

On the mangrove class-level, the AREA(mangrove area) showed the negative correlations with the heavy metal contents in the soil. That is to say, with the decrease of mangrove area, the content of heavy metals in soil increased. It once again proved that mangroves are powerful metal hyper-accumulators.

MNN (Mean nearest neighbor distance of mangrove patch) and CONT (reflecting the coherent and dispersal between mangrove patches) presented negative correlation with Cu, Zn and Pb, meaning that scattered distribution of mangrove forest reduced its ability in accumulation heavy metals. Based on abundant case studies, Frazier (1998) had already proved the importance of keeping ecosystem integrity.

DLFD showed a negative correlation with the contents of Cu ($r=0.9874$; $P<0.05$) and Pb ($r=0.8608$; $P<0.05$), indicating that the more complex of the mangrove boundary was, the less Cu and Pb preserved in the soil sediments. The boundary shapes and the spatial pattern of an ecosystem could affect its ecological functions, such as protecting the coast from hurricanes (Rodgers and Gamble 2008) as well as forest fire (Chang et al. 2004). However, to the best of our knowledge, no detailed study about the correlations between the wetland pattern and its enrichment function has been reported. Mangrove plants can reduce both Cu and Pb contents in the soil to a certain extent during their metabolism processes. However, further study will be necessary for a fully understanding of the spatial pattern effects on heavy metal sedimentation.

Finally, LSI (characterizing the complexity of patch shape) showed a close correlation with Pb content, suggesting that the mangrove patch boundaries approaching to the square shape could lead to a significant increase in the Pb content in sedimentation. Forman and Godron (1986) suggested that most of linear boundary and square shape were connected with human activities, so it indicated again that human activities lead to the increase of heavy metal in soil. We can explain these in two ways, first, the nature mangrove was a narrow

band along the coastline, which implied the existence of a great variety of habitats. The diversified habitats tended to hold more species, especially microorganisms, which played an important role in transforming and absorbing heavy metals (Brix et al. 2003); second, the accumulation of heavy metals could be lessened by tide water from overlay. The narrow mangrove band ensured the metals in soil sediments more accessible by tide water. This might also explain that the increasing of mangrove caused by artificial *Sonneratia apetala* planting did not followed by a decreasing of heavy metal content since 1999.

Summary

The present study showed that the increase in the area of aquaculture ponds and residential regions resulted in the main changing in land use patterns, which, to a certain degree, influenced the distribution characteristics of heavy metal sedimentation in mangrove wetlands. The correlation analysis results showed that the landscape fragmentation caused by aquaculture activities, had a close relation to the increasing of Cu, Zn and Pb contents in the soil sediments. The indices that depicting the mangrove spatial pattern, including MNN, SHDI and DLFDD, also demonstrated a close relation to the heavy metal distribution in the mangrove.

Land use, not only land using mode, but also land spacial pattern, has close relation to heavy metal moving, transform and sedimentation. To avoid soil pollution, reducing pollution sources, i.e. aquaculture and residential, is the first thing we need to do, however, it is equally important to make land use spacial pattern reasonable, to keep landscape integrity and to avoid fragmentation.

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