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Tracing Heavy Metal Dynamics in Mangrove Sediments: A Study from Ujung Kulon National Park

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Abstract

The dynamics of heavy metal concentrations in the sediments of mangroves are crucial for understanding the health and sustainability of these ecosystems. This study, conducted in a conservation area of Ujung Kulon National Park (UKNP) in Indonesia, investigated the distribution of heavy metals in mangrove sediments, which are vital in trapping these potentially harmful substances due to their absorption, sedimentation, and bioaccumulation capabilities. By analyzing sediment cores from different intertidal zones (interior, fringe, and mudflat) through Inductively Coupled Plasma - Optical Emission Spectrometry (ICP-OES), this study examined concentrations and distribution patterns of six heavy metals: manganese (Mn), copper (Cu), zinc (Zn), cobalt (Co), chromium (Cr), and Iron (Fe). The results indicated that the average concentration of heavy metals followed a decreasing order of Fe> Mn > Zn > Cu > Cr> Co across all locations. All of the heavy metals in the interior and fringe increased gradually from the year 1900 to the present; on the contrary, the mudflat decreased over the same period. Enrichment Factor (EF) and Geo Accumulation Index (I_{geo}) were used to evaluate heavy metal pollution levels comprehensively. The study revealed temporal trends in heavy metal concentrations, highlighting the need for further research to mitigate these contaminant's effects on mangrove ecosystems. By integrating the EF, the I_{geo} index, and quality standards, the research demonstrates that UKNP is likely pristine. Furthermore, this work emphasizes the need to utilize dated environmental archives to reconstruct historical patterns in trace metal pollution in locations where long-term environmental studies remain particularly limited.

Keywords: *mangrove sediment, heavy metals, ICP-OES, geochronology, sediment flux*

Introduction

Mangrove ecosystems are highly productive coastal habitats that provide numerous ecological services, including shoreline stabilization, diverse flora and fauna habitat, and carbon sequestration (Spalding *et al*., 2014; MacKenzie *et al*., 2021; Ramli *et al*., 2022). Among their crucial roles, mangroves act as natural filters for pollutants, including heavy metals, which can be detrimental to both environmental and human health (Bastakoti *et al*., 2018; Kesavan *et al*., 2021). The dynamic interaction between mangroves and their surrounding environments enables these ecosystems to capture and store significant amounts of heavy metals from terrestrial and marine sources.

The sediments found in mangrove ecosystems can serve as a repository for heavy metals that originate from rivers and the sea (Abeywardhana *et al*., 2022). This is due to the fact that the sediment in mangroves ecosystems can capture heavy metals using various mechanisms, including absorption, sedimentation, and bioaccumulation (Analuddin *et al*., 2023; Majumdar *et al*., 2023). These mechanisms allow the sediment to act as a filter, trapping heavy metals and preventing them from spreading further into the ecosystem. Chromium (Cr), copper (Cu), zinc (Zn), and cobalt (Co) are frequently analyzed in studies of mangrove ecosystems due to their roles as key indicators of industrial and agricultural pollution (Shen *et al*., 2019; Hu *et al*., 2024). Additionally, manganese (Mn) and iron (Fe) also contribute valuable information for pollution indices, aiding in the assessment of the impact of human activities on these vital environments (Kowalska *et al*., 2018; Ranjan *et al*., 2018). Investigating the concentrations and impacts of these metals is critical for monitoring pollution levels and evaluating the associated environmental risks within mangrove ecosystems.

Research in Indonesia shows that mangrove sediments contain elevated levels of heavy metals,

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such as lead, cadmium, and mercury (Hamuna and Wanimbo, 2021; Sastranegara *et al*., 2021; Analuddin *et al*., 2023). These heavy metals can come from various sources, including industrial activities, mining, and urbanization (Hossain *et al*., 2023; Rahman *et al*., 2024). The accumulation of heavy metals in mangrove sediments can negatively affect the health of the mangrove ecosystem and the living creatures that rely on the mangroves for food and other resources. While current research has extensively examined surface sediment concentrations, there remains a significant gap in understanding the vertical distribution of these contaminants. This underscores the necessity for further studies to gauge the full extent of heavy metal pollution in Indonesian mangrove sediments and to develop effective strategies for mitigating its impact.

A study investigating the vertical trend of heavy metals (Co, Cr, Fe, Hf, Sb, Sc, Se, and Zn) in mangroves within protective areas was conducted in Nusa Lembongan, Indonesia using Neutron Activation Analysis (Lubis *et al*., 2023). The findings indicated that all detected heavy metals likely originated from natural sources. However, it is important to note that the sampling site was not strategically chosen based on the intertidal zone. In a related study, Shintianata *et al*. (2024) examined sediment accumulation rates in Ujung Kulon National Park (UKNP) to assess carbon accumulation. In this paper, the vertical distribution of trace metals (Mn Cu, Zn, Co, Cr, and Fe) in the same sediment cores dated by Shintianata *et al*. (2024) is analyzed using ICP-OES. This investigation into the vertical profile of heavy metals in the mangrove sediments in UKNP represents a significant step towards a comprehensive understanding of heavy metal contamination levels. Studying these metals helps monitor pollution and assess environmental risks in mangrove ecosystems. The selected sites represent diverse intertidal zones, providing a comprehensive view of heavy metal pollution in UKNP. Furthermore, this research stands as the first study to analyze the extent of heavy metal pollution in UKNP, paving the way for the development of more effective contamination mitigation strategies

Materials and Methods

Ujung Kulon National Park (UKNP) is a vital conservation area and habitat for the endangered Javan rhinoceros, which has a population of fewer than 100 individuals (Haryono *et al*., 2015). Located in Banten at the western tip of Java, the park spans 105,694.46 ha, as indicated in Decree No. SK. 3658/Menhut-VII/KUH/2014 from the Ministry of Forestry. The region has a tropical climate, with heavy rainfall from October to April during the northwest monsoon and a drier period from May to September during the southeast monsoon. Temperatures typically range from 25 to 30 °C, with relative humidity between 65-100% (Ramono *et al*., 2009). Mangrove ecosystems thrive along the northern coast, supporting various species, such as *Lumnitzera racemose, Avicena* spp*, Soneratia alba, Bruguera* spp*, Nypa fructicans, Rhizopora* spp*,* and *Acrostichum aureum* (Statistik 2023 Balai Taman Nasional Ujung Kulon, 2023). A previous study conducted near the UKNP in Suralaya, Banten, investigated the concentrations of several heavy metals, including U, Th, Cr, Hf, As, Sc, Zn, and Co, in sediment samples. The results revealed a gradual increase in the levels of chromium and arsenic with sediment accumulation, while the concentrations of uranium, thorium, hafnium, and cobalt remained relatively constant (Arman *et al*., 2013). Given its protected status of UKNP, it can provide valuable insights into heavy metal accumulation, offering a relatively undisturbed baseline that is crucial for conservation efforts and assessing anthropogenic impacts over time.

Figure 1. Map of sampling sites in UKNP highlighting three distinct intertidal areas: interior, fringe, and mudflat

Sampling was carried out in three different locations as depicted in Figure1: interior (6°49'31.3"S, 105°26'37.9"E), fringe (6°49'29.9"S, 105°26'40.9"E), and mudflat (6°49'28.0"S, 105°26'44.7"E covering a range of intertidal zones. The interior is characterized by dense vegetation and represents the highest point. It experiences less frequent tidal inundation, with waterlogging occurring only during high spring tides. In contrast, the mudflat, which has significantly less vegetation (dominated by *Avicennia spp*), is the lowest point in this landscape. It is completely submerged during high tide and exposed during low tide. The fringe zone, which is subject to daily tidal flooding, serves as a midpoint along this intertidal gradient.

Sample analysis

Three sediment core samples were collected from three distinct locations using a sediment corer (Eijkelkamp Gouge). The lengths of cores were 78 cm, 54 cm, and 54 cm of interior, fringe, and mudflat, respectively. Sediment cores were sub-sampled by slicing every 2 cm on the upper of 10 cm and 4 cm afterward to prevent mixing between samples. There were 45 total samples. Each sample was stored in plastic clips and labeled according to its depth before being sent directly to the laboratory in an icebox. Upon arrival at the laboratory, the sediment core samples were first oven-dried at 60°C, and their water content was measured both before and after drying until a consistent dry weight was achieved. After drying, the samples were crushed using a mortar and pestle. Five grams of the dried sediment was then combined with tracer Po-209 and dissolved using hydrochloric acid (HCl), nitric acid (HNO3), and water, along with H_2O_2, before being dried over a water bath. Next, HCl and distilled water (aquadest) were added to the dried sample and heated for ten minutes. The sample was then filtered using Whatman filter paper 42 and washed with 0.3N HCl. The filtrate was dried over a water bath. Once dry, a mixture of 1:1 HCl and 0.3N HCl was added until the solution volume reached 50 ml. From this solution, 2 ml was extracted for metal analysis and stored in a glass vial, while the remaining solution was used to analyze Pb-210. The solution was platted on the copper plate for two hours. The results in the copper plate were then analyzed using the Alpha Spectrometer.

The concentrations of six different metal elements (Mn, Cr, Zn, Cu, Co, and Fe) in the solution were determined using ICP-OES (Inductively Coupled Plasma - Optical Emission Spectrometry) Thermo Scientific iCAP 700 Series. For quantitative determination, ICP multi-element standard solution IV was used. The precision of the analytical method was consistently verified by testing samples of reference material SRM 158 (marine sediment) supplied by the International Atomic Energy Agency (IAEA). Both the reference samples and the standard sediment were treated in the same way. The obtained concentrations for certified reference material of heavy metals were close to the reported values with deviations of <10 %. The sediment dating and sediment accumulation rate (SAR) of each core was performed using excess Pb-210 activity and an alpha spectrometer obtained from Shintianata *et al*. (2024).

The heavy metal flux was calculated by multiplying the sediment accumulation rates (SAR) with sediment dry bulk density (DBD) and heavy metal concentration (HM) as follows:

$$Heavy\ Metal\ Flux\ (\mu g\ cm^{-2}\ year^{-1}) = SAR\ (cm\ year^{-1}) \times DBD\ (\ g\ cm^{-3})\ \times HM\ (\mu g\ g^{-1}) \qquad (1)$$

Pollution assessment

The Enrichment Factor (EF) and Geo Accumulation Index (I_{geo}) were used for pollution assessment to determine the level of heavy metal pollution in UKNP. These measures have been used widely in several countries, such as Indonesia (Umroh *et al*., 2023), China (Tang *et al*., 2022; Hu *et al*., 2024), Malaysia (Khan *et al*., 2020; Krishnan *et al*., 2022), and South America (Fernández-Cadena *et al*., 2014). Muller (1979) developed I_{geo} as a commonly used approach to evaluating the degree of heavy metal contamination in sediments. The following is the formula used for calculation.

$$I_{geo} = \log_2(C_n/1.5B_n) \qquad (2)$$

Where C is the measured concentration of the heavy metal in the sample and B is the concentration of the heavy metal on the local background.

EF was used to determine whether heavy metals found in coastal sediments come from human activities or natural processes. The EF calculation followed the equation as below (Krishnan *et al*., 2022; Xie *et al*., 2022).

$$EF = \frac{\left(\frac{M_x}{M_y}\right)_{sample}}{\left(\frac{M_x}{M_y}\right)_{background}} \qquad (3)$$

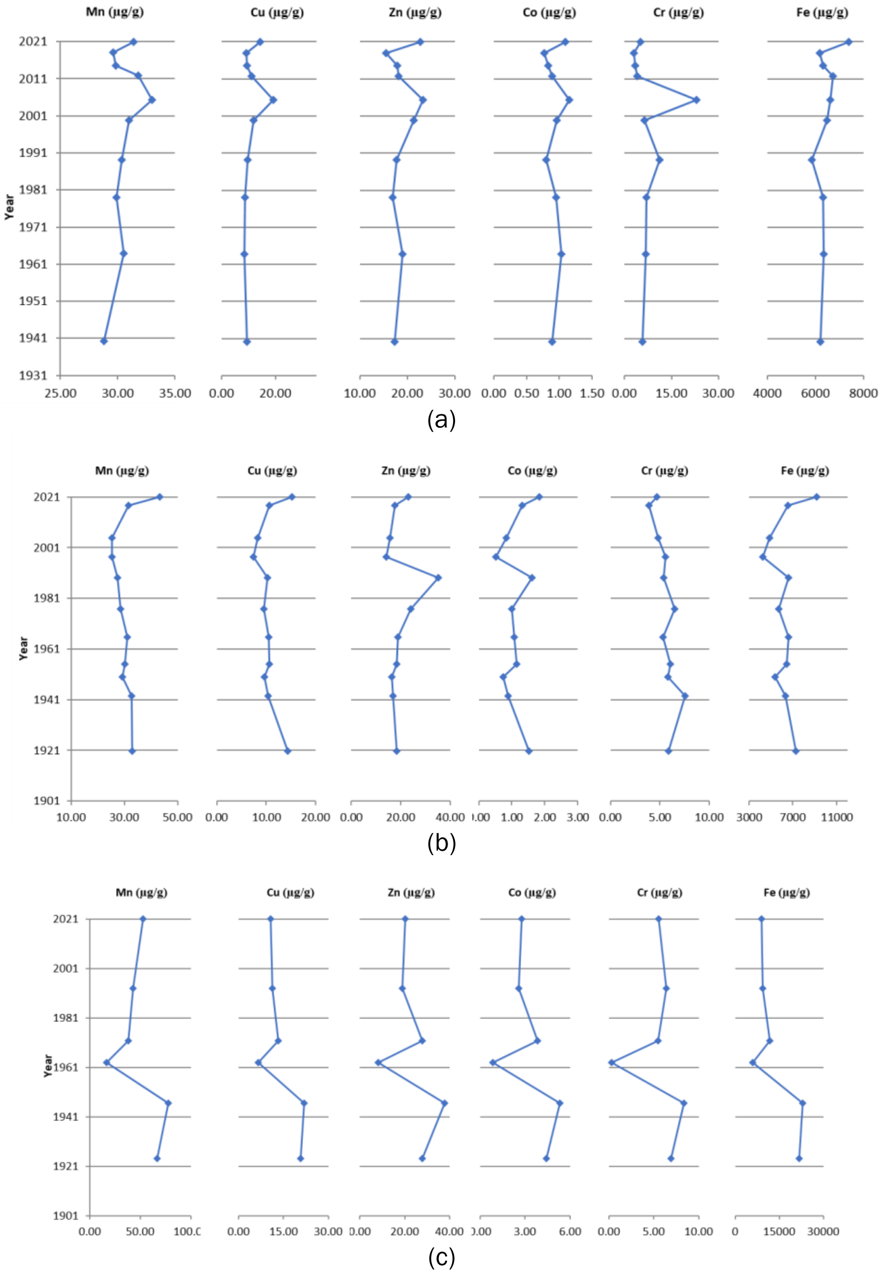
where M_x refers to heavy metal concentration, whereas M_y is the concentration of the reference baseline. Al and Fe are frequently utilized as references in EF calculation, primarily due to their abundant presence on Earth and their notable chemical stability (Kolibongso *et al*., 2017; Krishnan *et al*., 2022; Xie *et al*., 2022). Specifically, Fe was selected as the reference element in this research.

Results and Discussion

Heavy metal concentration and flux

The average concentrations of Mn, Cu, Zn, Co, Cr, and Fe in the interior, fringe, and mudflat are summarized in Table 1, with Fe being the most abundant element, followed by Mn, Zn, Cu, Cr, and Co, in that order. Specifically, the interior had Mn, Cu, Zn, Co, Cr, and Fe in average concentrations of 30.64, 11.01, 18.95, 0.94, 7.56, and 6437.46 µg.g^{-1}, respectively; the fringe had 30.62, 10.61, 19.85, 1.14, 5.58 and 6294.10 µg.g^{-1}, respectively; and the mudflat had 50.32, 12.95, 22.41, 3.12, 5.54 and 11886.34 µg.g^{-1}, respectively.

The historical trends of heavy metals in the interior, fringe, and mudflat are shown in Figure 2. Overall, most all metal shows a gradual increase in interior and fringe, but fluctuating in the mudflat. In the interior, the trends of Mn, Cu, Zn, Co, and Cr were similar, with a gradual increase from 1940 to 2005, followed a decrease after 2005. Meanwhile, Fe showed different trends with gradual increases, with the maximum peak observed at the surface.



(a)

(b)

(c)

Figure 2. Vertical heavy metal deposition over time in UKNP (a)interior, (b) fringe, and (c) mudflat

Similar to the interior, Mn, Cu, and Zn increased over time in the fringe, with Mn and Cu peaking at the surface in 2021 and Zn in 1989. The trends for Co and Cr showed fluctuations over time. This vertical distribution indicated that the heavy metals in the interior and fringe had increased gradually in the past few years. On the other hand, all heavy metals in the mudflat had similar trends, with the lowest peak in 1963. Contrary to the interior and fringe, the vertical distribution in the mudflat revealed an overall decrement in heavy metal concentrations, suggesting divergent depositional and post-depositional behaviors in this zone compared to the others. The reason might be because of the dynamic nature of mudflats, which are significantly influenced by tidal movements. These tidal actions could lead to the Fe had the highest concentration in all locations, consistent with its natural abundance on Earth as a major component of mineral rocks (Lubis *et al*., 2023). Meanwhile, Co had the lowest concentration in all zones, similar to the findings from Sepang Besar, Malaysia (Krishnan *et al*., 2022). Co is often introduced into ecosystems through industrial activities (Xu *et al*., 2015). The low concentration of

Table 1. Summarizes and comparison the concentrations of trace metals (Mn, Cu, Zn, Co, Cr, Fe) in sediment samples from mangrove sites in Indonesia and other countries

Location	Sediment	Mn (µg.g^{-1})	Cu (µg.g^{-1})	Zn (µg.g^{-1})	Co (µg.g^{-1})	Cr (µg.g^{-1})	Fe (µg.g^{-1})	Reference
This Study	Interior	28.34-33.00	8.33-19.07	15.50-23.26	0.77-1.15	2.95-22.83	5837.40-7376.73	
		(30.64)	(11.01)	(18.95)	(0.94)	(7.56)	(6437.64)	
	Fringe	25.28-43.20	7.42-15.23	14.20-35.15	0.52-1.84	3.90-7.52	854.20-1932.02	
		(30.62)	(10.61)	(19.85)	(1.14)	(5.58)	(6294.1)	
	Mudflat	16.69-77.80	6.50-24.80	8.04-37.80	0.85-5.37	0.33-22.83	5851.05-22882.88	
		(50.32)	(12.95)	(22.41)	(3.12)	(5.54)	(11886.34)	
Bangladesh	Core 1		24.0-31.6	49.1-11.9		56.9-124		(Islam *et al*., 2024)
			(28.8)[3.6]	(65.4)[69.0]		(85.0)[118]		
	Core 2		18.4-26.2	42.6-63.0		49.3-95.1		
			(23.2)[23.5]	(56.0)[58.4]		(71.5)[71.2]		
Kendari Bay - Indonesia	Surface	10.83-17.77	6.85-9.65	8.60-10.48				(Analuddin *et al*., 2023)
		[14.16]	[8.2]	[9.6]				
Tinanggea - Indonesia	Surface	5.98-9.95	2.03-4.09	4.62-6.40				(Analuddin *et al*., 2023)
		[8.37]	[3.07]	[5.31]				
Shankou - southern China	Surface		2.99-34.85	13.88-130.22	1.19-23.25	16.03-222.88		(Xie *et al*., 2022)
			[15.3]	[64.82]	[6.04]	[61.72]		
Sepang Besar River - Malaysia	Surface				1.43-3.12	5.34-15.82		(Krishnan *et al*., 2022)
					[2.44]	[11.62]		
Cochin - India	Core 1	9.3-136.7	2.1-27.0	11.3-84.9				(Passos *et al*., 2022)
		(72.7)	(17.1)	(45.3)				
	Core 2	9.3-171.8	2.1-27	11-95.9				
		(102.7)	(19.92)	(60)				

*The concentration values within the first brackets are mean values and data within the third brackets are concentrations of the corresponding metals in the surface sediments of the core periodic flushing and redistribution of sediments, thereby reducing heavy metal accumulation at depth.

Co in this site can be attributed to the region's designation as a conservation area, which reduces industrial and anthropogenic influences.

Mn recorded the second-highest concentration among all the metals examined in this study. Furthermore, its concentration was the highest when compared to Mn levels observed in other regions. The average concentration of Mn in the mudflat was almost two times higher than in the interior and fringe. Moreover, the average value in this site was higher than research conducted in Kendari Bay (14.16 μg.g^{-1}) and in Tinanggea (8.37 μg.g^{-1}), which is located around a marine pond and a nickel mining site (Analuddin *et al*., 2023), indicating there is a possibility of contamination Mn in the mudflat.

In contrast, Cu, Cr, and Zn concentrations were lower than those reported in the Shankou Mangrove National Nature Reserve in southern China, where Zn, Cr, and Cu mean concentrations were 64.82 μg.g^{-1}, 61.72 μg.g^{-1}, and 15.30 μg.g^{-1}, respectively (Xie *et al*., 2022). Elevated levels of Zn, though essential for plant growth, can be harmful to certain organisms (MacFarlane *et al*., 2003). Additionally, the presence of Cu, Cr, and Zn is frequently documented in industrial discharges and, to a lesser degree, in household wastewater (Kim *et al*., 2016; Hu *et al*., 2024).

The concentrations of most heavy metals in this study were notably low compared to other areas, as shown in Table 1, which might be attributed to the site's location within a conservation area characterized by minimal anthropogenic activities. Moreover, the observed concentrations of Cu, Zn, and Cr in the studied sediments are notably below the established threshold values defined by Washington State's Marine Sediment Quality Standards (WAC 173-204-320). Specifically, these standards set the permissible upper concentration limits for Cu, Zn, and Cr at 390 μg.g^{-1}, 410 μg.g^{-1}, and 260 μg.g^{-1}, respectively (Marine Sediment Quality Standards, 2013). This indicates that the levels of these metals across the evaluated areas did not exceed the defined environmental safety criteria. Moreover, the lower values of heavy metal concentrations indicate that UKNP does not bear strong human influence, which is possible because it is a conservation area.

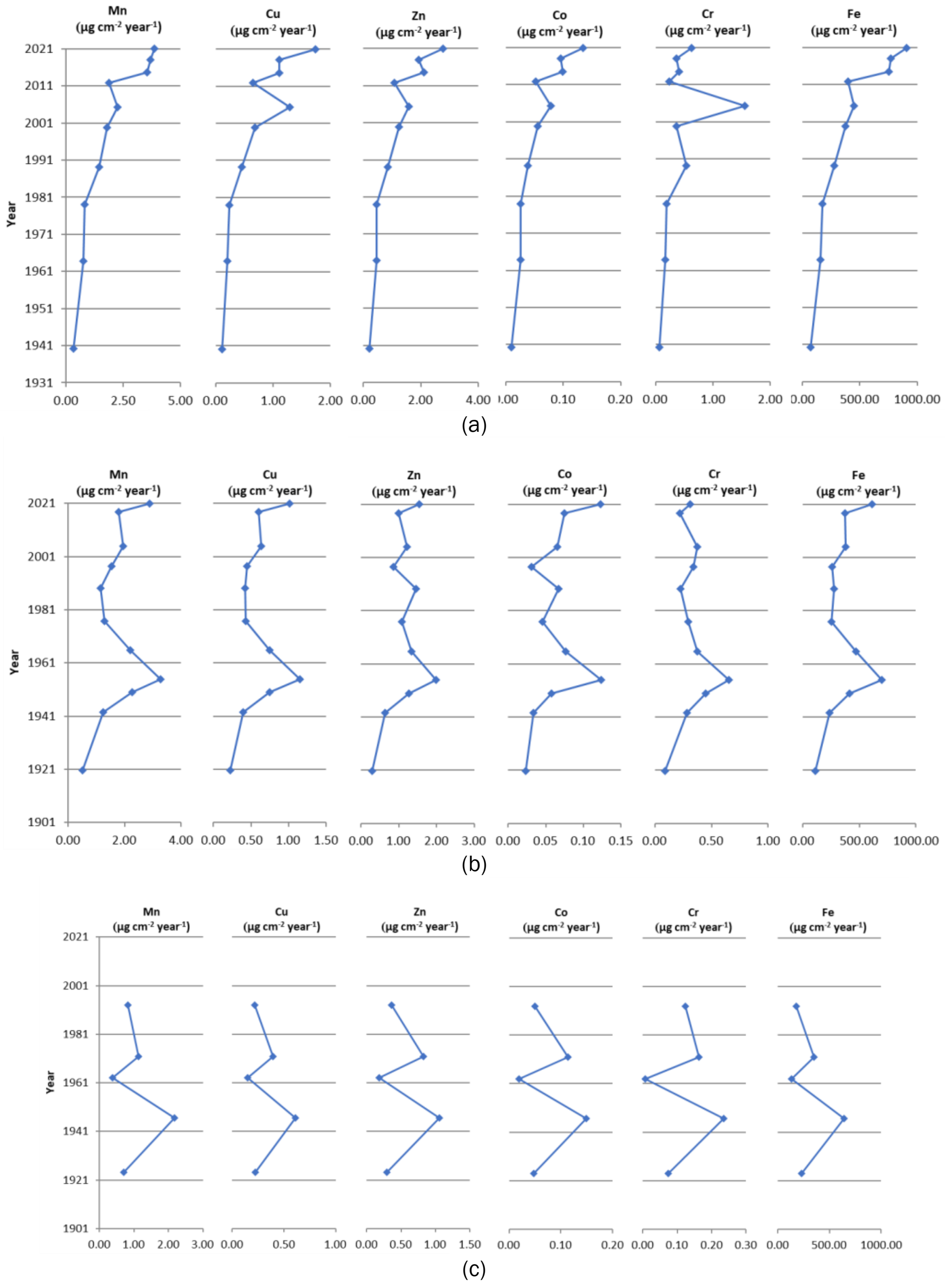
The data presented in Figure 3 illustrates the trend of heavy metal flux over time. All six heavy metals in the interior show a significant increase in flux over time, particularly from 1979 onward, indicating increased inputs of these metals into the environment. The marked increase in heavy metals after 1981 is possibly due to intensified industrial activities and urban growth in many regions, particularly around Southeast Asia. Additionally, studies conducted in Southeast Asia also indicate a potential rise in pollution from fossil fuel usage since 1960 (Engels *et al*., 2018). The flux of Mn and Fe showed a relatively steady increase, with a peak between 2012 and 2021. Cu, Zn, and Co have a similar flux trend, which remained low until 1979. After that, there was a small but noticeable rise until 2005 and continued fluctuating until 2021. Cr flux remained low until about 1979, but from 1979 to 2000, there was a noticeable rise, with fluctuations afterward.

On the other hand, the fringe showed a unique pattern, with a peak in heavy metal around 1955, followed by a decline until 1977. After that, the flux gradually rises, with a notable increase in 2021. In contrast with interior and fringe, all heavy metal flux in mudflat fluctuated, starting low in 1924, rising steadily until 1947, dipping around 1961, and then increasing again by 2001. This fluctuating trend, possibly influenced by tidal action, suggests that tidal dynamics play a significant role in redistributing heavy metals within the sediment. The steady rise from 1924 to 1947 could also be linked to human activities, while the dip around 1961 might indicate a period of reduced metal input or increased sediment flushing. The increase by 2001 could be related to changes in hydrodynamics, land use, or industrial processes that once again heightened metal deposition in the mudflat.

Polution assesment of heavy metal

The values of Enrichment Factor (EF) are shown in Figure 4. Values of EF less than 1.5 signify that the metal is sourced solely from crustal contribution, whereas values greater than 1.5 indicate the presence of a considerable amount of non-crustal sources of the element (Fernández-Cadena *et al*., 2014). The respective average EF values for Mn, Cu, Zn, Co, and Cr in the interior were 1.03, 1.15, 1.06, 1.02, and 1.30. Meanwhile, the same was observed in the Fringe, with total averages of EF of 1.10, 0.84, 1.29, 0.83, and 1.16, respectively. The average EF value in the interior and fringe of all heavy metals were under 1.5, which makes it possible to assume the heavy metal were provided from a crustal source. In contrast, the EF values for mudflat were mostly above 1.5, with respective average values for Mn, Cu, Zn, Co, and Cr being 1.54, 1.22, 1.61, 1.37, and 1.61. These results indicate that most metals (Mn, Zn, and Cr) in the mudflat are from non-crustal sources; hence, there is a possibility that these metals are caused by anthropogenic pollution. This is supported by the higher value of Mn found in the mudflat compared to other areas.

The value of Geoaccumulation Index (I_{geo}) are shown in Figure 5. I_{geo} values in the interior region were negative, ranging from -0.9 to -0.53. The average I_{geo} value of heavy metals in the interior region followed a decreasing order of Cu>Cr>Zn>Mn>Co>Fe. However, some results for Cr and Cu exceeded 0, with the highest values being 1.41 and 0.45, respectively. On the other hand, the average I_{geo} values in the fringe followed a decreasing order of Zn>Cr>Mn>Fe>Cu>Co. I_{geo} results in the fringe region mostly showed negative values except for Zn in 1989, with an I_{geo} value of 0.35. In contrast, the mudflat's I_{geo} values for heavy metals were all negative and following a decreasing order of Zn>Mn>Cr>Co>Cu>Fe. Overall, the results of I_{geo} calculations of heavy metals in the interior, fringe, and mudflat mostly showed negative values with averages of -0.81, -0.48, and -1.29, respectively. These values indicate that the sediment was unpolluted with heavy metals. Similar results were reported in Matang Mangrove Forest Reserve research, with average I_{geo} values for all heavy metals of less than 0 (Khan *et al*., 2020).



(a)

(b)

(c)

Figure 3. Heavy metal flux over time in UKNP (a)interior, (b) fringe, and (c) mudflat

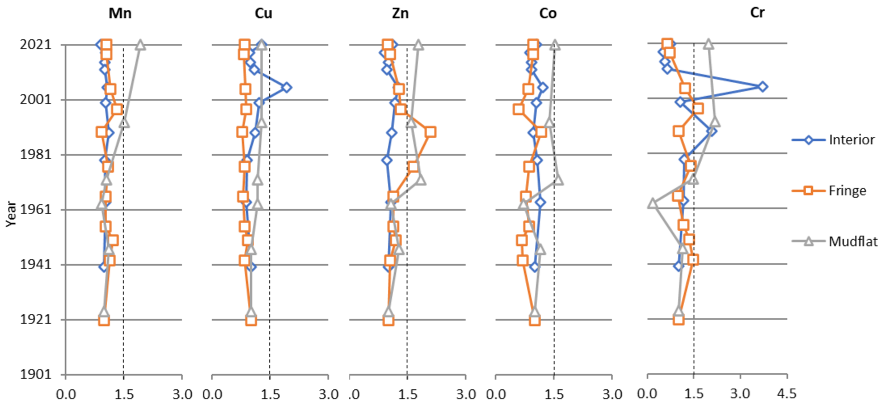


Figure 4. Profile of Enrichment Factor (EF) values of the metals over time in Interior, Fringe and Mudflat

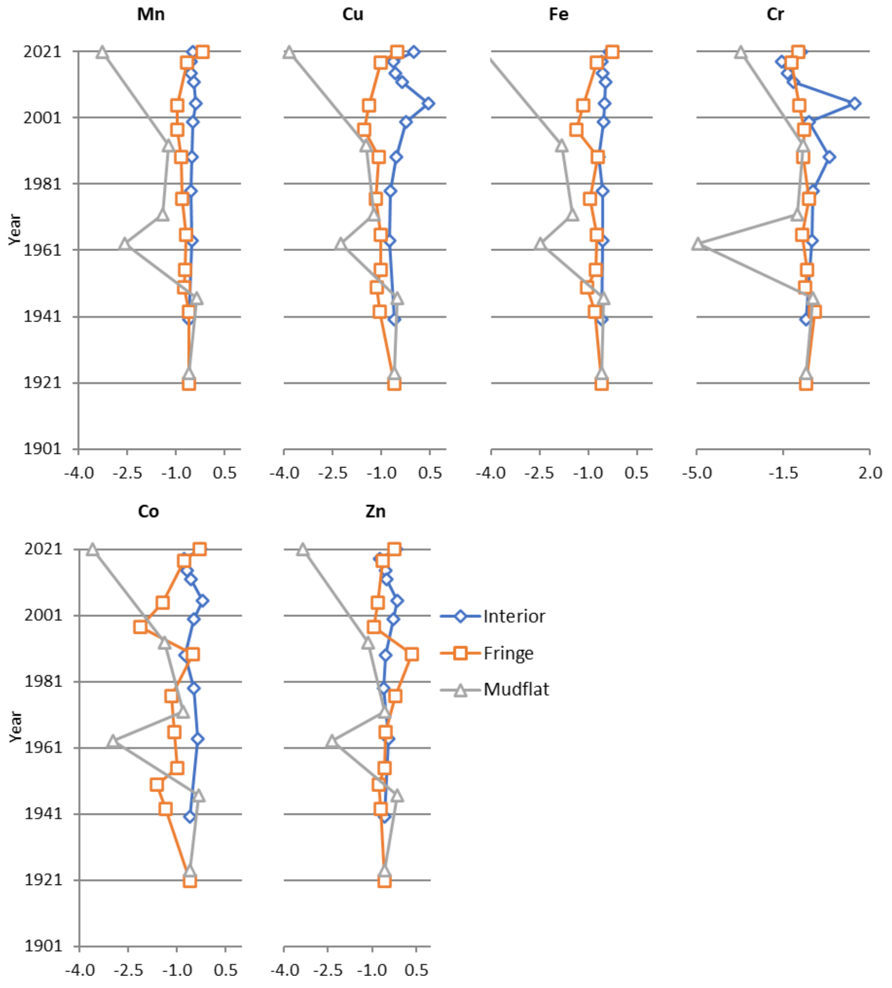


Figure 5. Profile of Geoaccumulation Index (I_{geo}) over time of each metal in Interior, Fringe, and Mudflat

Conclusion

The study conducted in Ujung Kulon National Park provided a detailed insight into the dynamics of heavy metal accumulation within mangrove sediments. Through the analysis of sediment cores from various intertidal zones, it was discovered that these sediments acted effectively as environmental filters, sequestering heavy metals like manganese

(Mn), copper (Cu), zinc (Zn), cobalt (Co), chromium (Cr), and iron (Fe). This is significant because these metals, originating from both natural and human activities, can be harmful to the ecosystem and human health if allowed to circulate freely. Fe was identified as the most abundant metal across all samples, indicating its natural prevalence and perhaps its role in the environmental dynamics of the study areas. The study's use of pollution assessment measures such as the Enrichment Factor (EF) and the Geo Accumulation Index (I_{geo}) further clarified the extent to which these sediments were impacted by heavy metal pollution. Combining the EF, I_{geo}, and quality guidelines, this study indicated that UKNP mostly presents pristine conditions, highlighting the effectiveness of protected area management in mitigating anthropogenic impacts. However, the higher contamination levels in the mudflat warrant further investigation to identify potential sources and mitigate their effects. While the research underscores mangroves' role in capturing and storing heavy metals, it also highlights the necessity of ongoing monitoring and research. Understanding the full scope of heavy metal contamination, its sources, and effective mitigation strategies is crucial for preserving the health of mangrove ecosystems, which are vital for biodiversity conservation and coastal protection.

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