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ASSESSMENT OF METAL CONTENTS AND LEAD ISOTOPE FINGERPRINTS IN SOUTH CHINA SEA SEAWATER AND SEDIMENTS

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Abstract

To investigate both anthropogenic and natural sources of metals in the South China seawater, the study analyzed metal content and Pb isotopic compositions in surface sediment and seawater from Taiwan and Indonesia. This analysis aimed to assess potential environmental pollution in the region. The dryness sediment sample was characterized of metal content mapping and distribution using SEM-EDX and was leached using a microwave digestion for metal contents and Pb isotope ratios ($^{207}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$) using ICP-MS. The distribution of metal contents in sediments from South China ranged from lowest to highest concentrations as follows: Na (0.4–3.8 at.%), Mg (0.6–1.2 at.%), Al (4.2–7.3 at.%), Si (14.4–20.6 at.%), K (0.7–1.8 at.%), Ti (0.3–0.4 at.%), Ca (0.2–1.1 at.%), and Fe (1.3–2.0 at.%). The contents of Na, Mg, Al, Si, K, Ti, Ca, and Fe in South China Sea sediments fall within the natural background range of marine sediments and do not indicate anthropogenic enrichment. The Pb isotope ratios of the seawater samples reveal distinct patterns and variations among the 5 groups. Notably, these samples deviate from the typical Pb growth curve. This suggests that Pb in unpolluted seawater may originate from natural sources, such as freshwater input and mineral weathering within local sediments. Furthermore, seawater around Taiwan has Pb isotope ratios of ~1.15–1.17, suggesting a strong mixing signal from Pb derived from Malaysia, Indonesia and China.

Keywords: *South China Seawater; Sediment; Metal contents; Pb isotopes; Natural source*

Introduction

Anthropogenic activities such as ore mining, metal smelting, fossil fuel combustion, and machine use in big cities are inputs of anthropogenic activities of big cities that indirectly pollute the regional, national, and international environment. This can be proven by research that informs the accumulation of heavy metals in various environmental media [1], [2]. Roadside dust is the first media that records contaminants of anthropogenic activities of cities through a combination of air and wind that settle in sediments. Sediment is the main indicator for assessing heavy metal pollution in aquatic environments. Metal contents in sediments can be released into water when chemical reactions occur [3]. Therefore, metal contents in sediments can pose a threat to the safety

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of water quality and aquatic organisms [4]. One of the fastest-growing cities in Southeast Asia is the city of Kuala Lumpur, which is famous for its steel and glass structures [5], [6]. Based on their geographical area, the cities of Kuala Lumpur and Jakarta are occupied by various industrial/trade activities, traffic loads, and development in a number of residential areas. The release of pollutants from the urbanization process into the surrounding environment is believed to pose a significant threat to humanity [7]. One of the main pollutants in urban areas today is heavy metals. To overcome this pollutant, it is important to know the source of the pollutant using Pb isotopes for anthropogenic sources and Sr isotopes for natural sources [2], [5], [8]. In addition, roadside dust and sediment are believed to provide a reflection of environmental conditions related to the health of marine biota [9]. Several researchers have shown that roadside dust data comes from urban emissions of industrial exhaust, household heating, waste burning, and agricultural side effects [10].

Lead (Pb) contamination in the atmosphere has a long historical presence, with significant increases during the 20th century [11]. The rise in Pb levels within aquatic environments, largely driven by escalating human activities, is a global concern [12]. Identifying Pb sources is essential for effective pollution assessment. Due to the isotopic stability of Pb in sediment, even amid ongoing physical and chemical processes, Pb isotopes have become a powerful tool for source identification [13]. Studies suggest that alternative Pb sources gained prominence after the phase-out of leaded gasoline between the 1970s and 2000 [14]. In China, rapid industrialization and high coal consumption maintained elevated Pb levels in aerosols, even post-phase-out [15]. Since the 1980s, anthropogenic Pb emissions from China have likely been transported atmospherically to the western Pacific's surface [13], [14], [15]. As marine sediments act as significant Pb sinks, capturing historical trends, Pb isotopic analysis in China's marine sediments has become a focus for researchers [14]. Some researchers analyzed Pb in South China Sea surface sediments and found no significant anthropogenic Pb pollution [15]. However, in coastal areas like the Pearl River Estuary, anthropogenic Pb represented about 30% of sediment Pb in the 1990s [14]. Currently, Pb isotopic data are still lacking for the northwestern South China Sea and nearshore regions heavily affected by human activity, which await further evaluation.

The selection of Chu Wei Beach in Taoyuan, Taiwan, as a sampling site for metal contents and Pb isotope analysis is well-suited due to its strategic location near heavily trafficked maritime routes in East Asia and the significant influence of industrial activities and urbanization in the area. This region is critical for studying pollutant distribution, as sediment transport here is influenced by the Kuroshio Current circulation system, allowing insights into pollutant spread over broader areas, including the South China Sea. Geographically, Chu Wei's coast features environmental characteristics favorable for sediment accumulation relevant to this analysis, such as tidal dynamics and diverse coastal substrates. Although it is geographically distant from the South China Sea, comparing these two locations provides essential insights into pollutant sources and heavy metal distribution within environments impacted by human activities. Additionally, this site benefits from prior studies, offering valuable baseline data for comparative analyses and long-term pollution trends.

Chuwei Beach is located in Dayuan District, Taoyuan City, Taiwan, facing the Taiwan Strait on the island's northwest coast, approximately at coordinates 25.0885° N, 121.2386° E. This beach features an extensive, natural coastal environment with a substrate ranging from sandy to muddy and is home to mangrove vegetation that supports the local ecosystem. Despite its proximity to urban centers and Taoyuan International Airport, the area has retained its natural character, though it is influenced by nearby industrial and residential activities. The subtropical climate, with high rainfall, affects erosion and sedimentation processes along the coast. Chuwei

Beach is easily accessible and popular for recreation, yet it also serves as a vital site for environmental research, particularly in studies of pollutant distribution and coastal ecosystem changes.

The selection of Chu Wei Beach in Taoyuan, Taiwan, as a site for mapping metal contents and Pb isotope analysis in sediments and for examining interactions with sediments from the South China Sea is grounded in several key scientific considerations. Situated near the CPC Corporation's Sharon Filling Plant, a large industrial facility handling various fuels like LPG, gasoline, and diesel, this location is at high risk for heavy metal contamination. Emissions and potential leaks from these industrial operations can elevate heavy metal concentrations, including Pb, in coastal sediments. Given the intensive industrial activities surrounding this area, Chu Wei Beach serves as a strategic point to study the impact of industrial pollution on the coastal environment.

The strategic geographic position of Chu Wei, near busy maritime routes and the South China Sea, enhances the research value of this site. This location enables studies on the distribution of heavy metals that may be influenced by maritime activity and discharge from major rivers emptying into the South China Sea. As part of a sensitive coastal ecosystem, sediments in this area frequently accumulate pollutants from urban areas through surface runoff and river flow. Taoyuan's status as a rapidly urbanizing region adds to the pollution potential, making Chu Wei an ideal site for analyzing how urban and industrial activities impact the marine environment. Research at this site also has significant implications within the context of global environmental studies, especially concerning cross-boundary pollutant transport. By comparing data from diverse locations like Kuala Lumpur, Jakarta, and Chu Wei, along with sediments from the South China Sea, this study aims to build a comprehensive database on the distribution and sources of heavy metals across various environmental types. This research will help identify key contamination sources and understand heavy metal transport mechanisms and their effects on broader ecosystems. Therefore, selecting Chu Wei Beach as a research site is crucial not only for regional studies but also for gaining insights into the global impact of pollutants on ecosystems.

Experimental part

Sampling

The collection of 32 sediment and 24 seawater samples from South China seawater close to Chu Wei Beach, Taiwan, located near Zhubei Fishing Harbor, and The Gulf of Prigi, Indonesia, respectively, was conducted to gain a comprehensive view of pollutant distribution, specifically metal contents and Pb isotopes, in a coastal area influenced by maritime and fishing activities (Fig. 1). These sites were chosen because harbors often serve as potential contamination sources, from shipping activities and seafood processing to runoff carrying industrial and urban pollutants. Sampling sediments and seawater around the harbor allows for an in-depth analysis of how human activities impact coastal environmental quality. Additionally, the varied sampling locations from areas surrounding the harbor to points further offshore provide rich data for mapping the spatial distribution of contaminants and understanding sediment deposition dynamics in relation to ocean currents and coastal interactions. With a total of 56 samples, the data were collected to be sufficiently representative to analyze pollutant accumulation patterns and offer insights into the environmental impact of fishing and maritime activities in this region.

Fig. 1. Sampling point location

Distribution and Elemental Mapping by SEM-EDX Analysis

The sediment samples were dried at 60–80°C for 2–4 hours to remove moisture prior to SEM-EDX analysis using an FEI Inspect S50 operated in low-vacuum mode. A small amount of the dried sample was mounted onto an aluminum stub using double-sided conductive carbon tape. To minimize charging effects, a thin carbon coating of approximately 10–20 nm was applied using a sputter coater. Carbon coating was preferred over gold coating when analyzing Na, Mg, Al, Si, K, Ti, Ca, Fe, and Pb to avoid peak interference with Au. The accelerating voltage was set to 15–20 kV with a working distance of 10 mm to optimize both imaging resolution and X-ray generation for elemental detection. After imaging, elemental analysis and mapping were performed using the EDX detector. For elemental mapping, a region of interest was selected and target elements including Na, Mg, Al, Si, K, Ti, Ca, Fe, and Pb were defined in the EDX software. The mapping resolution was set to 256 × 200 or 512 × 400 pixels with a dwell time of 100–500 µs per pixel. The total acquisition time ranged from 15 to 60 minutes depending on the required signal-to-noise ratio. During acquisition, the software collected characteristic X-rays from each pixel to generate 2D distribution maps for each element. Following mapping, semi-quantitative data in wt.% and at.% were obtained using ZAF correction. The resulting maps, spectra, and quantitative data were then exported to evaluate the spatial distribution and elemental associations within the sample matrix.

Analysis Method

Samples selected for grain size analysis were pretreated with 10 mL of 30% H_2O_2 and 3N HCl to remove organic matter and carbonates. Next, 5 mL of a 0.5 mol/L sodium hexametaphosphate solution was added to prevent flocculation. The samples were then dispersed and homogenized using ultrasonic agitation for 30 seconds before analysis with a laser grain size analyzer (MAM 5005, Malvern Ltd., UK) covering a measurement range of 0.2-2000 µm.

Pb content was measured using inductively coupled plasma mass spectrometry (ICP-MS, X Series II, Thermo Fisher Scientific Inc.). Frozen, dried, and ground samples were fully digested in Teflon vessels with HF, HNO_3, and $HClO_4$ before analysis. Analytical procedures and quality control protocols followed Zhang et al. (2019) using standard sample GBW07309 (GSD-9, Institute of Geophysical and Geochemical Exploration, Chinese Academy of Geological Sciences) to ensure analysis accuracy, with concentration discrepancies between measured and certified values under 5%.

Stable Pb isotope ratios were determined using a high-resolution multi-collector ICP-MS (MC-ICP-MS) reference according to methods from Zhang et al. (2019). Briefly, approximately 0.2 g of sediment was digested with HF, HNO_3, and $HClO_4$ in a closed Teflon vessel at 190°C. The resulting dried residue was redissolved in HCl and HBr, then passed through an AG 1-X8 anion-exchange resin column for purification, followed by a second AG 1-X8 column for further purification. The blank Pb levels, determined by isotope dilution, were minimal and did not influence the isotope ratios in the samples. The international standard NBS981 was used, yielding mass ratios of $^{206}Pb/^{204}Pb$ = 16.9343 ± 0.0003, $^{207}Pb/^{204}Pb$ = 15.4897 ± 0.0003, $^{208}Pb/^{206}Pb$ = 2.1682 ± 0.00002, and $^{207}Pb/^{206}Pb$ = 0.91468 ± 0.00001, which closely matched the reference values ($^{206}Pb/^{204}Pb$ = 16.9309, $^{207}Pb/^{204}Pb$ = 15.4850, $^{208}Pb/^{206}Pb$ = 2.16810, and $^{207}Pb/^{206}Pb$ = 0.91464) [16].

Results and discussion

Metal contents in sediment

As shown in Figure 2, the source of Na on the sediment is very high. The primary source of Na content (0.50-4.40 wt.%; 0.40-3.80 at.%) in the sediment is attributed to the surrounding marine environment, particularly the Taiwan Strait, which delivers substantial saline input. This influence is supported by research indicating that sediments in proximity to the Taiwan Strait show elevated levels of Na due to direct seawater influx and deposition of marine aerosols. The primary source of Mg content (0.70-1.50 wt.%; 0.60-1.20 at.%) in the sediment is linked to the Taiwan Strait, where seawater influx significantly contributes to the sediment's Mg content. Studies indicate that proximity to marine sources like the Taiwan Strait increases Mg concentrations in coastal sediments due to seawater deposition and the influence of marine aerosols. Main elements are presented in the Figures 2 to 9.

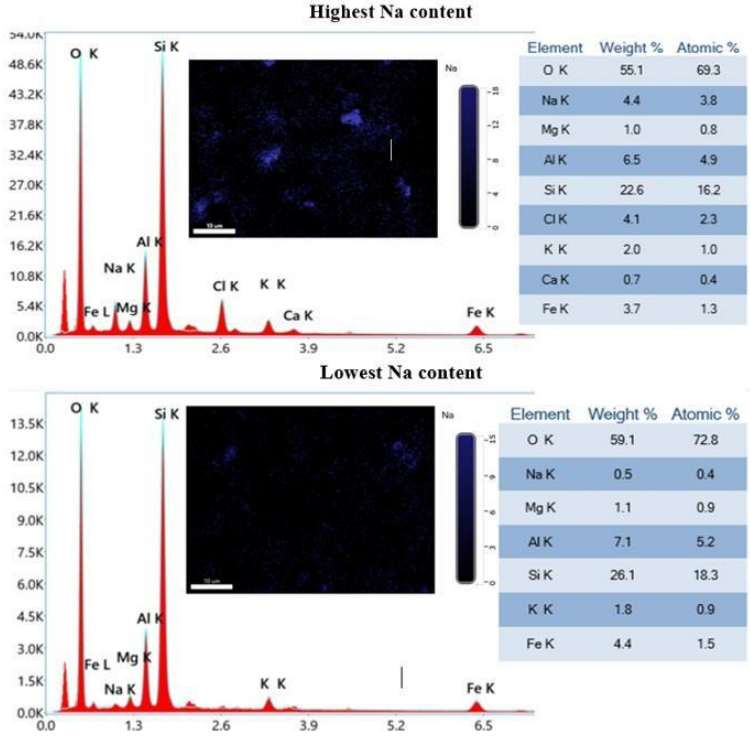


Fig. 2. The distribution Na contents in Taiwanese sediment

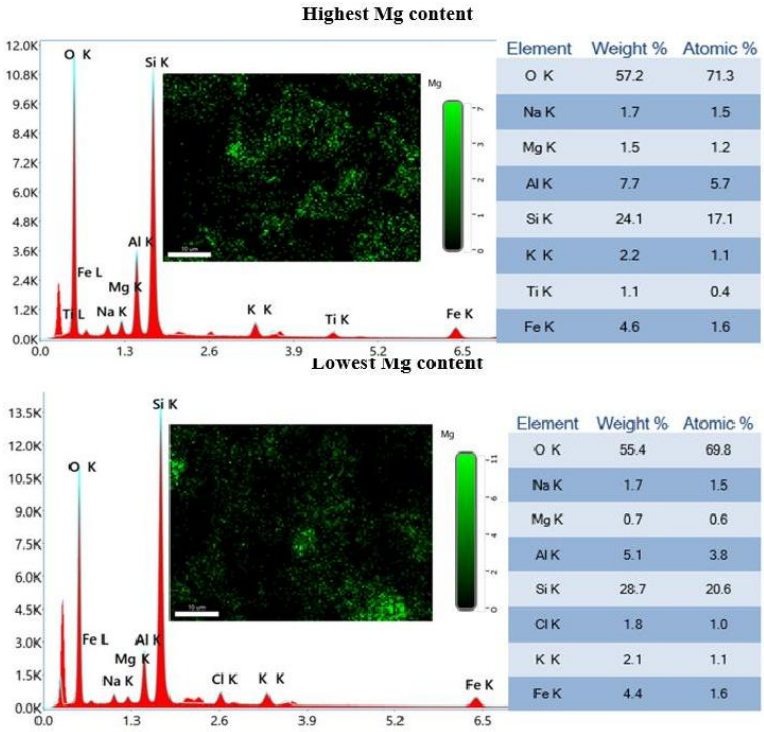


Fig. 3. The distribution Mg contents in Taiwanese sediment

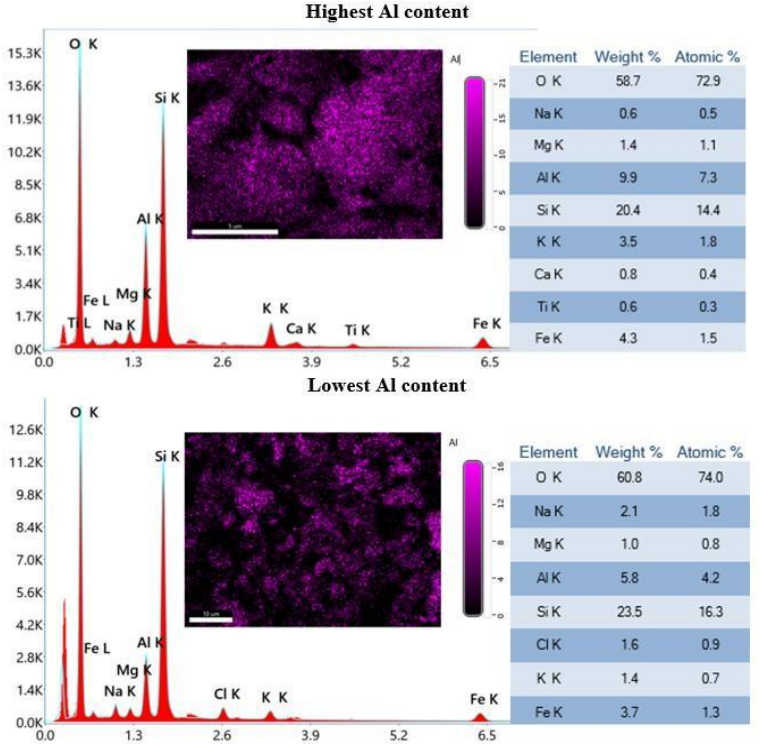
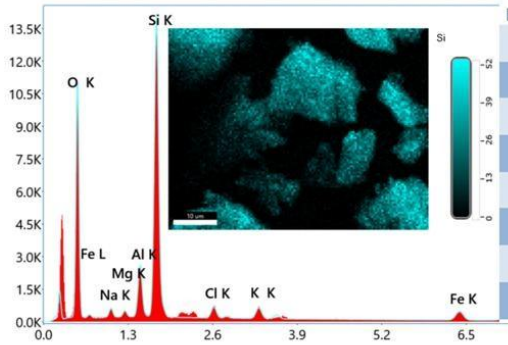


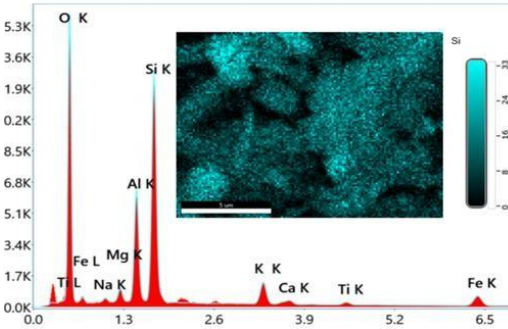
Fig. 4. The distribution Al contents in Taiwanese sediment

Highest Si content



Element	Weight %	Atomic %
O K	55.4	69.8
Na K	1.7	1.5
Mg K	0.7	0.6
Al K	5.1	3.8
Si K	28.7	20.6
Cl K	1.8	1.0
K K	2.1	1.1
Fe K	4.4	1.6

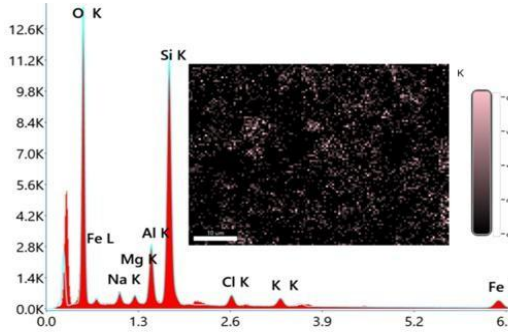
Lowest Si content



Element	Weight %	Atomic %
O K	58.7	72.9
Na K	0.6	0.5
Mg K	1.4	1.1
Al K	9.9	7.3
Si K	20.4	14.4
K K	3.5	1.8
Ca K	0.8	0.4
Ti K	0.6	0.3
Fe K	4.3	1.5

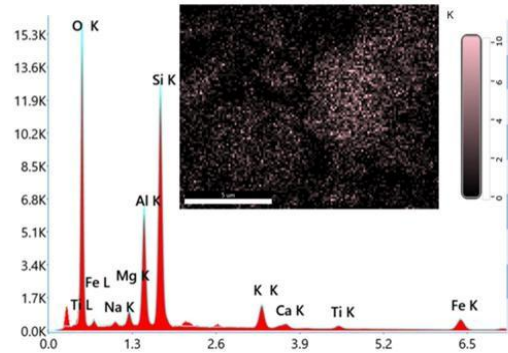
Fig. 5. The distribution Si contents in Taiwanese sediment

Highest K content



Element	Weight %	Atomic %
O K	60.8	74.0
Na K	2.1	1.8
Mg K	1.0	0.8
Al K	5.8	4.2
Si K	23.5	16.3
Cl K	1.6	0.9
K K	1.4	0.7
Fe K	3.7	1.3

Lowest K content



Element	Weight %	Atomic %
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Na K	0.6	0.5
Mg K	1.4	1.1
Al K	9.9	7.3
Si K	20.4	14.4
K K	3.5	1.8
Ca K	0.8	0.4
Ti K	0.6	0.3
Fe K	4.3	1.5

Fig. 6. The distribution K contents in Taiwanese sediment

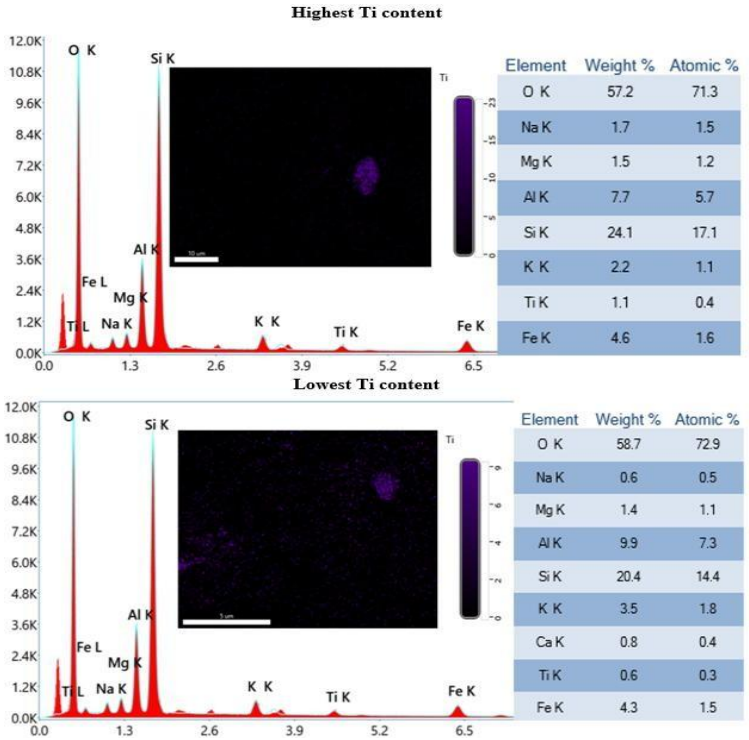

Fig. 7. The distribution Ti contents in Taiwanese sediment

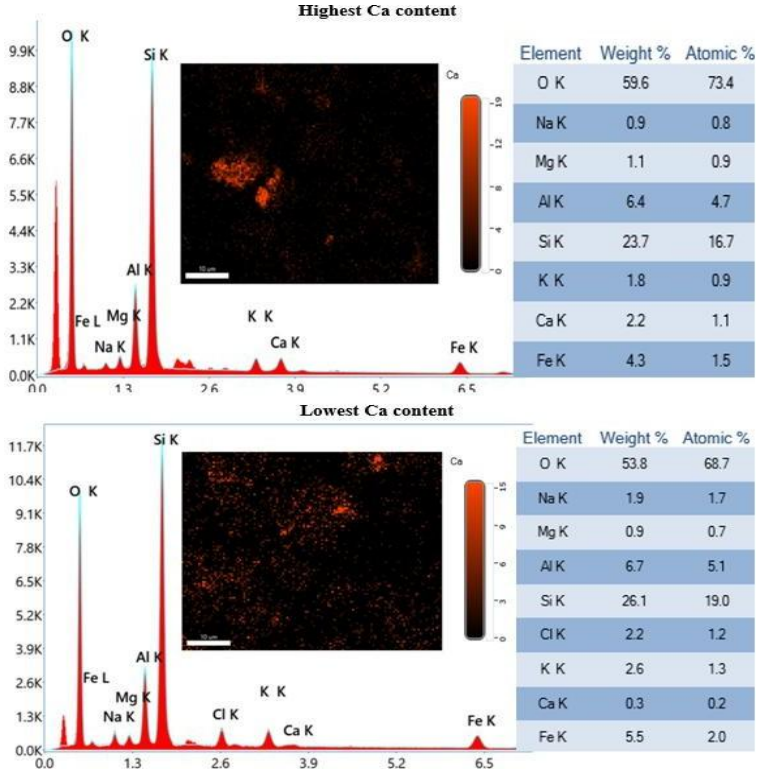

Fig. 8. The distribution Ca contents in Taiwanese sediment

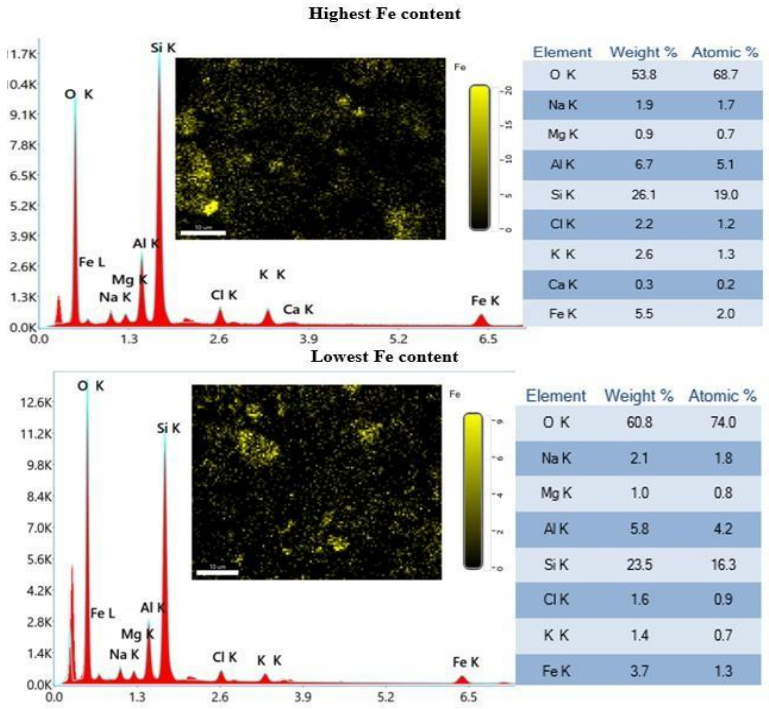

Fig. 9. The distribution Fe contents in Taiwanese sediment

Al content (5.80-9.90 wt.%; 4.20-7.30 at.%) in marine sediments along the coast of Taiwan primarily originates from terrestrial sources, with rivers serving as the main transport pathways. Taiwan's rivers, particularly those draining mountainous areas rich in aluminum-bearing minerals, carry substantial amounts of Al as suspended particles or dissolved ions into coastal and offshore regions. Weathering of aluminosilicate minerals, such as feldspar and kaolinite, is accelerated by Taiwan's humid, subtropical climate and frequent typhoons, resulting in high Al concentrations in riverine sediments. These sediments are subsequently deposited in coastal areas, contributing to the Al content in marine sediments. Studies have shown that the input from river discharge, particularly from larger rivers like the Danshui and Zhuoshui, is a major factor influencing Al levels along the northern and western coasts of Taiwan. Additionally, atmospheric deposition is an important secondary source of Al in Taiwan's marine sediments. Taiwan's location downwind from East Asia exposes it to airborne dust and particles, especially during seasonal monsoon events and dust storms from the Asian continent. This atmospheric input, enriched with Al from both natural dust sources and anthropogenic activities, settles into the marine environment and is incorporated into the sediments. Although the contribution from atmospheric deposition is generally smaller than riverine sources, it can still be significant, particularly in areas distant from direct river input. These combined terrestrial and atmospheric inputs of Al play a critical role in the geochemical composition of Taiwan's coastal and offshore sediments, reflecting both regional climatic conditions and land-based activities.

Si content (20.4-28.7 wt.%; 14.4-20.6 at.%) in marine sediments along the coast of Taoyuan, Taiwan, primarily originates from terrestrial sources, specifically from riverine and land-based erosion processes. The rivers draining through Taoyuan, which pass through regions with silicate-rich bedrock, transport considerable amounts of silicon to the coast in the form of

suspended particles, such as quartz and other silicate minerals. This silicon-rich sediment is deposited along the coastline, creating a high Si concentration in nearshore marine sediments. Taoyuan's seasonal rainfall, combined with intense weather events like typhoons, accelerates the erosion of silicate rocks, enhancing the riverine transport of Si to the marine environment. Consequently, the sediment composition near Taoyuan reflects significant terrestrial influences, with high levels of silicon-rich minerals derived from the regional geology and climatic patterns. In addition to riverine input, silicon in Taoyuan's marine sediment also includes contributions from biogenic sources, such as diatoms and other siliceous marine organisms. As these organisms grow, they absorb dissolved silicon from seawater to build their siliceous structures, which, upon death, settle to the seafloor and become part of the sediment matrix. This biogenic silicon input is particularly significant in productive coastal waters influenced by nutrient upwelling or human-induced nutrient input from nearby urban areas. Thus, the silicon content in Taoyuan's marine sediments reflects a complex interaction of terrestrial inputs from silicate-rich erosion and in situ contributions from biogenic silicon sources, both of which contribute to the region's distinctive sedimentary composition. Cl content in marine sediments along the coast of Taoyuan, Taiwan, primarily originates from seawater, where Cl is a major dissolved ion. The proximity of Taoyuan to the Taiwan Strait allows for continuous exposure to saline water, leading to the deposition of Cl through processes such as seawater infiltration, wave action, and tidal mixing. As seawater interacts with coastal sediments, Cl ions are deposited and incorporated into the sedimentary matrix. The high concentrations of Cl in these coastal sediments thus directly reflect the influence of the marine environment, where salinity levels are a primary determinant of Cl content. This process is particularly pronounced in nearshore areas and tidal zones, where frequent inundation ensures regular deposition of marine-derived Cl.

Additionally, atmospheric deposition plays a significant role in Cl accumulation in Taoyuan's marine sediments. The coastal location exposes Taoyuan to sea spray and marine aerosols, especially during monsoon seasons and typhoon events, which transport fine droplets of seawater containing Cl over land and coastal waters. These aerosols eventually settle into marine and coastal sediments, adding to the Cl levels from direct seawater interactions. This atmospheric input of Cl, while secondary compared to direct seawater deposition, can be substantial during storm events, which enhance the inland reach of marine aerosols. Together, these sources of marine water interaction and atmospheric deposition constitute the primary pathways for Cl in Taoyuan's marine sediments, reflecting both direct and indirect influences of the adjacent marine environment.

K content (1.40-3.50 wt.%; 0.70-1.80 at.%) in marine sediments along the coast of Taoyuan, Taiwan, is largely derived from terrestrial sources, particularly through riverine input and the erosion of potassium-bearing minerals from the surrounding landscape. Rivers draining the region carry sediments rich in feldspar, mica, and clay minerals, which are significant reservoirs of K. As these minerals weather and break down, they release K ions into the river waters, which subsequently transport them to the coastal sediments of Taoyuan. This process is intensified by Taiwan's subtropical climate and frequent typhoons, which accelerate erosion and sediment transport. Consequently, the high concentrations of K in Taoyuan's marine sediments reflect the geological makeup of the nearby land, dominated by silicate minerals that are naturally rich in potassium. In addition to terrestrial inputs, marine sources also contribute to the K content in Taoyuan's coastal sediments, though to a lesser extent. Seawater contains dissolved K ions, which can be deposited in sediments through tidal action, wave interaction, and seawater infiltration into the coastal substrate. While the concentration of K in seawater is relatively lower compared to other ions, such as sodium and chloride, continuous exposure to the Taiwan Strait

allows for a steady, though modest, deposition of K. This marine input, combined with the substantial terrestrial contributions, results in a distinct geochemical signature in Taoyuan's coastal sediments, illustrating a complex interplay between land-derived minerals and oceanic influences in shaping the sedimentary composition of this region.

Ca content (0.30-2.20 wt.%; 0.20-1.10 at.%) in the marine sediments along the coast of Taoyuan, Taiwan, originates primarily from terrestrial sources, especially from riverine transport of weathered carbonate minerals from nearby upland regions. Rivers draining the Taoyuan area carry sedimentary particles rich in calcite, aragonite, and other Ca-bearing minerals derived from the local geology, which includes limestone and other calcareous deposits. As these minerals undergo chemical and physical weathering, Ca is released and transported downstream, where it eventually settles in coastal marine sediments. The intensity of weathering and erosion in Taiwan's subtropical climate, coupled with seasonal typhoons, facilitates this influx of Ca-rich materials to Taoyuan's coast, creating sediment deposits with substantial terrestrial-derived calcium content. Additionally, biogenic sources from marine organisms contribute to the Ca levels in Taoyuan's coastal sediments. Marine organisms, such as shellfish, corals, and certain plankton, utilize calcium carbonate to form their shells and exoskeletons. Upon their death, these calcareous structures settle and accumulate on the seafloor, adding biologically sourced calcium to the sediment matrix. This biogenic calcium is particularly pronounced in productive coastal waters, where nutrient influxes from nearby urban and agricultural areas can stimulate biological activity. Therefore, the calcium content in Taoyuan's marine sediments reflects a dynamic interplay between land-based mineral sources and marine biogenic inputs, offering insights into both geological and ecological processes impacting the region's coastal environment.

Fe content (3.70-5.50 wt.%; 1.30-2.00 at.%) in marine sediments along the Taoyuan coast of Taiwan primarily originates from terrestrial sources, particularly through riverine transport of iron-rich minerals eroded from the island's mountainous regions. Taiwan's rivers, which flow through iron-bearing rock formations, transport significant quantities of Fe in the form of suspended particles, such as hematite, magnetite, and other iron oxides, directly into the coastal zones. The subtropical climate, marked by frequent heavy rainfall and typhoons, accelerates the weathering of these minerals and enhances their transport to coastal environments. These iron-rich sediments, originating from Taiwan's rugged geology, become deposited in marine sediments near Taoyuan, where they contribute to the high Fe concentrations characteristic of the region's coastal sedimentary makeup.

In addition to riverine input, atmospheric deposition also contributes to the Fe content in Taoyuan's marine sediments. Iron-rich dust, carried from mainland Asia during seasonal monsoons and dust storms, settles over the Taiwan Strait and nearby coastal regions, adding a significant external source of Fe to the sediment. This atmospheric iron deposition is especially pronounced during the northeast monsoon season when dust-laden winds from China reach Taiwan's coastal zones. Together, these sources, terrestrial riverine input and atmospheric deposition, create a complex Fe enrichment in Taoyuan's marine sediments, reflecting the combined impact of local geology and transboundary atmospheric processes on the region's sedimentary composition. This finding is related by the researchers to the reported distribution of Fe, and to the distinction between anthropogenic and natural sources of Fe [17], [18]. All measured contents of Na, Mg, Al, Si, K, Ti, Ca, and Fe in South China Sea sediments fall within the natural background range for marine sediments and show no evidence of anthropogenic enrichment. International guidelines such as those from WHO do not provide threshold values for these major elements in sediments. Therefore, these data can serve as a reference for background, non-anthropogenic metal contents in the region.

Pb concentration and isotope analysis

Figure 10 presents Pb isotope data for all seawater samples. Total Pb concentrations were measured and plotted against individual Pb isotope ratios, specifically ^{207}Pb and ^{208}Pb. The modeled total Pb content in seawater ranges from 0.0005 to 0.0020 wt.%, with one outlier at 0.0035 wt.% (Figure 10). A moderate negative correlation is observed between total Pb content and Pb isotope ratios, described by the equation: y = -17.785x + 0.2057 with R^2 = 0.5469. The pattern negative similar trend among the isotope ratios suggests that the Pb in surface seawater originates as Pb isotope ratios from the natural sources such as upper crust (^{207}Pb/^{204}Pb = 15.6-15.7); basalt rock/mantle (^{207}Pb/^{204}Pb =15.45-15.55); Saharan dust (^{207}Pb/^{204}Pb = 15.66-15.690; sea aerosol (^{207}Pb/^{204}Pb = 15.55-15.60), and (^{207}Pb/^{204}Pb =15.55-15.68). Together, these mixed sources constitute a global Pb signal in seawater influencing both Taiwan and Indonesia, with radiogenic Pb produced by decay of ^{235}U and ^{237}Th in parent rocks. Natural U and ^{204}Pb is primordial and non-radiogenic, while ^{207}Pb and ^{208}Pb indicate a minor radiogenic contribution. We propose that seawater Pb predominantly results from natural weathering, sediment leaching, and aerosol deposition.

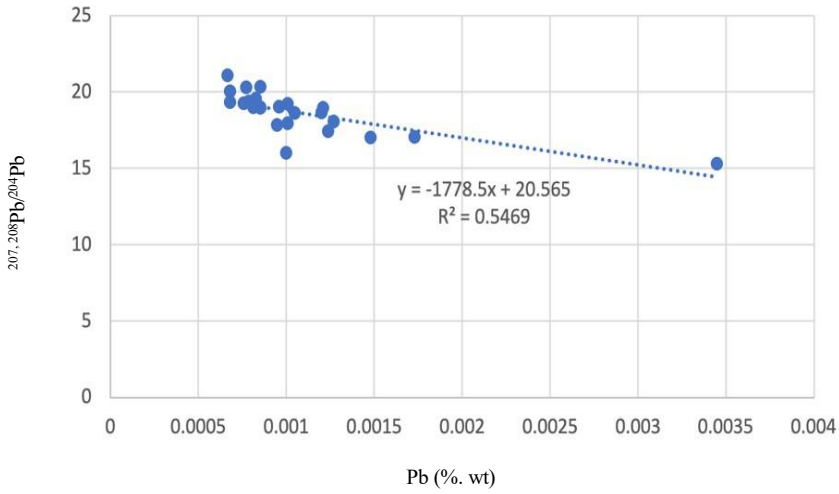

Fig. 10. Total Pb concentration (%. wt) vs $^{207;\,208}$Pb/^{204}Pb

Figure 11 presents the ^{208}Pb/^{204}Pb and ^{207}Pb/^{204}Pb growth curve, which traces the evolution of ore Pb through time. The left end represents older ore Pb, and the right end represents younger, modern Pb. By incorporating time-dependent decay, this curve allows for the identification of Pb ore sources and provides age information for Pb-bearing materials. It is generated by plotting isotope ratios along the global Pb ore growth line. The model is applied to seawater using measured Pb isotope ratios to assess anthropogenic Pb contributions.

As shown in Figure 11, it shows the evolution of Pb isotope ratios over geological time, driven by the decay of unstable U and Th to stable Pb isotopes. Specifically, ^{235}U decays to ^{207}Pb with a half-life of 703.8 million years, and ^{232}Th decays to ^{208}Pb with a half-life of 14.010 billion years. ^{204}Pb is primordial and its abundance remains constant over geological time. The Pb isotope growth curve therefore provides constraints on the age of Pb-bearing materials. This curve is generated by plotting isotope ratios against a global reference line defined by known Pb ore sources. To apply this growth curve model to seawater in the Gulf of Prigi, we correlated

measured Pb isotope ratios with reference data from global Pb ore deposits. Previously published age data for each source were integrated, yielding a correlation coefficient of $R^2 = 0.99$.

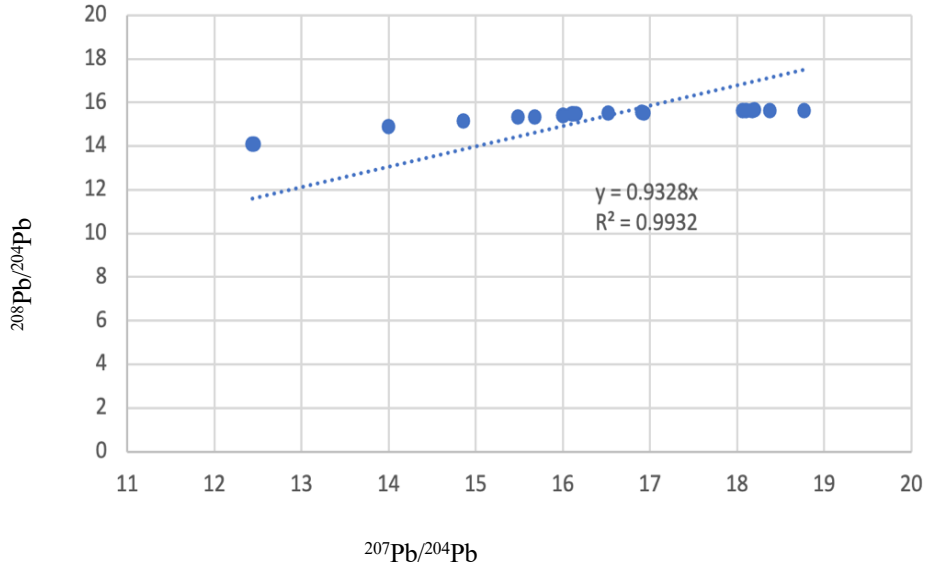

Fig. 11. Correlation of $^{207}Pb/^{204}Pb$ vs $^{208}Pb/^{204}Pb$ associated with ore age

Figures 10 and 11 were used to distinguish between anthropogenic and natural sources of Pb by measuring and calculating Pb isotope ratios in seawater. As shown in Figure 10, the data are divided into 5 groups: 3 clusters and 4 individual sources. One source plot far from the other sources. Based on the growth curve, most source groups appear to have Pb derived from old ore. One group of seawater samples has Pb isotope ratios ranging from 17.0 to 22.0 with Pb contents of 0.00055–0.0015 wt.%. Another group Pb isotope ranges from 17.0 to 20.0 with Pb contents of 0.00125 wt.%. The four individual sources, from left to right, have values of Pb isotope ratios ~0.15 (0.0010 wt.%), 0.16 (0.0015 wt.%), 0.16 (0.00175 wt.%), and 0.15 (0.0035 wt.%). The Pb isotope ratios of the seawater samples reveal distinct patterns and variations among the 5 groups. Notably, these samples deviate from the typical Pb growth curve. This suggests that Pb in unpolluted seawater may originate from natural sources, such as freshwater input and mineral weathering within local sediments.

A s shown in Figure 10, the seawater Pb isotope ratios are relatively low and indicate an old source age. For comparison, typical anthropogenic end-members include: leaded gasoline $^{207}Pb/^{204}Pb = 16.10–16.40$; unleaded gasoline $^{207}Pb/^{204}Pb = 16.00–16.20$; smelter/metal smelting $^{07}Pb/^{204}Pb = 16.30–18.50$; coal-fired power plant $^{07}Pb/^{204}Pb = 18.50–19.20$; waste incinerator = $^{07}Pb/^{204}Pb = 16.50–17.50$; and paint/industrial sources $^{07}Pb/^{204}Pb = 16.20–16.80$. Anthropogenic inputs are generally characterized by relatively high Pb isotope ratios, particularly $^{207}Pb/^{204}Pb >$ 16.0. This reflects the use of younger-age Pb ores in industrial processes and urban emissions. We suggest that there is no significant contamination from anthropogenic sources. The elevated Pb and other metal concentrations are more likely due to the natural abundance of metals in oxide phases mixed with silica and alumina, which are leached from local sediments into seawater.

In addition, the Pb isotope values in seawater are consistent with mixing via natural processes. The South China Sea Current and the Kuroshio Current can transport water masses from Indonesia and Malaysia toward Taiwan. Upper crust and basaltic sources are characterized by $^{207}Pb/^{204}Pb$ ~15.23 – 16.08. In contrast, Indonesia members include seawater Pb isotope ratios ~1.158, and Singapore aerosol with Pb isotope ratios ~1.147. Therefore, if seawater around Taiwan exhibits Pb isotope ratios ~1.15–1.17, it strongly indicates mixing of Pb from Malaysia, Indonesia and China sources [19].

Conclusions

All measured contents of Na, Mg, Al, Si, K, Ti, Ca, and Fe in South China Sea sediments fall within the natural background range. These values can serve as a reference for marine sediments in the region. Furthermore, the sediments can be used to reconstruct the historical record of seawater chemical composition. The relationship between total Pb content and isotope ratios suggests that the China Sea water predominantly receives Pb from natural sources associated with parent rocks, such as U and Th. Analysis of $^{208}Pb/^{204}Pb$ and $^{207}Pb/^{204}Pb$ ratios conforms to the natural Pb growth curve, confirming that Pb in this region is mainly derived from natural leaching of marine sediments rather than pollution sources. The use of Pb isotopic analysis in this region's seawater is effective for environmental monitoring.

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