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Microplastics leaving a trace in mangrove sediments ever since they were first manufactured: A study from Indonesia mangroves

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ABSTRACT

Mangrove environments have been well recognized as marine litter traps. However, it is unclear whether mangrove sediments sink microplastics more effectively than other marine sediments due to active sedimentation. Furthermore, microplastics archives in mangrove sediments may provide quantitative data on the impact of human activities on environmental pollution throughout history. Microplastic abundance varied markedly between high and low anthropogenic activities. Both mangrove and adjacent mudflats sediments act as microplastic sequesters, despite having similar microplastic abundances and depth profiles. The decreasing trend of microplastics was observed until the sediment layers dated to the first-time plastic was manufactured in Indonesia, in the early 1950s, but microplastics remained present beneath those layers, indicating the downward movements. This discovery highlighted the significance of mangrove sediments as microplastic sinks. More research is needed to understand the mechanisms of microplastic deposition in sediments, as well as their fate and potential impact on mangrove sediment dwellers.

1. Introduction

The suggested era of Anthropocene defines the geological period during which escalated human activities have brought about significant alterations in Earth-system processes (Lewis and Maslin, 2015). This particular time frame has been marked by the production of novel substances that hold vital importance for our societies (Elhacham et al., 2020). Plastic, among these materials, stands out due to its singular attributes and highly adaptable material, which has resulted in its volume surpassing that of living creatures on Earth and potentially leaving lasting traces in sedimentary archives (Andrady, 2015; De-la-Torre et al., 2021). The growing incidence of plastic pollution in marine environments raises apprehension regarding its detrimental impact on ecosystem dynamics, encompassing modifications to the oceanic carbon cycle and toxicological injury to organisms (Huang et al., 2022b; Miller et al., 2021). Regrettably, current scientific research has accumulated significant evidence concerning the presence of small-sized plastic debris, i.e., microplastic, in marine ecosystems, particularly in tropical environments such as mangroves, seagrass beds, and coral reefs. Whereas, these fragile ecosystems serve as major reservoirs for the accumulation of microplastic pollutants (Chen, 2022; Li et al., 2022; Martin et al., 2020). The primary mechanism by which these contaminants enter these valuable habitats is through their buoyancy (Yuan et al., 2023); floating particles are expected to remain at sea until they

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eventually reach these locations (Cordova et al., 2021b). Nonetheless, it is worth noting that mechanisms other than buoyancy also contribute to the transport of microplastics into the water column and subsequently via ocean currents (Iskandar et al., 2021, 2022). Moreover, various physical and chemical transformations resulting from processes like weathering, fragmentation, biological interactivity, aggregation with other substances, and biofilm formation may promote further deposition of microplastic particles into sediment layers (Chubarenko et al., 2018; Van Sebille et al., 2020).

Microplastic particles that are deposited in sediment have the potential to interact with organisms found at the bottom of bodies of water (Saley et al., 2019; Zhang, 2017). These pollutants can serve as indicators of time within sedimentary records and be used as markers for determining the chronological sequence of such deposits (Bancone et al., 2020). Although microplastics could potentially function effectively as temporal tracers, their longevity and breakdown processes in sediments remain largely un-investigated. Depositional environments like river prodeltas provide an opportunity for detailed analysis due to their ability to offer high-resolution stratigraphy spanning several recent decades, which makes them ideal locations for monitoring the patterns and changes associated with microplastic pollution over time.

In recent years, researchers have found evidence of microplastic contamination in various marine and freshwater sediments from different regions worldwide. For instance, studies conducted in areas such as the urban lake in London (Turner et al., 2019), Southern Ocean regions (Cunningham et al., 2020), Tokyo Bay (Matsuguma et al., 2017), the Gulf of Thailand (Chaisanguansuk et al., 2023; Matsuguma et al., 2017), the Straits of Johor in Malaysia (Matsuguma et al., 2017), Durban Bay in South Africa (Matsuguma et al., 2017), Wuliangsuhai Lake (Mao et al., 2021) and Taihu Lake (Huang et al., 2022a) in China have revealed the presence of microplastics at different depths within these sediments. These findings indicate that microplastics can accumulate and persist in deep sediment layers to a greater extent than observed near the surface or in overlying waters alone. As a consequence of this discovery, it becomes essential to assess the amount and distribution pattern of microplastics throughout vertical profiles within sediment samples. By analyzing these profiles comprehensively, a more accurate estimation can be obtained regarding total quantities present as compared to conclusions based solely on surface sediment analysis.

Indonesia possesses a significant share of the world's mangrove ecosystems, accounting for approximately 20–25 % of these ecologically important habitats (Rahadian et al., 2019; Sidik et al., 2018). The total area covered by Indonesian mangroves is estimated to be around 3,364,076 ha (Kementerian Lingkungan Hidup dan Kehutanan Republik Indonesia, 2021). Unfortunately, anthropogenic factors pose substantial threats to this precious ecosystem; specifically, plastic waste emerges as a major concern. The inadequate management and collection of plastics in Indonesia make it particularly susceptible to the accumulation of microplastics within its vast expanse of mangrove forests (Cordova et al., 2021b; Richards and Friess, 2016). The origin of this plastic debris is primarily attributed to human activities in the vicinity of the mangrove forest, mainly from domestic sources (Cordova et al., 2022a; Iskandar et al., 2021, 2022). It is crucial that authorities recognize the significance of addressing this issue promptly and effectively, given that coastal fisheries serve as vital sources of sustenance and economic stability within the country (Phillips et al., 2015). Consequently, controlling plastic pollution in these valuable wetlands should assume paramount importance for government institutions and inhabitants. Nevertheless, there is a paucity of research conducted on the presence and characterization of microplastics in mangrove sediment. In particular, there is limited information regarding the specific types of polymers found in these sediments as well as quantitative assessments of mangroves' role as sinks for microplastics. Furthermore, studies investigating microplastic distribution within stratified sediment cores are particularly scarce. Such investigations are crucial since they allow us to gain valuable insights into historical trends related to contamination levels over time.

In order to enhance our comprehension of microplastic contamination within mangrove ecosystems, this investigation examined the vertical distribution of microplastics within sediment profiles at four different mangrove sites. These sites encompassed two localities with varying magnitudes of human-induced activity - two areas with high levels (Jakarta and Surabaya) and two areas (Cilacap and Berau) with low levels. This study presents the outcomes derived from assessing polymer types found in sediment cores collected from Indonesian mangroves and mudflat habitats. It offers a detailed quantitative assessment concerning how sediments function as sinks for microplastics within these regions. The significance lies in the fact that measuring microplastics within marine environments is a relatively nascent field, leading to limited availability when it comes to time-series data. Consequently, the objective of this study was to examine the effects of anthropogenic activities and sampling location (specifically in mangrove and mudflat areas) on the presence of microplastic contamination.

2. Methodology

2.1. Study area

At the beginning of 2023, a survey was conducted in Jakarta and Surabaya - areas with high levels of anthropogenic activity - as well as Cilacap and Berau, where such activities were relatively low (Fig. 1). Jakarta and Surabaya are prominent urban centers within the nation of Indonesia, whereas Cilacap and Berau are administrative divisions located in the regions of Java and Kalimantan, respectively. The remnant patches in Jakarta and Surabaya, characterized by tidal ranges of <2 m and silty sediment, are exclusively inhabited by *Avicennia marina* and *Rhizophora mucronata* (Fitriana et al., 2022; Latief et al., 2018). The mangrove habitats in the two major cities were originally mangrove coverage and then converted into fishponds that were later abandoned, transformed, and recolonized by mangroves (Mayalanda et al., 2014; Prasita et al., 2019). In contrast, the mangroves chosen for this study were located in the Segara Anakan Lagoon (SAL), Java's largest remaining mangrove forest. These mangroves have experienced relatively minimal disturbances and are situated adjacent to the capital city of Cilacap. This particular geographical area exhibits a greater diversity of mangrove species compared to the cities of Jakarta and Surabaya. Noteworthy species found in this location include *Rhizophora apiculata*, *Bruguiera gymnorrhiza*, *Xylocarpus granatum*, *Xylocarpus moluccensis*, *Aegiceras corniculatum*, *Ceriops decandra*, and *Ceriops tagal*. According to Khadami et al. (2020), the amplitude of the SAL tide during the spring tide is approximately 2 m, whereas it is approximately 1 m during the neap tide. The study included Delta Berau in Kalimantan, situated east of Tanjung Redeb, the capital city of Berau. This area was chosen due to its undisturbed mangroves. The observed species at the sampling sites were limited to *Sonneratia alba* and *Avicennia alba*. According to Kusumaningtyas et al. (2019), the tidal range of the delta can reach a maximum of 3 m.

2.2. Sampling methods

In this research, we collect two types of sediment samples to investigate microplastics' prevalence in mangrove sediment and analyze their deposition rates. To facilitate sediment dating, a total of eight cores were acquired, with each sampling region yielding one core from both mangrove and mudflat areas. In order to analyze microplastics, a comprehensive set of twenty-four sediment cores was obtained, with three cores being collected from each of the four sampling regions, specifically the mangrove and mudflat areas. Stainless steel tubes (with a length of 120 cm and an inner diameter of 5.65 cm) were employed to gather sediment cores. The identified core exhibiting stable patterns in sediment accumulation rate over time was selected for further

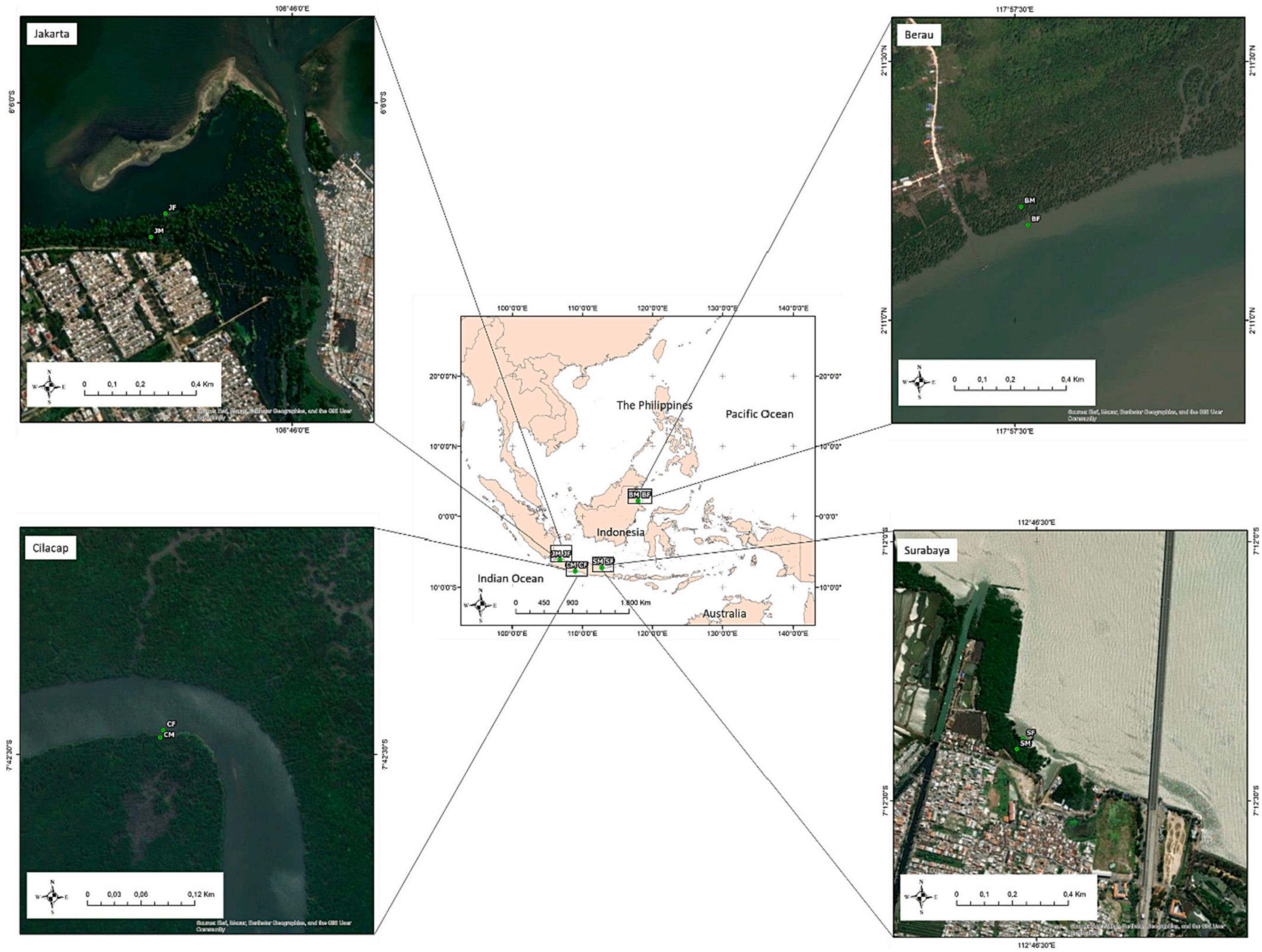

Fig. 1. Study locations with contrasting levels of anthropogenic activity. Jakarta and Surabaya represent the high level, while Cilacap and Berau represent the low level. The sampling sites in each location were in mangrove habitats and mudflats separated by varying distances of 9 m (Cilacap), 42 m (Surabaya), 70 m (Berau), and 94 m (Jakarta).

examination after age-profiling all cores via established protocols. The length of the central part of each area differs. Samples collected from Jakarta are within the range of 60–88 cm, while those taken from Surabaya fall between 72 and 80 cm. The sediment cores obtained from Cilacap share a common extent measuring precisely 80 cm, whereas samples extracted in Berau oscillate approximately between 80 and 88 cm in length. The samples were partitioned into 2 cm segments (for sediment dating) and 8 cm for microplastics analysis (triplicate) and subsequently placed in a sterile plastic ziplock bags (for sediment dating) and sterile aluminum foil bag (for microplastics analysis), followed by preservation at a temperature of 4 °C. Given the limited facilities available for this research, we made adjustments to optimize our analysis procedure for segmenting microplastics from sediment cores. Although it would have been ideal to carry out the segmentation at smaller (1–2 cm) intervals, we conducted representative replications and used the resources that were accessible to us.

2.3. Sediment dating determination

The sediment ages were determined by ^{210}Pb, which is an environmental isotope; a radionuclide (via decay of ^{238}U and ^{222}Rn) has a half-life of 22.3 years. It is commonly used to estimate the date of sediment layers in aquatic environments over a timespan of from ~100 to 150 years with a precision of a few years (Barsanti et al., 2020). The applicability range of ^{210}Pb method is coincidence with the time during which appreciable environmental changes occurred due to industrialization (Krishnaswamy et al., 1971). Analyses of total ^{210}Pb (^{210}Pb$_{tot}$) were adopted in this study, following Lubis (2006) and Sanchez-Cabeza et al. (1999). Briefly, 0.3 g of dried homogenized sediments were digested in closed vessels and high-pressure using microwave. Prior to the digestion, dried sediments dissolved with HNO_3, HCl, H_2O_2, and aqua bidest. Internal standard ^{209}Po was spiked to each sample for quantitative determination of ^{210}Pb. After digestion, the samples were dissolved in 0.3 N HCl which then heated and added ascorbic acid to complexify any iron present. ^{209}Po and ^{210}Pb (assumed in equilibrium with ^{210}Po) were spontaneously subsequently deposited onto copper disks while stirring for about 4 h. Both isotopes were counted using alpha spectrometers equipped with PIPS (Passivated Implanted Planar Silicon) detectors (Canberra A450-20AM). Supported ^{210}Pb (^{210}Pb$_{sup}$) was obtained from the constant value observed in the core bottom measured by alpha spectrometry (de Carvalho Gomes et al., 2011; Sanchez-Cabeza et al., 1999). In addition, ^{210}Pb$_{sup}$ (in equilibrium with ^{226}Ra in sediments) was also measured from the top, middle and bottom of each core using gamma spectrometer. The gamma spectrometer was equipped with a high purity germanium detector, which has 30 % efficiency and the connected to the GENIE 2000 spectrum master and Multi-Channel Analyzer. Prior to the measurement by gamma spectrometry, the sediment samples were sealed for 4 weeks to achieve equilibrium with ^{210}Pb. The excess ^{210}Pb ($^{210}Pb_{ex}$) was calculated by subtracting ^{226}Ra from the $^{210}Pb_{tot}$. A downcore profile of $^{210}Pb_{ex}$ was employed to estimate the chronological age of sediment deposition at varying depths.

2.4. Microplastic extraction, identification and characterization procedure

The laboratory technique used to evaluate microplastics in the core sediments was adopted from prior research (Abayomi et al., 2017; Curren et al., 2021). In brief, sediment samples (49–51 g) were dried in an oven at 40 °C for 72–96 h to minimize the moisture contents without deforming plastic material and causing minimal structural damage to the polymer composition. The sediment was then treated with a $ZnCl_2$ solution (ρ = 1.5 g/ml, Merck Millipore EMSURE®, ACS, ISO, Reag. Ph Eur), stirred, and left overnight. The supernatant solutions were then transferred to a test tube (Pyrex, 100 ml) and dried at 40 °C for 24 h. Fenton's reagent prepared from Fe(*II*)solution (20 ml Fe(*II*)SO4, 10 mg/ml Merck Millipore, EMSURE® ACS, ISO, Reag. Ph Eur) and H_2O_2 (20 ml, 30 % Merck Millipore, Emprove® Essential, Ph Eur, BP, USP) was applied to dried samples in a test tube. The test tube was incubated in a water bath (Shibata water bath WB-6C) at 40 °C for 24–48 h. Afterward, the samples were then transferred to sterile filter paper (Merck Whatman™ cellulose nitrate filter paper 47 mm, pore size 0.45 m, grid) and covered with a sterile petri dish at room temperature prior to form, size, and chemical composition analysis. The filter paper membrane was observed using a microscope (Oympus CX31) and a camera (Sony IMX307).

We detected possible microplastics were detected using established methods for identification (Cordova et al., 2019; Dehaut et al., 2019; Piehl et al., 2018). The identified particles were characterized by their consistent coloration, the absence of organic or cellular characteristics, and an undivided structure (Cole et al., 2013; Cordova et al., 2022a; Peng et al., 2017). The shape and size measurements of these determined particles were promptly recorded. This study set a detection limit at 200 μm which was marginally below the size range boundary (212 μm) portrayed in Lao and Wong's (2023) investigation conducted among different environmental matrices. The smallest microplastic observed through microscopy, verified with Attenuated Total Reflectance - Fourier Transform Infrared Spectrometer (ATR-FTIR) procedure, measures 200.1 μm, while the largest measured at 4494.9 μm. From the samples, a representative of the identified microplastic particles was selected (80 %, 240 out of 300 particles), and its chemical composition was evaluated using an ATR-FTIR (diamond crystal material, Agilent Cary 630 with Microlab Expert Software). The 240 particles used for the chemical composition analysis were collected using a method of systematic random sampling that represented each sampling area. The FTIR was configured to operate at a resolving power of 4 cm^{-1}, encompassing the spectrum range from 650 to 3000 cm^{-1} with 32 scans. The scrutiny for polymers centered on inspecting specific band regions to identify features representative of CH_2 bending vibration level, CF_2 stretching vibration level, C═O stretching vibrations and that characteristic of CH/CH_2/CH_3 groups' stretching vibrations (Jung et al., 2018; Piehl et al., 2018; Wagner et al., 2017). Polymers were detected by analyzing unique signatures manifested in various frequencies within these bands' ranges combined with a standard and advanced library provided by FTIR.

2.5. Quality control and quality assurance

In order to ensure the precision of sediment dating, we addressed the issue of uncertainty estimation in sediment age by employing several techniques. These techniques included using certified references, selecting undisturbed sediments, slicing sediments at a thickness of 2 cm, and correcting mass depth as Barsanti et al. (2020) recommended. The accuracy of ^{210}Pb measurements was assessed by comparing the radioisotopes ^{210}Pb and ^{226}Ra activities in standard reference materials IAEA-447, provided by the International Atomic Energy Agency (IAEA). In this study, the measured activity concentrations for both radionuclides closely aligned with the certified values, exhibiting <10 % deviations. Ideally, ^{137}Cs should be measured alongside ^{210}Pb measurements as a validation for sediment dating (Barsanti et al., 2020), but we do not have access to that facility in our study, and ^{137}Cs is difficult to detect in the Southern Hemisphere, as it is in the majority of study locations. To ensure the integrity of our microplastic samples and prevent any potential contamination, rigorous precautions were taken prior to sampling. The spatula used was washed meticulously with filtered water three times using sterile filter paper. Similarly, all glassware was rinsed thoroughly three times with filtered water and then covered in aluminum foil. Moreover, we followed strict protocols for personal hygiene by wearing 100 % cotton clothes while handling the samples, further minimizing the risk of foreign particulates entering our samples. Additional measures were implemented during sample treatment processes to maintain a controlled environment in the laboratory setting. After each treatment step, samples were promptly wrapped to minimize contact with external elements. Furthermore, all instruments involved were diligently sanitized and cleaned before commencing any laboratory studies. Moreover, to eliminate even trace amounts of microparticles that might impact experimental results or introduce bias into our analysis, chemical solutions underwent filtration using sterile filter paper.

In order to evaluate the effectiveness of the microplastic extraction treatment, a recovery test was conducted (Cordova et al., 2022b; Nurhasanah et al., 2021; Weber and Kerpen, 2022). This involved creating a homogeneous mixture consisting of seven different synthetic polymers, with a size range of 400–1000 μm, commonly utilized globally (PlasticsEurope and EPRO, 2021). The polymers (i.e., polypropylene, high-density polyethylene, low-density polyethylene, polystyrene, polyvinyl chloride, polyamide, and polyurethane) were thoroughly blended with pure water sourced from Milli-Q® and 6 mg/l Now Solution® Red Clay Powder. This concentration was chosen as it mirrors the average levels of total suspended solid content found in Jakarta Bay (Koagouw et al., 2021). The material blend underwent identical protocols as the original sample, which involved separation based on density using a solution of ZnCl2 and digestion facilitated by Fenton's reagent. Repeating the density separation procedure three times and implementing one round of biological digestion found that all polymers were recovered at a 100 % rate. The outcomes of the investigation on the chemical composition using FTIR were juxtaposed against Agilent, Thermo-scientific, and Shimadzu's standard polymer catalogs. In addition to that, evaluations were also conducted by referring to FLOPP Spectral Libraries for Plastic Particles (De Frond et al., 2021) as well as a plastic benchmark acknowledged by the Research Center for Geosciences, University of Bayreuth, Germany. A blank procedural measure was devised to quantify the level of contamination introduced during these processes and mitigate potential errors arising from sampling and laboratory procedures. This procedure involved collecting blank samples during field sampling and conducting laboratory analyses, which consisted solely of cotton fibers within a size range of 839.22–3824.2 μm. In this study, two cotton particles were observed in the blank samples collected during field sampling, while seven cotton particles were detected during the laboratory work analysis.

2.6. Data analysis

The age of the sediment was estimated by analyzing the concentration profiles of $^{210}Pb_{ex}$. The estimation of this feature has been commonly conducted using three models, i.e., CF-CS (Constant Flux and Constant Sedimentation Rate), CF-CIC (Constant Flux and Constant Initial Concentration), and CRS (Constant Rate of Supply) (Sanchez-Cabeza et al., 1999). The age of the sediments was determined using the CRS model due to the observation that the $^{210}Pb_{ex}$ profiles, as revealed by our measurements, did not exhibit an exponential decrease.

The analysis of microplastic abundance and characteristics was carried out using PAST software version 4.03. To determine the relationship between the number of microplastics found in the sediment and the depth in each sampling area, a significant test was conducted utilizing non-parametric analysis (Kruskal-Wallis test followed by Dunn's post hoc analysis). A significance level of 0.05 was adopted for all statistical tests undertaken.

3. Results and discussion

3.1. Sediment archive

All sediment cores exhibited a discernible decline in the concentration of $^{210}Pb_{ex}$ as the depth increased, except the core obtained from the mudflat habitat in Jakarta(Fig. 2). The observed decline in $^{210}Pb_{ex}$ levels within sediment cores would be unlikely to occur if rapid physical and biological disturbances caused substantial sediment mixing. Therefore, the decreasing $^{210}Pb_{ex}$ profiles in Fig. 2 can be used to determine sediment age. This approach has been widely used including for sediment dating of mangrove habitat (Minu et al., 2018; Sanders et al., 2012).

Sediment layers, dated back 70 years, were identified at varying depths between 16 and 50 cm (Fig. 3). Moreover, our dating approximation aligns with prior scholarly investigations. Based on the findings reported from other countries, for example Breithaupt et al. (2012), it has been determined that the global sedimentation rate in mangrove sediment is approximately 2.8 mm per year. This information suggests that the sediment layer at a depth of 19 cm can be estimated to be approximately 70 years old. Furthermore, in their study in Kuwait Bay, Uddin et al. (2021) utilized the radioactive isotope ^{210}Pb to identify the

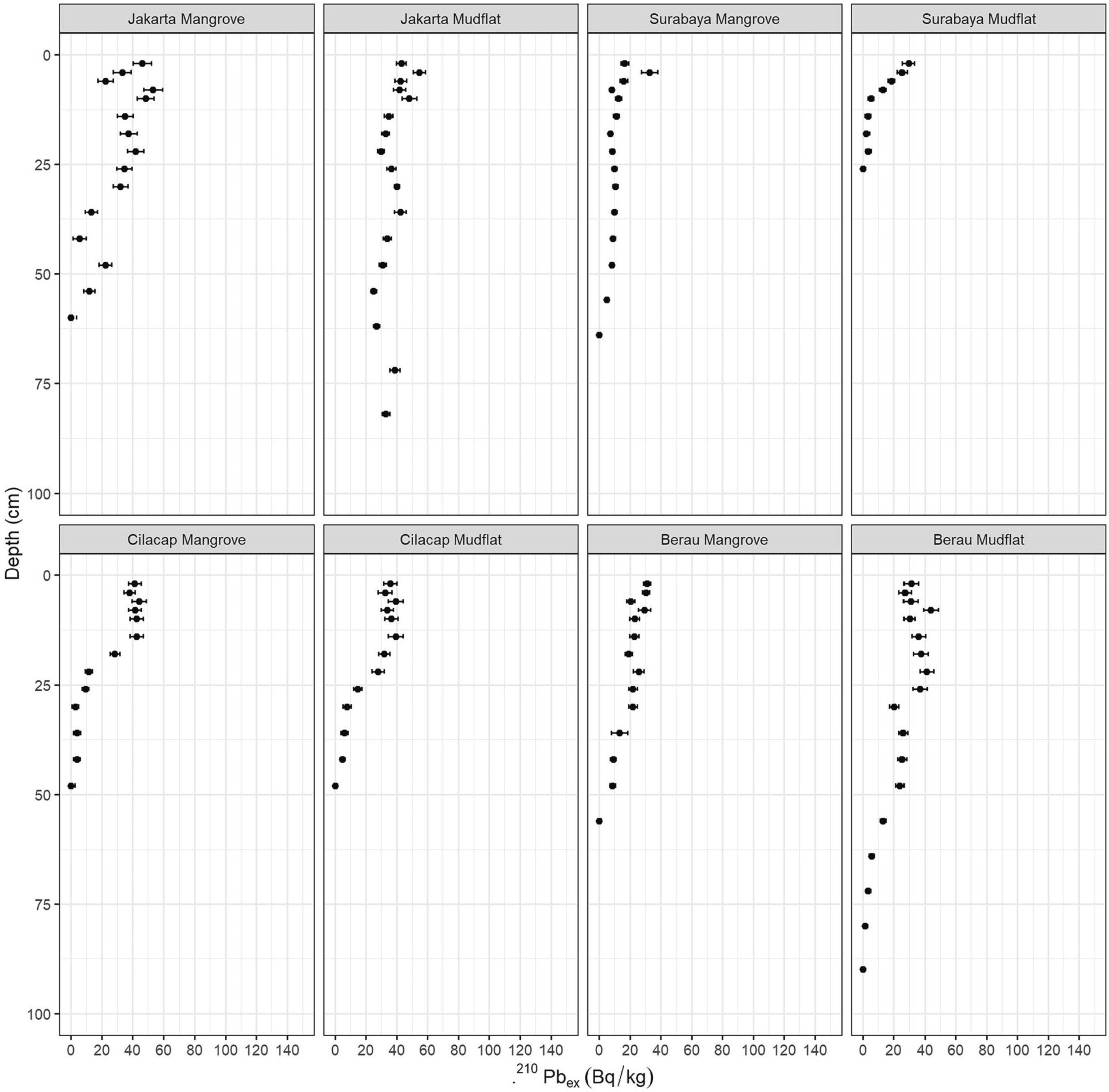

Fig. 2. Excess ^{210}Pb profiles used to model sediment age in mangroves and mud flats of the four different locations with high (Jakarta and Surabaya) and low (Cilacap and Berau) levels of anthropogenic activity. Bq/kg, Becquerel per kilograms, the unit for radioactive decay.

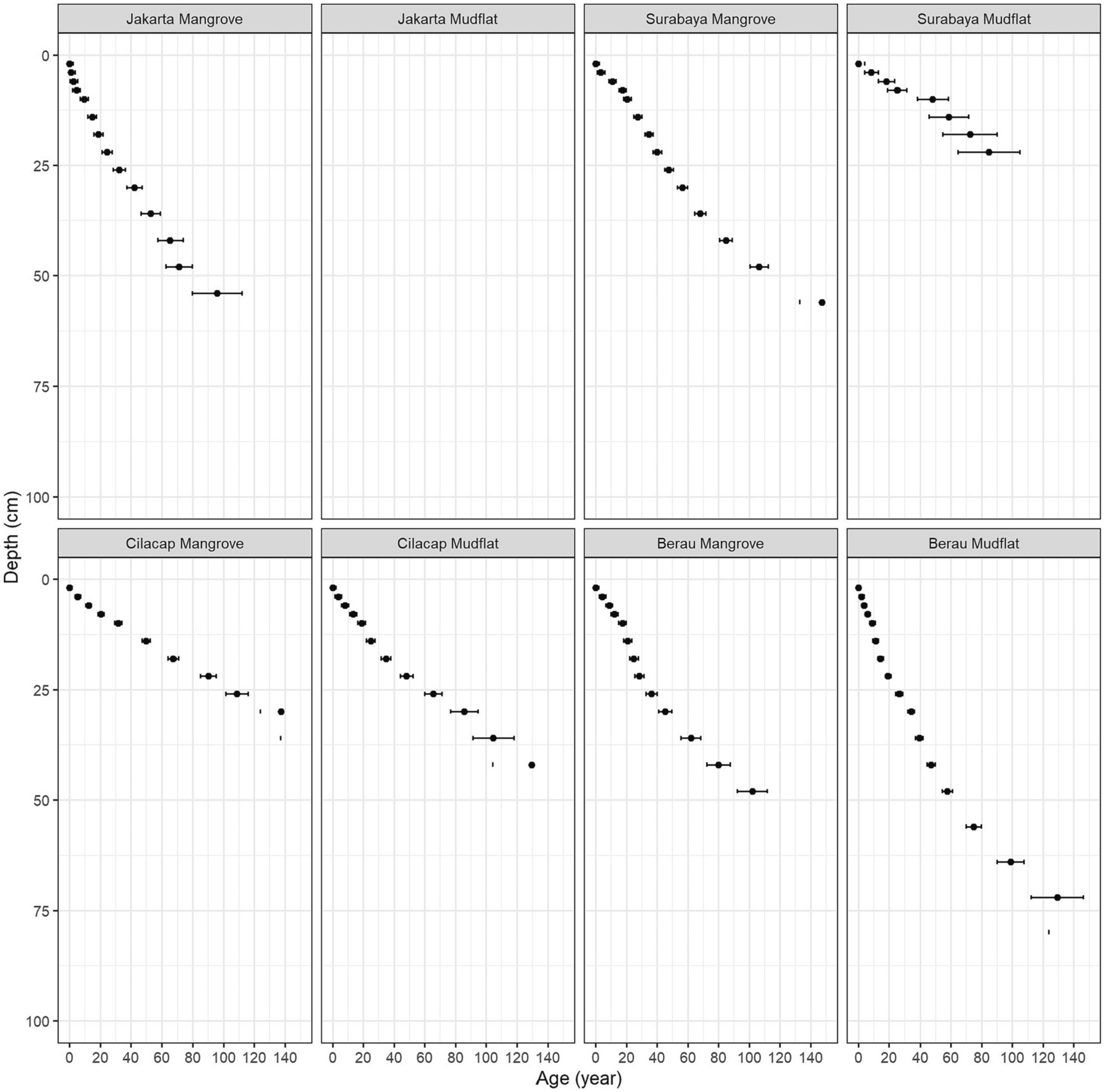

Fig. 3. Modelled age of sediment cores in mangroves and mud flats of the four different locations with high (Jakarta and Surabaya) and low (Cilacap and Berau) levels of anthropogenic activity.

presence of a sediment layer spanning 60 years, which was found at a depth of 50 cm. Hapsari et al. (2020) reported their findings in Segara Anakan Lagoon, Cilacap, that a core sample at a depth of 50 cm corresponds to a 21-year-old. In this case, the core sample is young, having been collected from an area with active sedimentation (Ardli and Wolff, 2009). Meanwhile, we obtained a core sample from Segara Anakan Lagoon, Cilacap but from a different corner, where the sedimentation rate is relatively low; and a 100-year-old sediment was discovered in the core sample at depths of 25 and 30 cm (mangrove and mudflat respectively). The dating of sediment cores in Surabaya is comparable to that reported in Rositasari and Purbonegoro (2021), who discovered a 100-year-old sediment at a depth of 50 cm from a core sampler collected at Lamong Bay, Surabaya. Our sediment dating for Berau sediment cores produced the same result as Dewi et al. (2020), with sediment layers <45 cm having an age of 100 years.

3.2. *Microplastic abundance*

Microplastics were successfully isolated and examined in all sediment core samples, with their presence observed in 189 out of the total 242 segments investigated within the sediment core. This research is the initial recording of microplastics found in sediment layers as deep as 10 cm within the mangrove region of Indonesia. To our knowledge, no prior research has documented this occurrence before. The levels of microplastic abundance varied across different layers of the sediment core (Fig. 4) collected from mangrove and mudflat regions spanning Jakarta (69.86 ± 27.53 and 78.52 ± 29.48 pieces/kg-dry sediment), Surabaya (103.17 ± 30.80 and 103.00 ± 30.38 pieces/kg-dry sediment), Cilacap (10.51 ± 5.06 and 9.62 ± 7.56 pieces/kg-dry sediment) and Berau (19.69 ± 10.93 and 16.78 ± 12.83 pieces/kg-dry sediment), respectively.

Based on the statistical analysis using mean values and conducting a

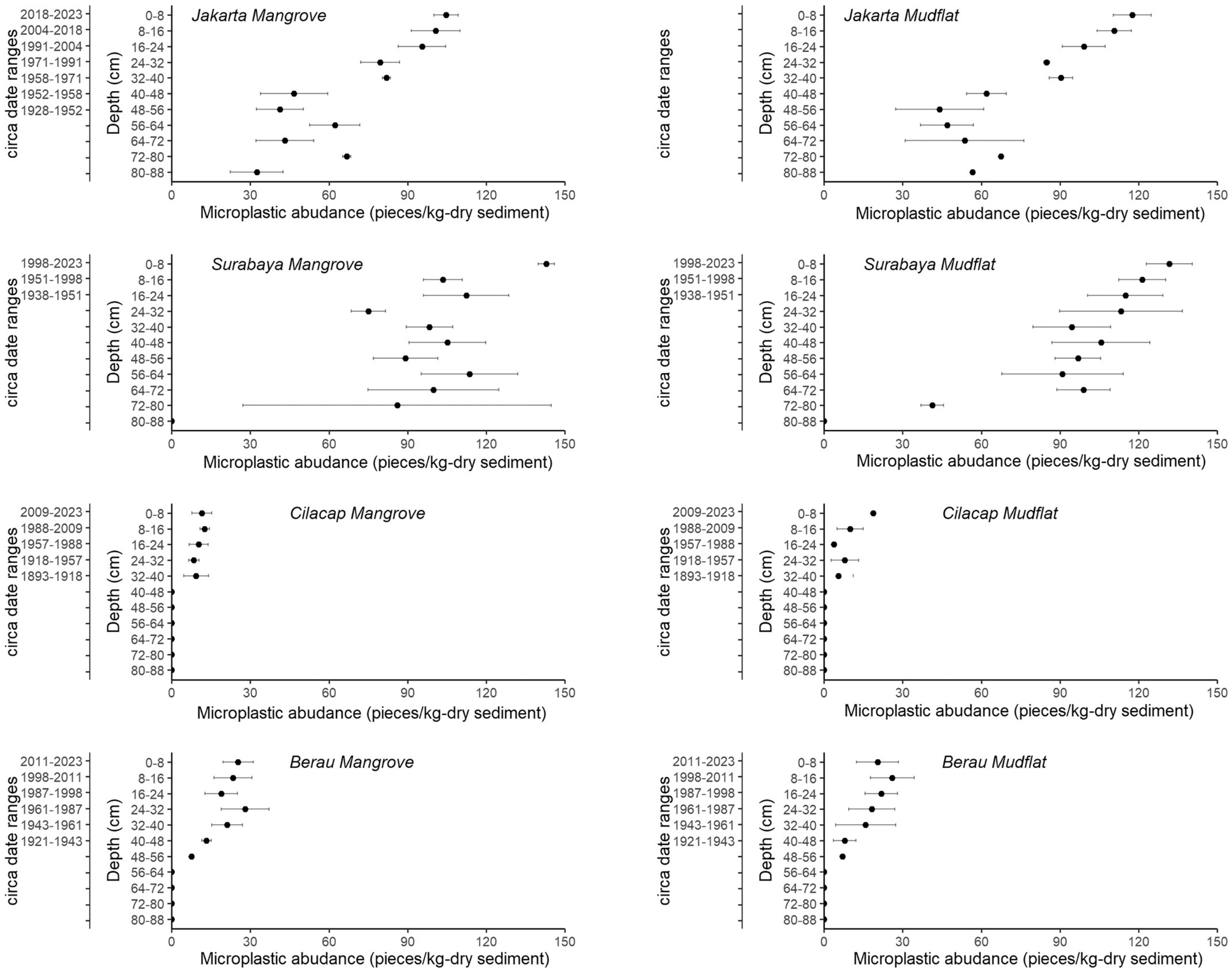

Fig. 4. The depth profile of microplastic abundance from all sediment cores obtained from mangroves and mud flats of the four different locations with high (Jakarta and Surabaya) and low (Cilacap and Berau) levels of anthropogenic activity.

Kruskal-Wallis test followed by Dunn's post hoc analysis ($p < 0.01$), it is evident that there is variation in microplastic abundance within sediment cores across. The ranking from highest to lowest abundance is as follows: Surabaya > Jakarta > Berau ≈ Cilacap. The microplastic in the sediment core in Surabaya (103.08 ± 30.32 pieces/kg-dry sediment) shows the highest abundance, followed by Jakarta (73.97 ± 28.56 pieces/kg-dry sediment). The levels of microplastics in Berau (18.24 ± 11.86 pieces/kg-dry sediment) and Cilacap (10.06 ± 6.35 pieces/kg-dry sediment) were found to be approximately similar. There is a significantly higher abundance ($p < 0.01$) of microplastics in mangroves and mudflats located in areas characterized by high levels of anthropogenic activity, e.g., Jakarta (69.86 ± 27.53 and 78.52 ± 29.48 pieces/kg-dry sediment, respectively) and Surabaya (103.17 ± 30.80 and 103.00 ± 30.38 pieces/kg-dry sediment, respectively). These regions exhibit microplastic concentrations that are 6–10 times greater compared to areas with lower anthropogenic influence, e.g., Cilacap (10.51 ± 5.06 and 9.62 ± 7.56 pieces/kg-dry sediment, in mangroves and mudflats respectively) and Berau (19.69 ± 10.93 and 16.78 ± 12.83 pieces/kg-dry sediment, in mangroves and mudflats respectively). It has been well-established that human-related activities play a crucial role in the presence of microplastics within various environmental settings (Browne et al., 2011; Luo et al., 2021; Yonkos et al., 2014). Previous studies have consistently identified sites proximate to anthropogenic sources as locations with elevated levels of microplastic content (Barboza and Gimenez, 2015; Browne et al., 2011; Cole et al., 2011; Li et al., 2018; Yonkos et al., 2014). Anthropogenic activities, such as human manufacturing and the utilization of plastic products, constitute the primary origins of microplastics (Lin et al., 2022). The escalating size of the population in the region is a potential factor contributing to an upsurge in plastic waste generation due to short product lifespans (Cai et al., 2018; Cordova and Nurhati, 2019; Häder et al., 2020; Iskandar et al., 2022; Lahens et al., 2018; Thompson et al., 2004). Notably, it should be highlighted that Jakarta and Surabaya are two major economic hubs in Indonesia. The Greater Jakarta area alone boasts a population exceeding 30 million people, while the greater Surabaya region is home to >10 million individuals (Statistics Indonesia, 2021).

Microplastics have also been detected in relatively pristine areas, including mangrove regions in Cilacap and Berau. These microplastics' primary source is the nearby vicinity, particularly through the transportation by adjacent river flows. In the case of the Cilacap mangrove area, microplastic particles likely originated from streams located on southern Java and Bali Island (Iskandar et al., 2021, 2022). They are transported along with plastic materials and eventually end up deposited in Cilacap's mangroves as a result of coastal accumulation processes. Furthermore, investigations indicate that microplastics accumulate within different layers of sediment found within mangrove ecosystems under the influence of ocean currents (Chen, 2022; Onink et al., 2019; Wang et al., 2020; Zhang, 2017). As for Berau, which directly adjoins the Celebes Sea, this region experiences complex patterns of oceanic currents carrying diverse substances - primarily originating from the Western Pacific via Mindanao Current and subsequently entering Makassar Strait through Indonesian Throughflow. Consequently, due to this convergence process involving mass transport and material confluxation, the levels of microplastic abundance significantly increase (Purba et al., 2021; Yuan et al., 2023). Current research on the correlation between human activities and microplastic contamination in highly populated regions is still progressing, necessitating a more comprehensive investigation into the association between these factors. In order to explore microplastic pollution concerning spatial elements, studies should exhaustively analyze the interconnection between human activities and this form of environmental degradation.

The abundance of microplastics in the sediment cores from Jakarta, Surabaya, Cilacap, and Berau mangrove areas was generally lower compared to Songkhla and Mae Klong mangrove areas in Thailand (Chaisanguansuk et al., 2023; Pradit et al., 2022), Tokyo Bay in Japan (Matsuguma et al., 2017), Pearl River estuary, Andong salt marsh, and the southeast coast of China (Fan et al., 2019; Li et al., 2020a; Yu et al., 2023). Similarly, these abundance was found to be approximately equal to the Pattani mangrove area and the Gulf of Thailand in Thailand (Chaisanguansuk et al., 2023; Pradit et al., 2022) as well as the Kuala Gula mangrove area in Malaysia (Mohamed et al., 2023). It is worth noting that this study observed a trend where the abundance levels varied across different sampling locations within these regions. According to the observed pattern, it can be inferred that there is a similar trend in the accumulation of microplastics in the mentioned areas. The concentration of microplastics tends to decrease as sediment depth increases.

3.3. Driving forces of microplastic deposition in sediment

The distribution of microplastics in each region exhibits a consistent pattern, whereby their abundance decreases with increasing sediment depth (Figs. 2 and 3). In the Jakarta and Surabaya areas, microplastics were detected in almost all segments of the sediment core, specifically 59 out of 66 segments for Jakarta and 58 out of 60 segments for Surabaya. However, certain portions within the sediment cores from these regions did not contain any microplastics; this was observed in several segments within the depth range of 64–88 cm. Slight variations were noted in Cilacap (30 out of 60) and Berau (42 out of 66). In Cilacap's mangrove and mudflat zones, samples collected beyond a depth exceeding 40 cm no longer exhibited any presence of microplastic particles. Similarly, for Berau's sediment core sample collection, no further traceable presence of microplastic after reaching 56 cm. Our study findings suggest that the small-sized plastic particles in Indonesia's mangrove area, which encompasses both mangrove forests and mudflats, are likely significant accumulators of the buildup of plastics in sediment. Our analysis of sediment cores revealed a substantial presence of plastic waste in the uppermost layer (up to 16 cm) of sediments. This level of plastic accumulation is similar to what has been observed in other mangrove sites, such as those found along the Arabian Gulf coast (Martin et al., 2020), southern Thailand's mangroves (Pradit et al., 2022), and Malaysia's Kuala Gula Mangrove (Mohamed et al., 2023). Accordingly, it can be inferred that Indonesian mangroves harbor comparable amounts of microplastics compared to these regions.

Microplastic distribution within depth was generally expected to be linear with sediment ages (Simon-Sánchez et al., 2022), but this is not always the case, as a review study discovered (Martin et al., 2022). Microplastic abundances decreased in this study as sediment layers aged up to the 1950s (dated ~70 years), when Indonesia's plastic industry began, but microplastics were still detected in sediment layers aged 80–100 years or between 1923 and 1943 (Fig. 4). Numerous studies have reported a strong relationship between microplastic abundance and the age of sediment layers, for example, in the Tien Bay and Red River Delta, Vietnam (Dung et al., 2021), and Kuwait Bay (Uddin et al., 2021), where the abundance decreases with age and disappears when it exceeds the first year of plastic production in these countries. In contrary, Xue et al. (2020) discovered microplastic at a depth of 60 cm, which corresponds to the 1897 CE deposit. Microplastics were also discovered in sediment layers of 80–110 cm in depth, dating between 1900 and 1945, or when plastic material was first industrialized globally (Belivermiş et al., 2021). Therefore, microplastics were deposited chronologically with an increasing trend in sedimentary environments, but may also experience downward migration, which Martin et al. (2022) have also been noted.

Numerous investigations have provided extensive evidence of the long-term accumulation of microplastics in sedimentary environments. Again, this study highlights the distinctive nature of microplastic aggregation in sediments based on chronological analysis, revealing their presence at depths corresponding to an age exceeding 70 years. The proliferation of plastic usage began exponentially approximately 70 years ago, coinciding with the advent of mass production during the 1950s. Contrasting somewhat with prior research conducted in Thailand (Chaisanguansuk et al., 2023; Matsuguma et al., 2017), where microplastics were solely identified within a shallow layer (above 12 cm), absent below this depth, our findings from mangrove sites encompassing Jakarta, Surabaya, Cilacap and Berau diverge slightly. Nonetheless, our data remain consistent with studies from Türkiye (Belivermiş et al., 2021), Kuwait (Aba et al., 2014) and Japan (Matsuguma et al., 2017) which also describe early onset accumulation predating widespread plastic manufacturing.

The reasons for the presence of microplastics in sediment layers that are >80 years old found in this study are still not clearly understood. There are several premises to explain this phenomenon. One assumption is related to the history of aquaculture ponds in Jakarta and Surabaya, where these areas were previously used for such purposes (Mayalanda et al., 2014; Prasita et al., 2019). It is possible that during those times, the sediments were disturbed, causing microplastics to become buried deeper within them (Hargan et al., 2020; Yang et al., 2022). Another premise suggests that infiltration may play a role in transporting microplastics into deeper sediment layers (Coppock et al., 2021; Waldschläger and Schüttrumpf, 2020). This process can be influenced by various factors, including properties of the microplastics themselves, soil texture, flow patterns, and disturbances caused by organisms living within the ecosystem (Dong et al., 2022; Dunn et al., 2019; Guo et al., 2022; Xu et al., 2020). Mangrove regions, including mudflat zones, are affected by tidal movements and are typically composed of loose sediments with high porosity (Al-Khayat and Alatalo, 2021; Dunn et al., 2019). Microplastic particles have been observed moving through cracks within soils or transferred from surface layers down into deep sediment layers via leaching or bioturbation processes facilitated by benthic invertebrates (Coppock et al., 2021; Huerta Lwanga et al., 2016; Riani and Cordova, 2022). Given the limited amount of research on this topic, further in-depth investigations are required to fully understand and elucidate the underlying processes associated with this issue. The scarcity of scholarly studies addressing this matter necessitates a comprehensive exploration to provide more extensive knowledge and insights into its mechanisms.

3.4. Microplastic characteristic

The physical characteristics, size distribution, and chemical composition of microplastics have been proposed as factors that can help identify their sources (Auta et al., 2017). Through our analysis, four distinct shapes of microplastics were observed: fragments, fibers, foam particles, and granules. Among these shapes, fragments were the most prevalent in sediment cores, accounting for 52.21 % of the total particles identified (Fig. 4). Fibers followed closely behind at 44.55 %. These two types - fragments and fibers - have consistently emerged as dominant forms among numerous studies conducted in coastal areas, estuaries, and deep-sea environments (Chaisanguansuk et al., 2023; Kukkola et al., 2022; Martin et al., 2019, 2020; Pradit et al., 2022; Simon-Sánchez et al., 2022). Fragments are commonly derived from secondary microplastics that originate from various applications such as packaging materials, instant food or beverage containers, cushions, and insulation materials (Andrady, 2017; Liu et al., 2019; Nurhasanah et al., 2021; Sulistyowati et al., 2022). Fiber-shaped microplastics, conversely, tend to be generated through textile manufacturing processes where staple fibers or filaments are released during washing activities (Courtene-Jones et al., 2020; Dong et al., 2020). This may include fibrous waste discarded by fishing-related operations (Cordova et al., 2022b; Jeyasanta et al., 2020).

Microplastic infiltration was observed to vary across the four sampling areas, with Jakarta and Surabaya exhibiting deeper levels of infiltration. In both Cilacap and Berau sediment cores, microplastics in the shape of granules and foam were found at depths up to 16 cm and 24 cm respectively, relative to the top layer of sediment. Interestingly, a distinct feature is observed in the Surabaya sediment core where granule and foam microplastics are discovered as deep as 48 cm; notably deeper than those found in Jakarta which reach only up to a depth of 32 cm. This discrepancy can potentially be attributed to the concentration of plastic production facilities along with their recycling operations predominantly conducted within Greater Surabaya since around 1960 until present day, whereas Greater Jakarta exhibited similar activities until mid-2015. Nonetheless, there was no difference in the depth profiles of microplastic shapes collected from mangrove habitats and mudflats in all sampling locations (Fig. 5).

Microplastics of varying sizes were detected in all sampling areas and also no difference in depth profiles between mangrove habitats and mudflats was observed (Fig. 5). The most commonly observed size category was within the range of 200–500 μm, accounting for 36.42 % of the total microplastic count. This was followed by sizes ranging between 500 and 1000 μm (28.36 %), 1000–2000 μm (20.78 %), and 2000–5000 μm (14.44 %). Notably, an increase in depth corresponded to a higher prevalence of microplastics in the smaller size category (200–500 μm), while near-surface sediment cores exhibited a larger proportion of particles falling within the range of 1000–2000 μm. Previous studies have indicated that small-sized microplastics tend to dominate due to their strong affinity for natural colloids, leading to increased precipitation rates with sediments over time (Besseling et al., 2017; Filella, 2015).

According to the findings of this study, it is postulated that the high proportion of small plastic sizes in mangrove ecosystems can be attributed to fragmentation experienced by larger plastics on the surface (Koelmans et al., 2017; Song et al., 2017). Larger-sized microplastics (>1000 μm) can be present as a result of mangrove roots acting as traps for relatively bigger debris (Okuku et al., 2023; van Bijsterveldt et al., 2021). These fragmented particles are likely to infiltrate deeper into sediment layers. The density of microplastics plays a crucial role in determining their vertical transport and behavior, including whether they float or sink (Kooi et al., 2017). Typically, microplastics with higher densities sink, while those with lower densities remain near the water surface (Ding et al., 2019; Willis et al., 2017). Environmental factors such as weathering and biofouling can affect the ability of microplastics to remain buoyant or become denser, leading to fluctuations in their overall density (Li et al., 2020b; Matsuguma et al., 2017). This variability contributes to processes like flotation and sedimentation that influence the fate of microplastics (Hinata et al., 2023; Long et al., 2015). Further investigation is necessary to fully understand this phenomenon as the residence time of microplastics greatly influences their remobilization process and subsequent exposure (Rillig and Bonkowski, 2018; Waldschläger and Schüttrumpf, 2019), to organisms residing in mangroves ecosystem - habitats which support diverse marine organisms and play crucial ecological roles (Jiao et al., 2022; Li et al., 2020c; Luo et al., 2021). Moreover, a reduction in bulk density and decreased porosity within sediments similar to fine sediment infiltration processes results in the infiltration of microplastics (Ballent et al., 2013; Dong et al., 2022; Horton et al., 2017). The sediments examined during this study primarily consisted of silt and sand with limited amounts of clay, ranging from fine-grained sand to very fine-grained sand. Given these characteristics, it is highly probable that the tropical mangrove ecosystem area, specifically within the study site, experiences significant microplastic infiltration.

A total of 240 particles out of the recovered 481 particles (49.90 %) were found to be composed of nineteen different polymer groups (Fig. 6). The highest number of polymer types was observed in sediment cores collected from mangrove areas in Jakarta and Surabaya, with 16 and 15 types, respectively. On the other hand, sediment cores from

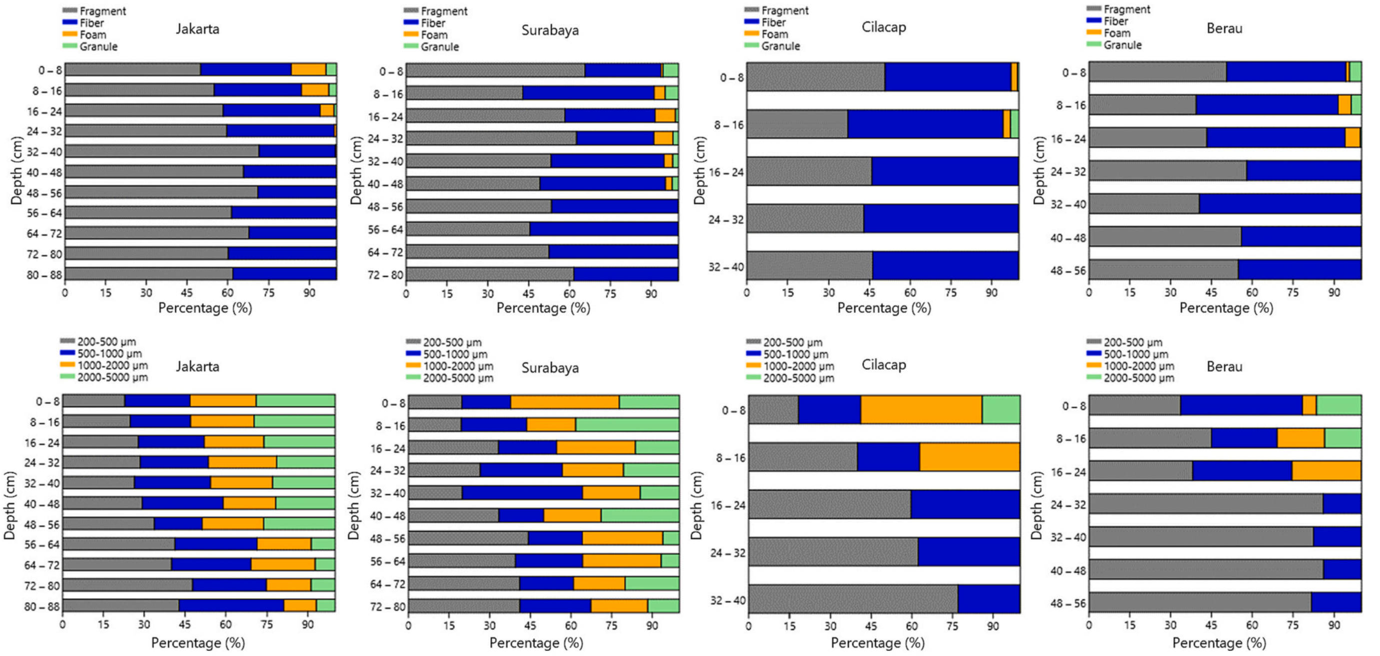

Fig. 5. The depth profiles of microplastics based on shapes and size.

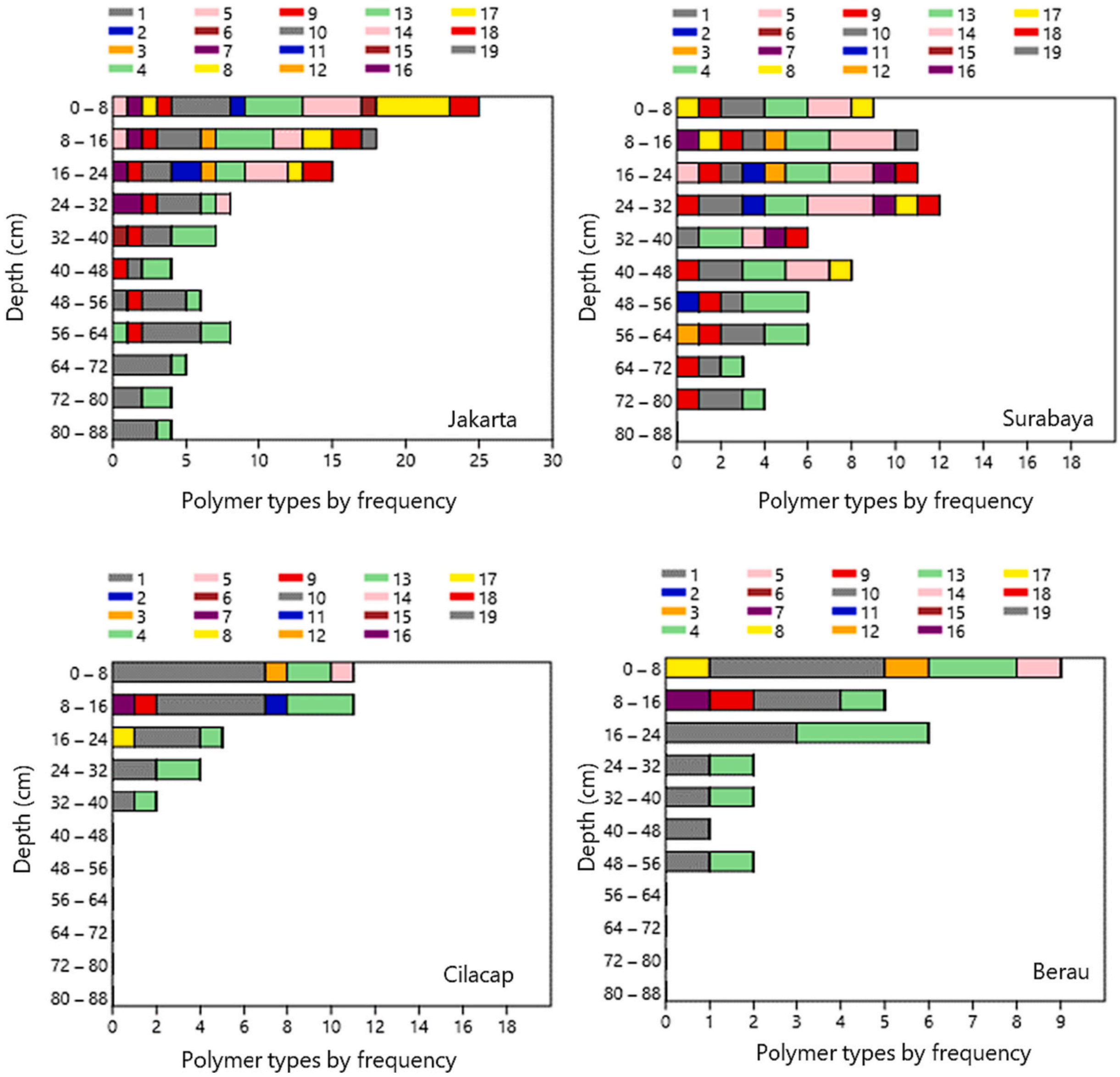

Fig. 6. The depth profiles of microplastics based on polymer types. (1) Butyl octyl phthalate, (2) Polyolefin (Oleafin fiber), (3) Peroxide Cure EPDM (ethylene propylene diene monomer rubber), (4) Poly(butylene terephthalate), (5) Polybutadiene, (6) Polychloroprene, (7) Polyester, (8) Polyethylene, (9) Polyethylene high density, (10) Polyethylene low density, (11) Polyethylene propylene, (12) Polyethylene terephthalate, (13) Polypropylene, (14) Polystyrene, (15) Polystyrene butadiene, (16) Polytetrafluoroethylene, (17) Polyurethane, (18) PVC (Polyvinyl chloride), (19) PVDC (polyvinyldine chloride).

Cilacap and Berau had a lower diversity, with only eight and seven types identified, respectively. When considering the depth of the sediment core, it was discovered that up to 13 different polymer types were present down to a depth of 32 cm. However, as we went deeper into the sediments, both the frequency and variety of polymers decreased. In this study, the majority of polymers identified were categorized into several types: low-density polyethylene (32.08 %), polypropylene (25.00 %), polystyrene (10.42 %), high-density polyethylene (7.92 %), polyurethane (4.58 %) and polyvinyl chloride (3.75 %). It is noteworthy that these three synthetic polymer groups, namely polyethylene, polypropylene and polystyrene, constituted >75 % of the total amount observed in this research. These particular types of plastics - polyethylene, polypropylene, and polystyrene - are widely prevalent as plastic debris in Indonesia's marine environment and within the larger global marine ecosystem (Erni-Cassola et al., 2019; Vriend et al., 2021). Gaining knowledge about the prevalent polymer groups within the sediment layer of mangrove areas is crucial in enhancing our comprehension of the specific plastic types found there (Cordova et al., 2022a). This information can then be correlated with findings from marine microplastic studies, further supporting the notion that fragmentation is a primary contributor to microplastic pollution in coastal and ocean environments (Chubarenko et al., 2018; Duis and Coors, 2016). Additionally, understanding which polymers are present offers benefits regarding environmental risk assessment and ecotoxicology due to unique additives associated with specific polymers that contribute to their ecological impact (Int-Veen et al., 2021). Microplastics, for example, have been found to accumulate in mangrove benthics (crabs, bivalves, and snails), with a high potential accumulation in mangrove crabs (Zhang et al., 2023). This is concerning for food security because mud crabs (*Scylla* spp.) are an important export commodity harvested from mangrove habitats (Hungria et al., 2017). Such insights enable researchers to conduct more thorough source tracing exercises for marine plastic debris (Gallo et al., 2018). Moreover, it is noteworthy that plastic pollution has persisted in Indonesia over an extended period and has permeated into the sedimentary layer. Consequently, there is a crucial need for comprehensive monitoring and effective management strategies (Cordova et al., 2021a). It is conceivable that the issue of plastic distribution and accumulation within sediment layers might possess intricate characteristics, potentially harboring risks beyond our current understanding.

4. Conclusion

Our research findings indicate that sediments serve as a valuable source of information on the historical and current release of plastic into marine ecosystems, particularly in mangrove regions. Microplastics were detected in nearly all samples, even in sediments dating back to periods before the mass production of plastics. Human activities have a significant impact on the presence of microplastics across various environmental contexts. It was observed that the ranking from highest to lowest abundance of microplastic is as follows: Surabaya > Jakarta > Berau ≈ Cilacap. In areas with strong anthropogenic influence (Surabaya and Jakarta), where levels of human-related activity are high, microplastic abundance is significantly higher (6–10 times) compared to areas with lower anthropogenic influence (Cilacap and Berau). The distribution pattern for microplastics within each region consistently reveals a decrease in abundance with increasing sediment depth. Moreover, the length of time that microplastics remain within a specific environment plays a significant role in their ability to be redistributed and subsequently exposed. In this study, we discover the presence of microplastics in sediment layers that are older than 80 years. The underlying causes behind these circumstances have yet to be comprehensively explained. One possible explanation pertains to the previous utilization of aquaculture ponds in Jakarta and Surabaya, which could have led to disturbances within these areas. As a result, microplastics may have become buried deeper into the sediments during those times. Another postulation suggests that infiltration could facilitate the transportation of microplastics into lower sediment layers. The presence of these microplastic characteristics highlights the significant contribution they make to plastic pollution in our oceans. As a result, there is an urgent requirement for thorough surveillance and efficient strategies to handle this matter. It is foreseeable that the problem of plastic dispersion and buildup within sediment layers may exhibit complex attributes, potentially carrying risks that surpass our current level of comprehension.

CRediT authorship contribution statement

Muhammad Reza Cordova: Conceptualization, Methodology, Data curation, Validation, Formal analysis, Investigation, Funding acquisition, Visualization, Supervision, Writing - original draft, Writing - review & editing. **Yaya Ihya Ulumuddin:** Conceptualization, Validation, Resources, Project administration, Funding acquisition, Visualization, Supervision, Writing - original draft, Writing - review & editing. **Ali Arman Lubis:** Data curation, Formal analysis, Validation, Investigation. **Muhammad Taufik Kaisupy:** Data curation, Formal analysis, Validation, Investigation. **Singgih Prasetyo Adi Wibowo:** Data curation, Formal analysis, Validation, Investigation. **Riyana Subandi:** Data curation, Formal analysis, Validation, Investigation. **Deny Yogaswara:** Data curation, Formal analysis, Validation, Investigation. **Triyoni Purbonegoro:** Data curation, Formal analysis, Validation, Investigation. **Jeverson Renyaan:** Data curation, Formal analysis, Validation, Investigation. **Doni Nurdiansah:** Data curation, Formal analysis, Validation, Investigation. **Untung Sugiharto:** Data curation, Formal analysis, Validation, Investigation. **Dienda Shintianata:** Data curation, Formal analysis, Validation, Investigation. **Sonia Saraswati Meiliastri:** Data curation, Formal analysis, Validation, Investigation. **Faza Putri Andini:** Data curation, Formal analysis, Validation, Investigation. **Suratno:** Data curation, Formal analysis, Validation, Investigation. **Muhammad Ilman:** Data curation, Validation, Investigation, Project administration, Funding acquisition. **Aji Wahyu Anggoro:** Data curation, Validation, Investigation, Project administration, Funding acquisition. **Basir:** Data curation, Validation, Investigation, Project administration, Funding acquisition. **Simon M. Cragg:** Data curation, Validation, Investigation, Project administration, Funding acquisition, Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found in the online version, at https://doi.org/10.1016/j.marpolbul.2023.115517. These data include the Google map of the most important areas described in this article.

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