# DISTRIBUTION AND GEOCHEMISTRY OF LEAD IN TIDAL CREEK SEDIMENT IMPACTED BY ANTHROPOGENIC ACTIVITIES

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**ABSTRACT**

Mangrove-dominated estuaries could play an important role in mitigating pollutants from the land to the sea. However, changes in the physicochemical characteristics of water and sediment could affect the spatiotemporal distribution of pollutants, including heavy metals which may induce ecological risks to organisms during tidal cycles and seasons. In the present study, we aimed to evaluate the lead (Pb) partitioning and its ecological risks in tidal creek sediments within the Can Gio mangrove estuary by using a method of sequential extraction. The results shown that the total lead content ranged from 16.92 – 24.55 mg/kg. We suggest that Pb originated mainly from natural sources during the weathering process that could be supported by Pb partitioning in residual fraction – silicate bonding i.e., F4 > F3 > F2 > F1 and geoaccumulation index (Igeo) < 0. The risk assessment code (RAC) was less than 10% at the sampling sites which indicated low ecological risks and anthropogenic activities were minor factors that may influence Pb geochemistry.

# PHÂN BỐ VÀ ĐỊA HÓA CỦA CHÌ TRONG TRẦM TÍCH KÊNH RẠCH BỊ ẢNH HƯỞNG BỞI HOẠT ĐỘNG DÂN SINH

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**TỪ KHÓA**

Chì
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Kim loại
Trầm tích
Cần Giờ

**TÓM TẮT**

Các cửa sông có rừng ngập mặn đóng vai trò quan trọng giảm thiểu chất ô nhiễm từ đất liền ra biển. Tuy nhiên, những thay đổi về tính chất hóa lý của nước và trầm tích có thể ảnh hưởng đến sự phân bố của chất ô nhiễm theo thời gian và không gian, bao gồm cả kim loại nặng, chúng có thể gây ra rủi ro sinh thái cho sinh vật theo thủy triều và mùa. Trong nghiên cứu này, chúng tôi sử dụng phương pháp chiết tuần tự các phân đoạn hóa học để đánh giá sự phân bố của chì (Pb) và rủi ro sinh thái trong trầm tích ở các con rạch bị ảnh hưởng bởi chế độ thủy triều vùng cửa sông có rừng ngập mặn Cần Giờ. Kết quả cho thấy hàm lượng chì tổng số dao động trong khoảng 16.92 – 24.55 mg/kg, có nguồn gốc chủ yếu từ các nguồn tự nhiên được phóng thích và tích lũy trong quá trình phong hóa. Kết luận này có thể được chứng minh qua hàm lượng Pb liên kết chủ yếu với khoáng silicate – F4, cụ thể F4 > F3 > F2 > F1 và chỉ số tích lũy địa lý (Igeo) < 0. Mã đánh giá rủi ro (RAC) < 10% tại các điểm lấy mẫu cho thấy rủi ro sinh thái của Pb ở mức thấp và các hoạt động nhân sinh không phải là nhân tố chính có thể ảnh hưởng đến địa hóa học của Pb.

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## 1. Introduction

Estuary is considered an effective buffer zone between land and sea, receiving large amounts of inorganic and organic matter, and protecting coastal water from pollution. Among pollutants, heavy metals are one of the main sources that could cause negative impacts on the aquatic and sediment ecologies [1]. The variability of trace metals is complex due to the influence of different biogeochemical processes throughout the topography and tidal regime. Because of their potential toxicity to biodiversity and ecological health, trace metal cycling has become an issue that scientists have been interested in for recent decades [2].

Mangrove-dominated estuaries provide a distinctive mechanism of trapping sediment that could carry pollutants and accelerate land-building processes in tide-dominated coastal and estuarine environments. Industrial wastewater, agricultural and fisheries production industries, wastewater treatment plants, and leaching from domestic and urban landfills have been reported as (non)point sources of heavy metals in estuaries that could decrease water and sediment quality, reproduction, biodiversity, coastal ecosystem function, and human health through the food chain [3]. During their transport, heavy metals are often distributed in water-soluble, colloidal, suspended, and sedimentary forms [4]. After deposition in sediments, trace metals can exist in different mineral bonds: amorphous crystals; adsorption on clay or iron/manganese oxyhydroxide surfaces; crystal lattice of carbonate, sulfate, or other elemental oxides; organic matter (OM) or silicate minerals [5] - [7]. Heavy metal association and distribution in sediment depend on pH [8], salinity [9], redox potential [10], and organic content [11], [12]. In contrast to organic pollutants, heavy metals are not degraded biologically and chemically and could be thus transported, deposited, and accumulated in sediments. The cycling, toxicity, bioaccumulation, persistence, and bioavailability of heavy metals e.g., Pb, Cd, Ni, Cr, and Co, etc. in mangroves have been studied and reported [13], [14]. However, mangrove sediments are rich in sulphide and organic matter which modify metal geochemistry after deposition [15], [16]. Therefore, determining the content and partitioning of heavy metals in the sediment is necessary [17].

In Viet Nam, a developing country, the capacity of water treatment factories is insufficient to treat the pollutant load e.g., metal(loid)s which are thus discharged to tidal creeks, rivers and estuaries, from anthropogenic activities. As most of the tropical estuaries are dominated by mangroves, metal(loid)s can be deposited in these ecosystems. Can Gio is the biggest mangrove reforestation area in Viet Nam. Many scientists have performed studies on various aspects of the Can Gio mangrove ecosystem. For example, factors affecting mangrove development and distribution, mangrove vegetation components and characteristics, and mangrove degradation [18], [19], the ecological values [20], the impact of hydrology on mangrove structure and function [21], accretion rates of sediment [22], [23] and also the influences of sedimentary-physical chemical parameters on the organic nutrient cycles. However, the research on metal geochemistry was still lacking. Costa-Boddeker et al. [24] reported the current status of metal accumulation in sediments at the edge of the Can Gio mangrove, showing that the distribution and accumulation of heavy metals were complex, especially in estuarine regions, where there was a reciprocal interaction between many factors that disturbed physicochemical properties. The problems of differences in physicochemical conditions between seasons of the year also help to better understand the state, evolution and bioavailability of heavy metals. Thanh-Nho et al. published research on metal content along the mangrove estuary in Can Gio mangrove estuary [25]. However, previous publications focused only on the total content of metals with out consideration of their various bond forms in the environment. So, the aims of the present study were to assess metals i.e, Pb distribution and its geochemistry in tidal creek sediments impacted by anthropogenic activities. Samples were collected at sites that receive effluents from the aquacultural and urban activities.

## 2. Methodology

### *2.1. Study area*

Can Gio is located in the Southeast of Ho Chi Minh City (10º22'14" - 10º40'09"N; 106º46'12 " - 107º00'59"E) which is a unique district bordering the Eastsea, surrounded by an intricate river system, such as Long Tau, Soai Rap, Cai Mep, Thi Vai, and Dong Tranh (Figure 1). The area of Can Gio district is 70,455.34 hectares and can be divided into 3 zones: (1) the core zone has an area of 4,720 hectares completely covered by mangrove forest which was established with the long-term purpose of landscape conservation, species biodiversity, and research. Human activities are prohibited in this zone; (2) the buffer zone with an area of 37,340 hectares is also covered by mangrove forests, providing a natural landscape, and serving as a cultural and ecological tourist destination; (3) the transition zone covers an area of about 21,331 hectares, which is the outermost area with low to dense coverage, and is important for maintaining socio-economic activities. For socio-economic development, a part of the Can Gio area was exploited and converted to shrimp farming and residential areas which were considered the direct cause of disturbing the properties of sediments. These activities can cause disturbance and increase the content of organic substances as well as the ability to accumulate metal(loid) in sediments. The process of forest exploitation and conversion to aquaculture leads to increasingly narrowing forest areas and disturbed sediment properties, increasing the mobility of metals and ecological risks. Due to their toxicity, bioaccumulation and persistence, trace metals can pose a major threat to mangrove biodiversity and also to human health. Therefore, it is necessary to better understand the behavior of metals in the Can Gio mangrove ecosystem.

### *2.2. Sample collections*

The location of sampling sites are exhibited in Figure 1 and the characteristics of each sample are shown in Table 1. Samples were collected according to TCVN 6663-3:2008 - Water quality - Sampling - Part 13: Guidance on the sampling of water, wastewater and related sludges. Sediment cores were collected using a specialized stainless steel tube, each core was divided into 5 sub-samples with depths (i.e., 0-10, 10-20, 20-30, 30-40, and 40-50 cm) and stored in polyethylene bags. The samples were kept in a cool box and transported to the laboratory. The sample was dried using a freeze-dryer for 48 hours. After that, the samples were ground and sieved through 100 μm before analysis. Samples were analyzed at the laboratory of the Department of Analytical Chemistry, University of Science, Vietnam National University Ho Chi Minh City.

**Table 1.** *Samples coordinate and characteristic.*

| Samples | Coordinates | Characteristic |
|---|---|---|
| $S_1$ | 10.618574, 106.841402 | The outlet area of the pond and residential area |
| $S_2$ | 10.590105, 106.852355 | Receiving effluent from shrimp farms and urban |
| $S_3$ | 10.571295, 106.814815 | The tributary, near shrimp farms and residential areas, connects the Long Tau River. |
| $S_4$ | 10.5127435, 106.75840 | Drains, near the wastewater discharge of shrimp ponds, connect small creeks and rivers. |
| $S_5$ | 10.463828, 106.776430 | Residential areas, near the outlets of shrimp ponds, and salt fields. |
| $S_6$ | 10.423134, 106.825155 | The mangrove forest is recovering, near the mouth of the river adjacent to the sea, surrounded by shrimp and salt farms. |
| $S_7$ | 10.543025, 106.830640 | Location is considered as a referent sample with limitation of anthropogenic activities collected in the buffer zone of the Can Gio mangrove forest. |


**Figure 1.** *Location of Can Gio mangrove and sampling sites*

### *2.3. Sequential extraction process*

Lead was divided into four operationally defined geochemical fractions: an exchangeable/carbonate fraction (acid-soluble phase), a Fe-Mn oxides fraction (reducible phase), an organic fraction (oxidizable phase), and a residual fraction (silicate bonding). Briefly, the various single extractions were performed as follows: 0.5g of sediments were put into 50 mL polypropylene tubes with caps, which were also used for shaking and centrifugation to minimize the possible loss of the centrifuge washing step. For the determination of the acid-soluble fraction (F1), we used 15 mL of 0.11 M $CH_3COOH$/pH = 5 at room temperature for 1.5h; for the reducible fraction (F2), 15 mL of 0.2M acid ascorbic was used during 1.5h; for the oxidizable fraction (F3), 5 mL of 30% $H_2O_2$ and 15 mL of 1 M $CH_3COONH_4$ were used at 85 $^oC$ during 2h; for the lead content in the residual fraction (F4), the samples was digested using 12 mL of concentrated $HNO_3$/HCl (3:1v/v) in polytetra fluoroethylene (PTFE) vessel at 110 $^oC$ for 24h. After that, the chemicals were eliminated at 160 $^oC$ to approximate a volume of 2 mL. The sample was filtrated and then adjusted into 25 mL using deionized water, which was stored in pre-cleaned polypropylene tubes at 4 $^oC$ until analysis [26].

## 3. Results and Discussions

### *3.1. Distribution of total lead content*

The variability of total lead contents in sediments is shown in Figure 2, ranging from 16.92 to 24.55 mg/kg, in detail: 24.55 ± 0.59 mg/kg (S1), 22.31 ± 0.62 mg/kg (S2), 22.45 ± 0.11 mg/kg (S3), 24.54 ± 0.53 mg/kg (S4), 21.94 ± 1.10 mg/kg (S5), 21.93 ± 2.71 mg/kg (S6) and 16.92 ± 0.67 mg/kg (S7). According to QCVN 43/2017 on lead content in saltwater sediments of 112 mg/kg, the lead content in Can Gio mangrove sediments of the present study was below the allowable level. The Pb content at site 7 was lower than other ones that could relate to this site being a forest area not affected by anthropogenic activities. However, it is seen that the increase of lead content in the study area tends to accumulate over time which could be supported by higher Pb content than those measured in the Can Gio mangrove (i.e. the lead content was 8 - 20.4 mg/kg) published by Costa-Boeddeker, et al. [27].

In addition, comparing to the Canadian Standards for evaluating risk level of Pb pollution to the environment, i.e., less than 32 mg/kg: unpolluted, while a higher content suggests 32 – 48 mg/kg: low risk; 48 – 64 mg/kg: medium risk; 64 – 96 mg/kg: high risk, 96 – 112 mg/kg: very high risk and > 112 mg/kg: level of influence [28]; for US EPA [29] with Pb risks, i.e., less than 40 mg/kg: unpolluted, 40 – 60 mg/kg: high risk and > 60 mg/kg: very high risk, all sites of the present study was at a weak level to no pollution. However, because of low flow and high sedimentation rates in the tidal creeks controlled by anthropogenic activities as well as lead to potential human health risks through food chains, it should be limited for land use change to aquacultural farming and urbanization in the future.

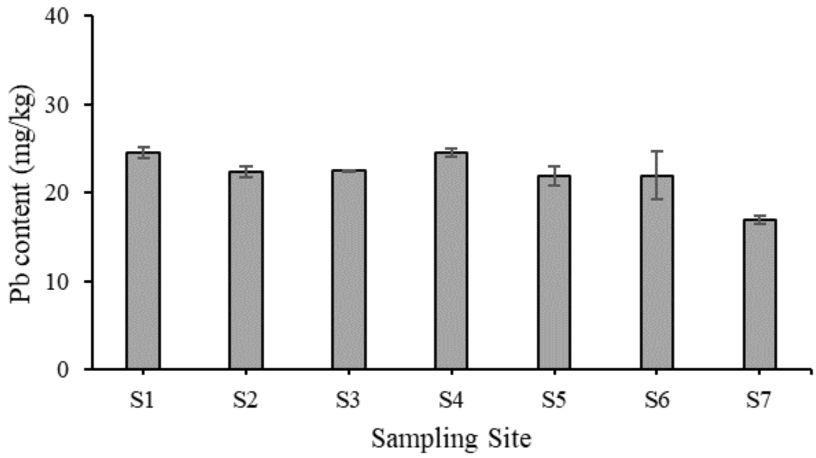


**Figure 2.** *Variability of total lead content in the creeks*

### *3.2. Partitioning of lead in sediments*

Lead contents in the different fractions are shown in Figure 3, mainly concentrated in a silicate-bound form (F4), decreased in the order F4 > F3 > F2 > F1, which was the form most tightly bound to the mineral matrix. In the residual fraction, the Pb content ranged from 8.07 – 24.23 mg/kg (i.e., 46.4 - 98.4% of the total content). Lead content in the form bound to organic matter varied from 0.019 - 8.94 mg/kg (i.e., 0.088 - 46.13%). For the fraction of Fe-Mn oxyhydroxide, Pb content fluctuated between 0.24 – 1.34 mg/kg accounting for 0.963 - 7.703%, while Pb content in exchangeable/carbonate phase changed from 0.068 - 0.991 mg/kg corresponding to 0.308 - 5.680% which is easily exchanged under the influence of factors as pH [4]. The lead distribution also changed between sites. At S1, S2 and S4, the Pb was mainly concentrated in a silicate-bound form with all depths, which may relate to the limit of influence by human activities [30]. However, for S3, S5 and S6, where directly received effluents from urban shown that Pb in the F4 fraction was more variable than those measured at S1, S2 and S4. Particularly at S7, situating in the mangrove forest shown that the lead distribution in different fractions was quite similar with all depths.

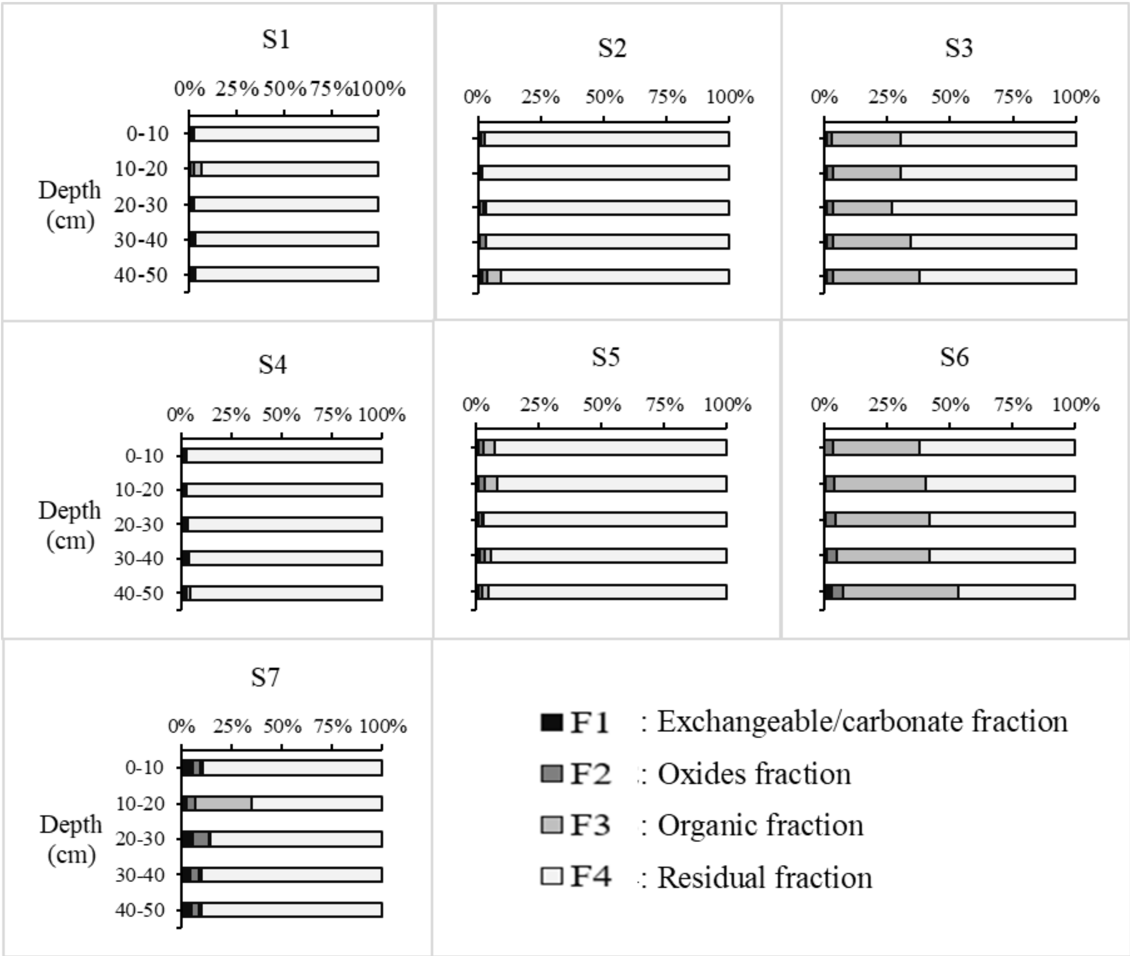


**Figure 3.** *Chemical fraction of lead in sediments*

### *3.3. Assessment of lead ecological risks*

Risk Assessment Code (RAC) is one of the important indices for assessing ecological risk which shows the availability of pollutants in sediment e.g., metals reported firstly by Perin et al. [31] and guidelines from previous publication by Passos et al. [32], Benson et al. [33]. The RAC is calculated based on the percentage of metal concentration that is representative in the bioavailable fraction (exchangeable/carbonate bound) to the total metal content, using the formula (1).

$$RAC = \frac{Metal\ content\ in\ \text{exchangeable/carbonate bound fraction}}{Total\ metal\ content\ in\ soil} \tag{1}$$

According to the RAC, metal content with less than 1% of exchangeable/carbonate fraction would be at no risk to the environment while a higher ratio suggests: 1% - 10%: low risk; 11%-30%: medium risk; 31% - 50%: high risk and > 50%: very high risk. In the present study, the RAC was < 10% at all sites (Figure 4a), of which the lowest value was at site S1 (0.66%) and the highest one was at site S7 (4.52%). The results showed that Pb contamination was at a low-risk level, however, should be taken to avoid activities that increase lead content in sediments.

$I_{geo}$ is a quantitative measure that evaluates the metal origin e.g., from natural and anthropogenic activities, the level of metal pollution in sediments. The results shown that the $I_{geo}$ index at all sampling sites (Figure 4b) was smaller than zero, indicating that there was no sign of lead pollution in the sediments. The results also reflect that anthropogenic activities were minor factors for influence on lead accumulation and pollution in the studied creeks.

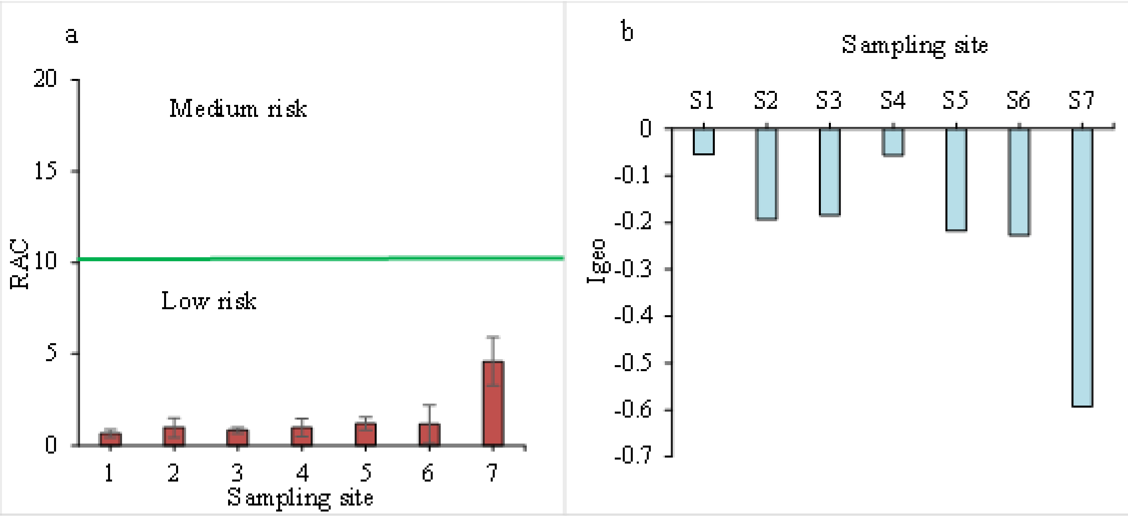


**Figure 4.** *Risk Assessment Code (RAC) and $I_{geo}$ Index of lead*

## 4. Conclusions

Although as main drainages of effluents from aquacultural and urban activities, the lead content in the tidal creeks in the present study ranged from 16.92 to 24.55 mg/kg which did not exceed the allowable threshold according to QCVN 43/2017. The lead partitioning in fractions was in descending: F4 > F3 > F2 > F1. Pb contents varied slightly between sampling sites and mainly concentrated in residual fraction (F4) that implied Pb origin from natural sources during weathering processes. Moreover, compared with Canadian and American standards, the lead content in Can Gio was unpolluted level and did not cause pollution. The geochemical accumulation index Igeo < 0 and RAC index < 10% for all sites indicated that the lead was a minor threat to organisms and human health.

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