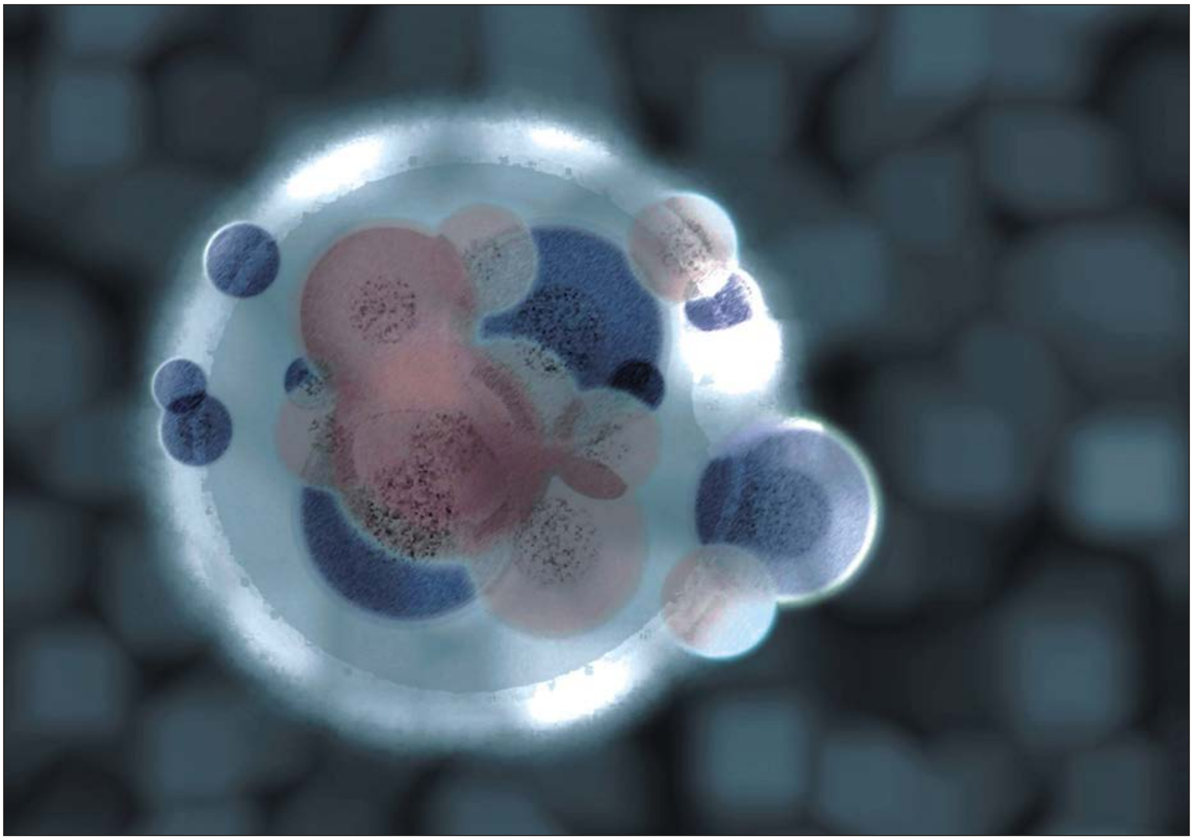

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Mini Review

Spatial and Temporal Changes of Heavy Metals in Coastal Mangrove Sediment in China: Review of Present Status

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Abstract

Mangroves are declined worldwide due to blowing up of population density as well as industrial development over the last decade. Heavy metal is a serious threat to the mangrove ecosystem in China owing to the industrial revolution. This review addresses the spatial and temporal changes of heavy metals in mangrove sediment in different provinces, sources of metals and also identified the phytoremediate mangrove species in China. The major sources of heavy metals in mangrove ecosystems depended on the local economy of the province including industrial effluent, agriculture and tourism. From the spatial variability of metals concentrations in mangrove, it was found that sediment has higher concentration at Guangdong and Fujian than Guangxi and Hainan province. This is because of Hainan and Guangxi are less developed than Guangdong and Fujian. Furthermore, from the temporal variation of metals within the same province demonstrates the enhanced concentrations at Guangdong, Hainan than Guangxi and Fujian. This review demonstrates that Guangxi and Fujian have less contaminated over time and have been improving their mangrove ecosystems, but in Fujian mangrove sediment, Cr and As concentration increased over the time period than all other studied metals. Phytoremediation capacity has supported that mangrove can tolerance and translocation of metals to alleviate pollution and Phytostabilisation of ecosystems, but their accumulation and translocation ability is mostly species specific as well as depend on sediment concentration of heavy metals.

Key words: Mangrove ecosystems, phytoremediation, heavy metals, spatial variation, industrial revolution, phytostabilisation of ecosystems

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Data Availability: All relevant data are within the paper and its supporting information files.

INTRODUCTION

The distribution of mangrove is in the tropical and subtropical intertidal regions of 118 countries of which about 75% of the world mangroves are found only in 15 countries[1]. Mangroves are ecologically important and one of the most yielding ecosystems in the world[2]. In the coastal environment, mangroves contributed a wide range of ecological, socio-ecological and economical services to the local populations, particularly coastal protection, nutrient retention, the refuge for fisheries resources and habitat for coastal communities[3,4]. The economic evaluation of ecosystem services from mangrove is estimated that coastal protection is the highest monetary value among all the services and followed by nutrient retention especially heavy metals and carbon sequestration[4,5]. Instead of enormous ecosystem services, 38% of the global mangrove has been lost over the last two decades and south Asia is the most vulnerable to mangrove loss. This is due to the urbanization, conversion for aquaculture and agriculture and industrial development[6]. Because of industrial development, a substantial amount of industrial sewage is discharged into the mangrove ecosystems, as they are considered as a dumping ground over the last decades in many developing countries[7]. In the coastal environment, heavy metals get significant attention during the present time due to their accumulation, persistence, toxicity and bioaccumulation to organisms[8-14]. Industrial discharges are the main anthropogenic source of heavy metals in the coastal mangrove ecosystem, but in the natural environment, rocks and soils are the natural constituents of heavy metals[8,9]. They enter the environment through the process of weathering and erosion[10]. Moreover, the input of heavy metals in the mangrove ecosystem is from different sources like industrial effluent, agricultural wastes, untreated domestic sewerage, oil spills and settlements in the coastal areas[9]. However, unnatural Arsenic (As), Cadmium (Cd), Copper (Cu), Mercury (Hg), Lead (Pb), Nickel (Ni) and Zinc (Zn) have been released from industries into the aquatic environment through storm water and wastewater discharges as well as from agricultural activities[11]. Zn and Cu are mostly used as fertilizers and As, Cd and Hg are used as fungicides in the agricultural fields[12,13]. Therefore, in the developing countries, high metals load from the industrial and domestic sources to the coastal environment, where has not enough treatment plants before reaching to the aquatic environment[5]. In addition, the rivers carry both domestics and industrial wastes from the upstream to the downstream and releases into the sea, but in the case of coastal areas bordered by mangrove vegetation, the mangrove soil acts as a biological filter and reduces the impact on pollution before reaching to the open waters[13-15]. Mangrove sediments are anaerobic and rich in organic matter, Sulphide, dense rooting system and high clay content in their sediment which is favourable for the retention of trace metals[16-18]. Moreover, the alternation of anoxic and anoxic condition of mangrove sediment, metals may have dissolved forms during the bearing phase and dispersed to the surrounding tidal areas[5].

However, in China due to the population boom and industrial development, a considerable amount of pollutants is discharged into the coastal mangrove ecosystems and it has reached[7] approximately 200 million ton. Among the discharges, heavy metals are of serious concern due to a large amount of cadmium, zinc and lead; potentially have an impact on human health and organism in the coastal biota[7,19]. Furthermore, heavy metals are the serious pollutants due to their toxicity, persistence and bioaccumulation[9,20]. Several studies indicated that the heavy metals concentration of China's mangrove is higher than sediment quality and has a considerable impact on mangrove ecosystem as well as coastal biota[7,9,21,22]. Therefore, in the mainland China 22700 ha mangrove have been declined[23] from 1980-2001.

Regular monitoring and assessment are important to evaluate the potential ecological risk and pollution status of metal pollution to the mangrove ecosystem. In addition, for better monitoring, assessment and changes of management strategy in the coastal areas, historical and spatial scale pollution status is a crucial criterion to evaluate and identify the changes of anthropogenic sources and contaminants. There are limited research reviews of the comparative monitoring of mangrove heavy metals pollution status in China except for some review of research on mangrove distribution and conservation strategies[23,24]. In this present study, heavy metals in the mangrove in mainland China were reviewed to investigate the current research on metals pollution, thereby to evaluate the change of metals pollution status with the temporal and spatial scale of provinces. Moreover, this paper was also investigated the phytoremediation capacity of mangrove species in China and possible recommendation of future prospective research.

HEAVY METALS

There is no universal definition for heavy metals. They are ions with a low number in the molecular table (>20) and their densities are greater[12,25,26] than 5 cm^{-3}. Most of the metals are in the D-block in the periodic table and form cations in the physiological condition. Furthermore, heavy metals can be classified into three categories:

- Non-critical
- Toxic but very insoluble
- Very toxic but accessible[13]

In the physiological condition of plants, few metals are soluble which are referred to as trace metals or micronutrients such as Fe, Zn, Ni and Cu, while the others such as As, Cd, Cr and Pb are toxic even present in trace concentration[13,27]. In addition, heavy metals present in soils is in different forms such as; free metal ions, as soluble metal complexes, associated with soil organic matter, as oxides, hydroxides, carbonates and incorporated into silicate material structures[25,26].

DISTRIBUTION OF MANGROVE AND SOURCES OF METALS IN MANGROVE SEDIMENT

Naturally, mangroves are distributed among Hainan, Guangdong, Guangxi, Fujian, Hong Kong, Macau and Taiwan in China[23]. In addition, Zhejiang province also has mangrove which was transplanted[23] in 1950s. In China, total areas of mangrove are in about 22752 ha of which 94% of the mangroves are in the three provinces of south China[23] (Table 1). The largest existing mangrove in China is in the Guangdong province (Table 1) followed by Guangxi, Hainan, respectively. The 24 true mangrove species are present in China, which represent one-third of the world true mangrove species[23].

In the Guangdong province the dominant species are in *Aegiceras corniculatum*, *Bruguiera gymnorrhiza* and *Sonneratia apetata*[28]. Marine culture and agriculture are the major financial sources to the local people surrounding Guangdong province mangrove[28]. In the Hainan province mangrove, *Rhizophora stylosa* and *Sonneratia apetata* are the dominant species, agriculture and tourism is the major pillar of the economy in this province[28]. *Kandelia obovata* is the dominant mangrove vegetation in the Fujian province mangrove, where local economical sources are mainly textile, steels and food industries[28]. *Bruguiera gymnorrhiza*, *Kandelia candel* and *Rhizophora stylosa* are the dominant species in the Guangxi province mangrove, where main economical sources are agricultural activities and tourism[24,29]. In the Zhejiang province, there is no natural mangrove, only one species *Kandelia candel* was planted in this mangrove forest[24]. Due to industrialization, economic growth and population boom, mangroves are in intense pressure due to human footprint. The land-use change in coastal areas is one of the main biotic pressures on the mangrove ecosystems which in turn also facilitate pollution[11]. In China, the metals

Table 1: Total areas of existing mangrove in China

Location	Total existing mangrove area (ha)	Total area (%)
Hainan province	3930	17.20
Guangdong province	9084	40.00
Guangxi province	8375	36.80
Fujian province	615	0.02
Zhejiang province	21	0.0001
Hongkong	380	0.016
Macau	60	0.002
Taiwan	287	0.01
Total area	22752	100

Source: Chen *et al.*[23]

Table 2: Sources of heavy metals in mangrove in mainland China

Location	Major sources of heavy metals	References
Hainan province	Natural sources such as geo-genic	Liu *et al.*[28]
Guangdong province	Industrial effluent, domestic sewage	Liu *et al.*[28]
Guangxi province	Agriculture and tourism	Zhang *et al.*[29]
Fujian province	Textile, steels and food industries	Liu *et al.*[28]
Zhejiang province	Agriculture and tourism	Zhang *et al.*[29]

input in mangrove ecosystems is dependent on the local economic sources, development and population density. In the Guangdong and Fujian provinces, the major heavy metals input in the mangrove ecosystems is due to industrial discharge and sewage effluent (Table 2).

The recent study on the Shenzhen mangrove found that high concentration of cobalt and tungsten due to the discharge of industrial wastewater and uncontrolled chemical alloys from chemical factories[30]. In addition, recent investigation on Futian mangrove of Guangdong province was observed the moderately contaminated level of Hg in sediments indicated the coastal based source of pollution[31]. These recent finding suggested that Fujian and Guangdong province mangroves are contaminated by metals due to land and coastal based anthropogenic sources[30,31]. In contrast, in the Zhejiang and Guangxi province, agricultural and tourism are the main source of heavy metals in mangrove ecosystems. In addition, the geo-genic origin is the main source of heavy metals in the Hainan province mangrove (Table 2).

HEAVY METALS ACCUMULATION AND POLLUTION STATUS OF MANGROVE

Mangrove sediment contains a mix matrix of inorganic and organic carbon, heterogeneous compounds and play a pivotal role in the coastal biogeochemistry cycles for holding more than 90% metals in the aquatic environment[8,32]. Heavy metals accumulation in mangrove sediment has been reported to several countries including Australia, Singapore, India, Brazil, Saudi Arabia, Hong Kong and Malaysia[9,33-37]. The sink of heavy metals in mangrove sediment is resulting in the baffle activities of roots and trunks[32,38]. The accumulation of fine sediment and organic-rich content is also influenced by

Table 3: Heavy metals concentrations (mean) (mg kg^{-1}) in mangrove sediment at different mangrove reserve in China

Province	Mangrove reserve	Cu	Pb	Zn	Cd	Cr	Hg	As	References
Hainan	Dongzhaigang	18.00	19.00	57.00	0.11	40.00	0.08	13.00	Qiu *et al.*[43]
	Sanya Bay	9.00	18.00	53.00	0.13	12.00	0.06	7.00	Qiu *et al.*[43]
	Wenchang	27.00	30.00	89.00	ND	109.00	0.06	15.00	Vane *et al.*[44]
	Dongzhaigang	22.93	28.87	49.63	0.28	61.13	0.20	19.23	Liu *et al.*[28]
Guangdong	Zhanjiang	16.90	32.80	49.00	0.20	5.12	0.00	0.00	Zhang *et al.*[29]
	Zhanjiang	18.30	41.90	42.10	0.36	44.60	0.16	14.00	Liu *et al.*[28]
	Futian	31.70	47.80	296.30	2.30	55.40	ND	ND	Li *et al.*[21]
Guangxi	Fangchenggang	5.70	23.60	49.00	0.12	12.20	0.12	4.70	Zhang *et al.*[29]
	Qinzhou	17.10	46.70	65.00	0.07	13.40	0.15	9.70	Zhang *et al.*[29]
Fujian	Quanzhou	34.00	167.00	107.00	0.06	18.60	0.04	5.30	Zhang *et al.*[29]
	Quanzhou	14.30	59.50	50.90	0.11	59.80	0.061	13.80	Liu *et al.*[28]
	*BG	10.1±5.18	26.5±12.4	51.9±29.6	0.004±0.03	47.2±26.9	0.035±0.04	11.8±3.87	Vane *et al.*[44]
	†MSQGs	35.00	60.00	150.00	0.50	80.00	0.20	20.00	Liu *et al.*[28]

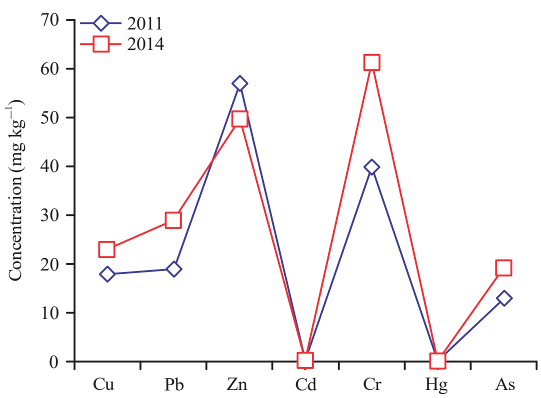

Fig. 1: Temporal change (between 2011 and 2014) of heavy metals concentrations in Dongzhaigang (DZG) MNR in Hainan province, China

Source: Liu *et al.*[28] and Qiu *et al.*[43]

the baffle activity of mangrove roots and trunks[32,38]. The high specific surface area of fine sediment and organic matter were facilitated to trap heavy metals from water and reduce transport to deeper layers of sediment[32,38]. As fine sediment, contained more metals than course sediment, it is an important factor for the distribution of metals in the mangrove areas[32]. Due to the high absorbing and holding capacity of heavy metals, they were contributed to binding with sediment. However, the disturbance such as prolonged dry periods, pH and salinity accelerates mobility of the metals and loosens the sediment-metal binding[36]. Therefore, mangrove sediment behaves like a heavy metals source because of human disturbances[39]. In addition, heavy metals accumulation and concentrations changed with the regional and local industrial development[40]. As the mangroves in China are in the different region and the development is not the same in all the regions, resulting in high variability in heavy metals concentrations according to the regional development. The heavy metal concentration of Cu, Zn, Cd, Cr, Pb, Hg and As ranged from 0.5-93, 9-410 mg kg^{-1}, nd (not detected) to 5.4 mg kg^{-1}, nd to 363, 7-180 mg kg^{-1}, nd to 0.47 and 1.2-19 mg kg^{-1}, respectively in the mangrove sediment in China[40], but pollution levels of metals in mangroves are confined to a narrow range compared with world mangrove values[41]. As a reference, the background values of China's coastal sediment were used due to the unavailability of mangrove background values[42]. In comparison with the reference value, great portion of heavy metal values are higher than the reference values indicated mangrove ecosystems have been contaminated with heavy metals (Table 3).

Furthermore, there is a spatial difference between heavy metals concentrations of China's mangrove sediment (Table 3). From Table 3, it is explained that Guangdong and Fujian have the high concentration of Cu, Zu Cd and Pb than Guangxi and Hainan.

This spatial variation of concentration is largely depending on the economic importance and developments, as Guangdong and Fujian are considerably developed and have higher population density than Guangxi and Hainan[43].

In the Dongzhaigang (DZG) mangrove reserve of Hainan province concentrations of Cu, Pb, Cr and As have considerably increased in 2014 than 2011 concentrations, but Zn concentration decreased within this period (Fig. 1). This is due to the lift of anthropogenic activities such as; land reclamation for the tourism industry. This province's main economic source is tourism and agriculture, but geo-genic also has influence the present pollution status[28,43,44].

Furthermore, in the Guangdong province, concentrations of Cu, Pb, Cd, Cr, Hg and As have enhanced considerably in the Zhanjiang mangrove reserve in 2015 than concentrations in 2008, but at the same time Zn concentrations decreased (Fig. 2). Increasing concentrations of heavy metals in mangrove areas is mainly due to industrial effluents discharges, but also marine culture is the main source of enhancing as in this mangrove[28,29,40].

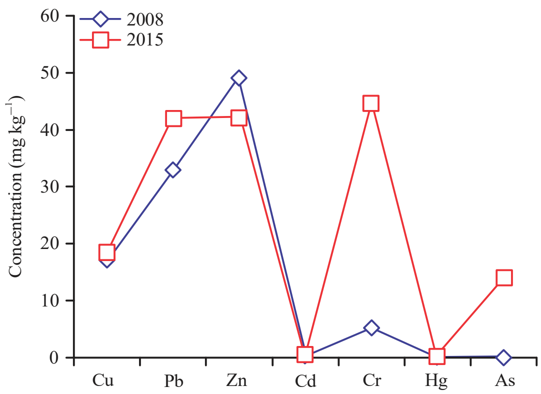

Fig. 2: Temporal change of (between 2008 and 2015) heavy metals concentrations in the Zhanjiang mangrove in Guangdong province, China
Source: Zhang *et al.*[29] and Wang *et al.*[40]

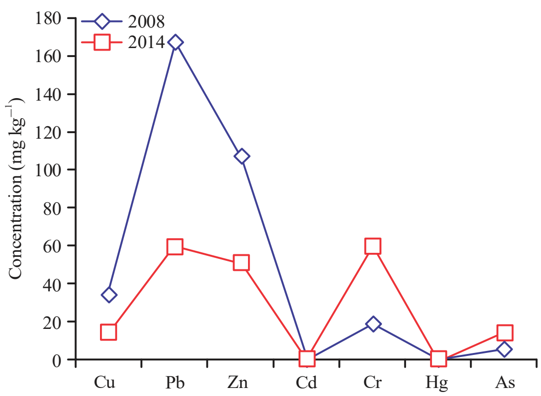

Fig. 3: Temporal change of (between 2008 and 2014) heavy metals concentrations in the Quanzhou mangrove forest sediment at Fujian province, China
Source: Zhang *et al.*[29] and Liu *et al.*[28]

Land-based sources and increasing use of pesticides also responsible for increasing metals concentrations[45]. The concentrations of Cr and As have increased in the Quanzhou mangrove forest sediment in the Fujian province in 2014 than sediment concentrations in 2008 (Fig. 3). Pb, Cu, Zn and Hg concentrations were reduced at the same time[28,29] (Fig. 3).

Another study demonstrated that metals concentration of Futian mangrove has lower than Quanzhou indicated that pollution level is decreased in Guangdong mangroves[19]. In addition, Hg concentration of Futian mangrove from the very recent study suggested that the concentration of Hg is increased in the Guangdong than Quanzhou mangrove sediments[31]. Overall the concentrations of heavy metals are considerably lower in Fujian province mangrove than all other provinces mangrove (Table 3), but increase in metals

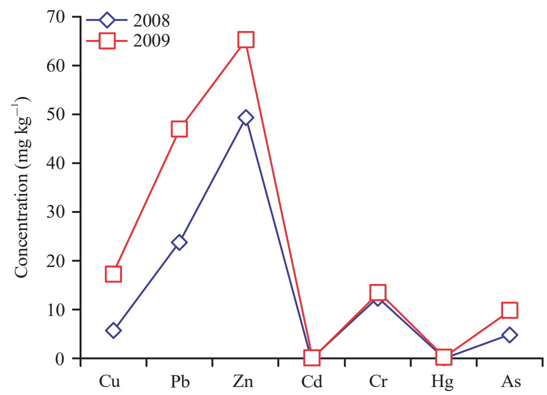

Fig. 4: Temporal variation of (between 2008 and 2009, blue line: Fangchenggang mangrove and Red line: Qinzhou mangrove) heavy metals concentrations of Guangxi province mangrove, China
Source: Zhang *et al.*[29]

concentrations of mangrove sediment is due to the major industrial development in Fujian province such as; steel, textile and food processing industries[28].

In the mangroves of Guangxi province, concentrations of Cu, Pb, Zn, Cr and As concentrations have increased in Qinzhou mangrove in 2009 than Fangchenggang mangrove in 2008 (Fig. 4). The overall concentration of heavy metals in this province is comparatively lower than Hainan and Guangdong (Table 3). This is because of newly developed provinces and relatively less developed compared to other provinces resulting in lower anthropogenic activities[43]. In the Zhejiang province, no data were found to identify the current temporal status of heavy metals pollution in this province's mangrove. So intensive research is recommended for evaluating the heavy metals concentrations in this province's mangrove sediment.

PHYTOREMEDIATION AND TOLERANCE CAPACITIES OF MANGROVE SPECIES

Phytoremediation is a way of technique to remove metals from the environment through plants without disturbing the ecosystems and comparatively less expensive method than the conventional physicochemical method of metals removing[26,46]. In the mangrove ecosystems, the most studied process of phytoremediation is phytoextraction, through metals absorbed in roots, transport and accumulates in shoot and leaves[47]. In addition, phytostabilisation is also an important process by which plants immobilize metals in their roots systems and reduce mobility[26,47]. Furthermore, rhizofiltration is a process by which metals are removed from surface water through adsorption and precipitation, but very few studies are conducted on this process in mangrove ecosystems compared to the other two processes of phytoremediation[47]. Mangrove ecosystem acts as a sink of heavy metals and mangrove has different excluding and regulators capacities of metals, being mangroves physiologically tolerant of the adverse polluted environment[16].

As a result, several researchers studied mangrove phytoremediation capability in the field and lab conditions. Usman *et al.*[12] mentioned that *A. marina* can reduce pollution through the process of phyto extraction while MacFarlane *et al.*[48] stated it as an indicator species for heavy metals accumulation. The accumulation and translocation of heavy metals in roots and root to different body parts (e.g. leaves, stems) of mangroves from sediment indicated bio-concentration and translocation factor, which also demonstrated the phytoremediation capacities of mangroves[9,20]. The accumulation of heavy metals in mangrove roots is higher than the surrounding sediment and it depends on the restriction capacity of plants roots and surrounding sediment concentration of metals[20,43]. Translocation of metals from mangrove roots to different body parts depends on the concentration of metals in the surrounding sediment, root anatomy, type of metals and pH[9,48].

Essential metals (e.g., Cu, Zn) have greater translocation factor than to non-essential metals as they are important for the plant physiological activities i.e., photosynthesis[49,50]. Lower pH in water increases the solubility of immobile metals and lift their concentration resulting in greater accumulation in roots and translocation to above ground body parts[9,20]. However, in mainland China, about 15 mangrove species tissues have been studied to evaluate the metals accumulation and translocation in their body parts[29]. Accumulation and translocation of metals in mangroves tissues depend on the sediment concentration and species tolerance and physiological capacities. Several studies showed different concentrations of different species and tissues[29]. In the mangrove tissues, Cu, Zn, Pb, Cd and Cr are most studied metals, As and Hg studies in tissues are rarely found[29]. In the Futian mangrove forest of Guangdong province, different accumulation and translocation of metals in the different body parts of eight mangrove species have been shown. Cu and Zn have significantly higher concentrations than non-essential metals except Cr. Higher accumulation of Cr in both stems and leaves of eight species but *B. gymnorhiza* exhibited the highest concentration in their stems among the species. In addition, Hg showed also greater concentration in leaves than stems and greater accumulation in the *E. agallocha* leaves[51].

Additionally, bio-concentration factor (BCF) of Cr value showed >1 and significantly enhanced stems and leaves than other metals which support the translocation and accumulation of Cr in these mangrove species of Futian mangrove forest[51]. Researchers showed the accumulation and translocation of metals concentrations of seven species of mangrove tissues (leaves, root, stems and fruit) were different along their species tissues in the Hainan province's mangrove forest. The metals concentrations of Cu, Pb, Zn, Cd, Hg and As in mangrove tissues were 2.8, 1.4. 8.7, 0.003, 1.1, 0.003 and 0.2 $\mu g\ g^{-1}$, respectively[43]. It is also indicated that Zn and Cu have greater accumulation in fruit whereas Pd, Cd and Cr were in the branch and As accumulated in the root. The BCF showed in the sequence of Hg (0.43)>Cu (0.27)>Cd (0.22)>Zn (0.17)>Pb (0.07)>Cr (0.06)>As (0.02)[44]. Since Hainan province mangrove forests are less impacted by human activities resulting in less accumulation and translocation of metals but Hg showed greater accumulation which indicates phytoextraction capacity of these plants in this forest. In the recent study on Hainan island, two species of mangroves (*Rhizophora stylosa* and *Sonneratia hainanensis*) suggested that accumulation of metals depends on the species and their mechanisms and additionally TOC of sediments which played an important role for metals regulating in the different part of mangrove tissues[52].

FUTURE PROSPECTIVE

From this review, it is clearly demonstrated that from the time scale of industrial development in China, heavy metals pollution stress on the mangrove ecosystem being enhanced but changes of concentrations are different in different provincial mangrove. In future, more focus should be given on the research on metals accumulation and translocation on mangrove plants and their life stages to evaluate phytoremediation capacity of mangrove species, improve modeling of metals transfer to different mangrove body parts as well as ecological risk to biota and local dependent people on mangrove. Furthermore, early life threshold metals tolerance level should be further studied through metals toxicity test. Finally, understanding the molecular mechanisms of tolerance of heavy metals, more research on stress gene to identify the complex tolerance structure of metals tolerance[53].

CONCLUSION

Sediments of the mangrove ecosystems in China were contaminated with heavy metals as most of the studied metals concentrations are higher than background values, indicated anthropogenic contaminants are major sources of metals in this ecosystem. Therefore, there was considerable spatial variability of metal concentration in sediments of different mangrove areas, suggested the development of local economy and industrial proliferation. Moreover, temporal changes of metals concentration in sediments of mangrove in different provinces indicated changes of anthropogenic sources over the time period in the respective province.

SIGNIFICANCE STATEMENT

This study discovered the spatial and temporal variability of heavy metals concentrations in mangrove sediments in China that can be crucial for the future management strategy and implementing a monitoring program. The study will help the researchers to uncover and identify the changes of heavy metals anthropogenic sources over the time periods. Thus, more research, monitoring and remediation will be the focus on increased heavy metals.

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