



Anthropocene microplastic stratigraphy of Xiamen Bay, China: A history of plastic production and waste management

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ABSTRACT

Microplastics (MPs) are considered one of the significant stratigraphic markers of the onset of the Anthropocene Epoch; however, the interconnections between historic plastic production, waste management as well as social-economic and timing of MP accumulation are not well understood. Here, stratigraphic data of MPs from a sediment core from Xiamen Bay, China, was used to reconstruct the history of plastic pollution. Generalized Additive Modeling indicates a complex temporal evolution of MP accumulation. The oldest MPs deposited in 1952 was $30,332 \pm 31,457$ items/kg•dw, coincide with the infancy of the plastic industry and onset of the Anthropocene. The Cultural Revolution (1966–1976) curtailed these initial increases. Subsequent rapid growth in MPs during the late 1970s was peaked at $189,241 \pm 29,495$ items/kg•dw in 1988 and was followed by a drastic decline in the late 1980s to a low value in 1996 ($16,626 \pm 26,371$ items/kg•dw), coinciding with proliferation of MP sources, coupled with evolution of plastic production, consumption, and regulation. Increasing MPs over the past decades implies that previous mitigation measures have been compromised by the escalated influx of MPs from increasing plastics production, legacy MPs remaining in circulation and insufficient waste management for a growing population. The present methodology and results represent a conceptual advance in understanding how changes in policy and economics over time correlate to changes in MP records in Anthropocene strata, which may help make decisions on plastic pollution mitigation strategies worldwide.

1. Introduction

Plastic, as one of four basic construction materials in modern society along with steel, wood and cement (China Plastics Processing Industry Association, 2002), has permeated environmental systems globally. Editorials (Edwards, 2018) in the past few decades, and become a persistent pollutant due to its durability (Andrady, 2011). Plastic waste has been dispersed into the environment since its inception (Chen et al., 2020; Eerkes-Medrano et al., 2015; Ivar do Sul and Costa, 2014; Kane and Clare, 2019; Liro et al., 2020), yet it was not until the 1980s that it

became increasingly recognized as a marine ecological hazard that had been apparent as early as the 1960s–1970s (Ryan, 2015). Plastic pollution has slowly become a global environmental concern since Thompson et al. (2004) coined the term microplastics (MPs), finding them in plankton samples back to the 1960s and with subsequent increasing abundance over time. MPs range from 1 μm to 5 mm and include primary plastics such as microbeads or microfibers, or secondary plastics formed by fragmentation of macroplastics into smaller particles (Frias and Nash, 2019). In recent years, MPs have become considered a marker of the Anthropocene Epoch dating from the mid-20th century,

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which closely coincides with the start of extensive plastic production (Zalasiewicz et al., 2016). The significance of MPs to the concept and understanding of the Anthropocene has led to the suggestion of the time interval being alternatively named the Plasticene (Haram et al., 2020). The development of the plastic industry and waste management practices (Lau et al., 2020) have driven the evolutionary history of plastic pollution by directly regulating its provenance, but the extent to which it temporally correlates with changing human activities remains poorly understood (Brandon et al., 2019; Dong et al., 2020; Matsuguma et al., 2017; Turner et al., 2019; Wang et al., 2018). There is therefore a pressing need to investigate the historical changes of plastic pollution and its coupled relationship with human activities, notably the effectiveness of waste management, which in turn can formulate targeted strategies to minimize plastic pollution (Rochman et al., 2013; Wang et al., 2020; Zalasiewicz et al., 2019; Zheng and Suh, 2019).

Plastic waste entering the ocean continues to increase with global plastic production and waste emissions (Hurley et al., 2018; Jambeck et al., 2015; Law and Thompson, 2014; Lebreton et al., 2017; Ryan and Moloney, 1993), with more than half of plastics produced calculated to be deposited on the seafloor (Frias et al., 2016; Moore, 2008), rendering it a key ultimate sink (Thompson, 2015). Due to the persistence of plastics in coastal and marine sediments (Ivar do Sul and Costa, 2014; Kane et al., 2020; Thompson et al., 2004) they will ultimately form distinctive technofossils (Zalasiewicz et al., 2014), and a unique geochemical signal of the Anthropocene (Crutzen, 2002; Waters et al., 2018). This suggests that interannual and decadal records of MPs in marine sediments (Bancone et al., 2020; Waters et al., 2016; Zalasiewicz et al., 2016) will allow us to reconstruct the history of plastic pollution during the Anthropocene. Recent global simulations of marine plastic transport, indicate that at least 77% of buoyant marine plastic debris released from land-based sources are trapped in coastal areas (Onink et al., 2021). In the coastal zone, MPs tend to accumulate within low-energy environments close to urbanized regions and anthropogenic sources (Ballent et al., 2016; Waters et al., 2018), especially the coastal reserves around rapid urbanization which provide ideal natural archives for MP records (detailed discussion in the section of *Sampling* in Supplementary Information, hereinafter SI).

While mapping MP pollution history through coastal sediment is challenging due to multiple mechanisms controlling spatial and temporal distributions of MPs and the time-consuming and labor-intensive procedure for extracting individual particles from environmental samples reliably, leading to often biased and incomparable results (Löder et al., 2015). Some literature has discussed the effect of global plastic output on the temporal distribution of MPs in coastal sediments (Brandon et al., 2019; Claessens et al., 2011; Martin et al., 2020; Matsuguma et al., 2017; Ostle et al., 2019; Wang et al., 2018), but well-known information about the multiple impacts of regional and local plastic production, use, and interventions to reduce plastic waste (Lau et al., 2020) on Anthropocene MP strata is scarce, which hinders our understanding of the potential hazards and fate of marine MPs. Beyond provenance, the influence of the sedimentary environment on the MP records remains unclear, and only a few studies have examined this issue; notably that sediment grain size is a critical proxy for MP abundance, at least for polymer types with higher densities when compared to seawater (Enders et al., 2019; Kane and Clare, 2019; Strand et al., 2013). Therefore, investigations on the collective mechanisms governing the potential historical inputs of MPs to coastal settings are warranted.

Here, we collected marine sediment cores at Xiamen Bay and dated the deposition rate through ^{210}Pb and ^{137}Cs analysis, using Focal Plane Array-micro-Fourier Transform Infrared (FPA- μFTIR) to investigate the characteristics of MP records. Based on the analysis of the sedimentary environment, we reconstructed the Anthropocene MP succession since the 1950s. Meanwhile, we identified and quantified the main responsible provenances of MPs and established their changing abundance since 1950. We then discuss the interconnections between the MP records in Anthropocene strata and historic plastic production, waste

management, and socioeconomics through a General Additive Model. The objective of this paper is to test the hypothesis that the depth distribution of MPs in marine sediments is primarily controlled by the historic human activities related to plastics, to evaluate the evolution of MP pollution during the Anthropocene, and to help formulate strategies for human interventions in mitigating plastic pollution.

2. Materials and methods

2.1. Study area and sampling methodology

Xiamen Bay of China (Fig. 1), a typical East Asian coastal bay, was chosen as the study site since it is home to nearly 2000 marine species, including protected species such as the little egret (*Egretta garzetta*), as well as a vital part of the surrounding human socioeconomic development, and approximately 76.67% of its plastic debris comes from human activities in the adjacent land (Chen et al., 2019). The sediments in Xiamen Bay, which come from the Jiulong River, adjacent land and the Taiwan Strait, exhibit a wealth of terrestrial characteristics attributed to intensive human activities surrounding the bay (Fang et al., 2010a) (e.g., the historical change in coastline and coastal land reclamation in recent decades, Fig. 1). The surface sediment migration trajectory is shown in Fig. 1, and the historic patterns of runoff and sediment discharge from the Jiulong River over the last 50 years are shown in Fig. S1. The salinity of Xiamen Bay is 26‰ to 30‰, with an average of 26‰ in the Jiulong Estuary controlled by tidal currents and freshwater upstream, decreasing from east to west.

Xiamen is one of China's four special economic zones and the largest coastal city near Xiamen Bay, with the 7th and 14th highest container throughputs in China and the world, respectively (JOC, 2019). Following China's economic reform and opening up that started in 1978, Xiamen experienced a rapidly developing residential and industrial economy (e.g., reclamation in Fig. 1), including the rise of the plastics industry. In recent years, the annual output of plastic products in Xiamen has been beyond 2.0×10^5 tonnes, and the competitiveness of the plastic industry has ranked first in Fujian Province for many years (Fujian Enterprise Information Center, 2017). The economic and industrial development record related to plastics in Xiamen facilitated the assessment of field observations of MPs in sediment cores in relation to historical human activities. If the Jiulong River is a major supplier of MPs to the estuary one might expect MPs to increase in the core at times of high runoff. If it is not a large supplier of MPs then conversely times of high sediment loads might be expected to result in dilution of the MPs in the core at that time. There is no evident systematic relationship between Fig. S1 and the MP abundance in Fig. 2a, suggesting that Jiulong river didn't have an important contribution. The population upstream Jiulong river is relatively small compared than that along the coast and Xiamen city. In addition, there is inadequate data on MPs from the catchment area of the Jiulong River, therefore, plastic discharges from the upstream Jiulong River were not considered herein. Four marine sediment cores were collected at Xiamen Bay using gravity corer with white polyvinyl chloride (PVC) tubes (outer diameter $\varphi = 75$ mm) on March 18, 2017 (Fig. 1, SI *Sampling* and Table S1). The cores were sealed immediately onsite using PVC end caps, which were secured with electrical tape to keep the original state of the sediment with minimal interference and avoid potential atmospheric plastic contamination. One of the sediment sites (GY) is situated in the middle zone of the Jiulong estuary close to the Jiyu Nature Reserve, although near a wastewater effluent site (Fig. 1). The vertical salinity in the GY core located area is sufficiently mixed and the stratification is not significant in the water column (Yu et al., 2012; Zhang et al., 2008). Sampling sites WY and HS were selected to avoid shipping lanes and are close to the undeveloped coastline, with HS located farther out into the bay. The characteristics of the sediment cores are shown in Table S1.

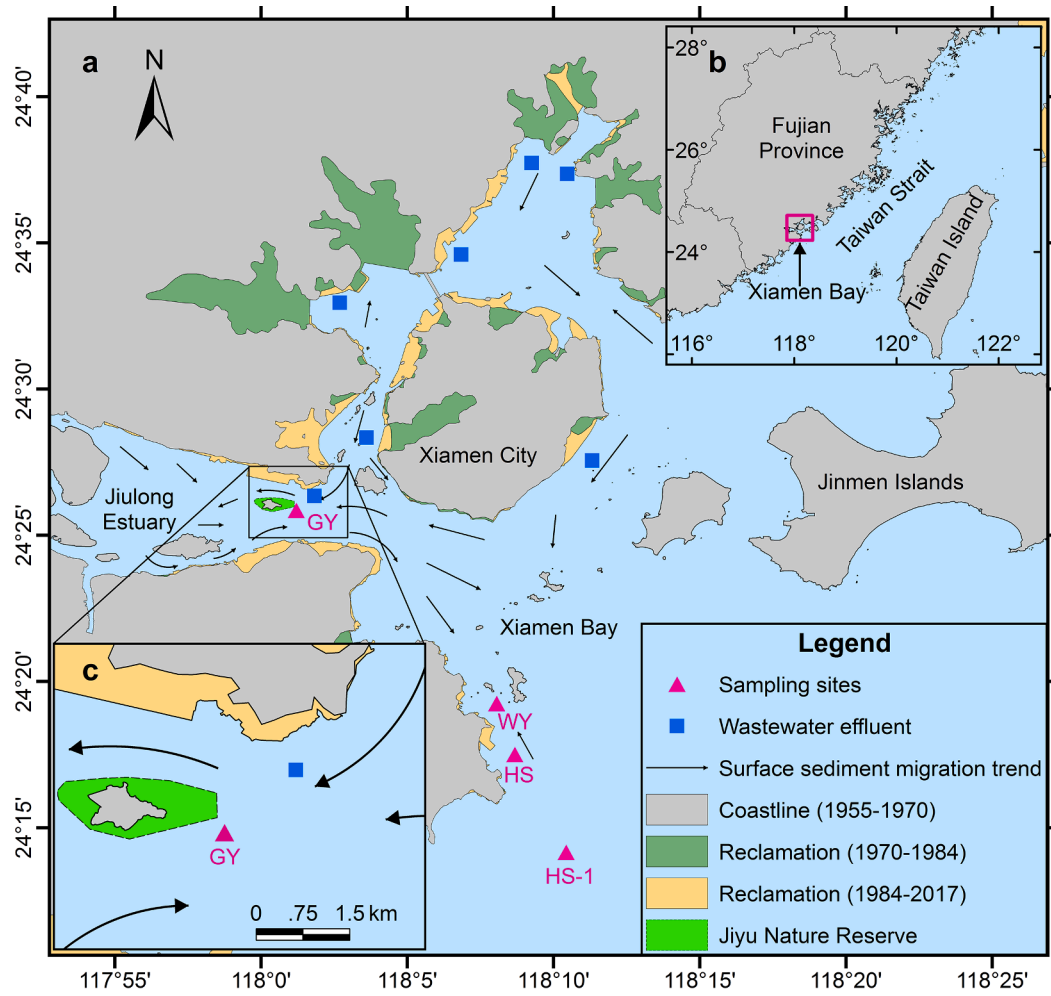


Fig. 1. Sampling locations for sediment cores, and the historical change of coastline and coastal land reclamations during 1970–1984 and 1984–2017 in Xiamen Bay (a–c) (Lu et al., 2011). The surface sediment migration trend was modified from (Cai et al., 1999; Fang et al., 2010b; Zuo et al., 2011). Sediment core sampling in Xiamen Bay, China (d,e).

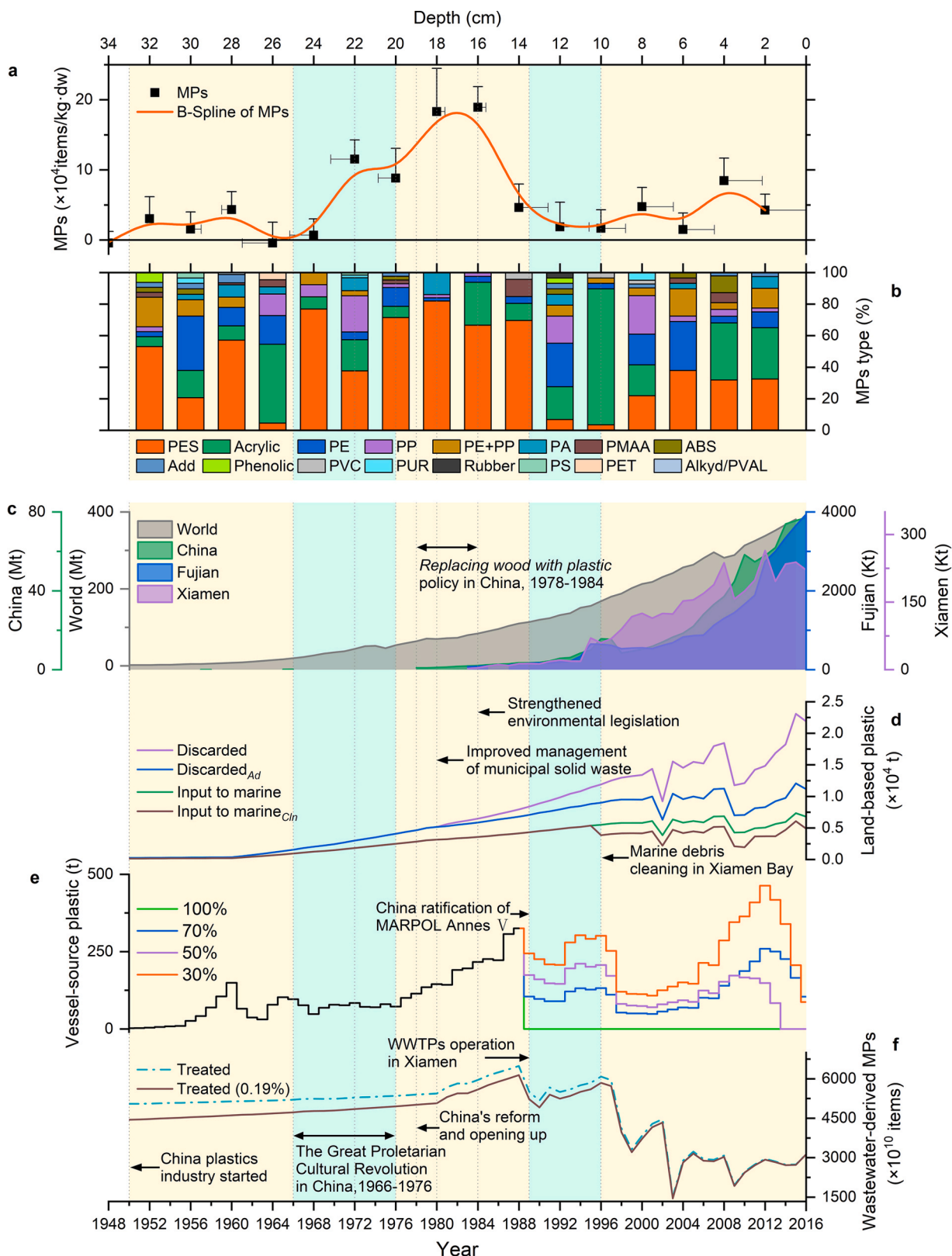


Fig. 2. Temporal variations of MP distribution in Xiamen Bay and key footprints of human plastic activities. (a) Stratigraphic profile of MP abundance. The X-axis error was based on the mass depth calculated from the moisture content and bulk density (Table S3). (b) Chemical composition of MPs in each stratified layer of GY core. For the meaning of the abbreviations, please see SI. (c) Plastic production globally, in China, Fujian, and Xiamen versus time. Note the different scales of the y-axes. (d) Land-based plastic. The pink and blue lines represent the land-based plastic debris discarded into the environment in two scenarios: without and with considering the recycling and incineration rates of plastic waste, respectively. The green and brown lines represent land-based plastic disposed to the sea without and with marine debris collection, respectively. (e) Ship-based plastic waste. The orange, pink, blue, and green lines are the ship-based plastic waste using four different settings (30, 50, 70 and 100%) of reduction. (f) Wastewater-based MPs. The brown and blue lines represent two conservative scenarios to deduce the MPs fluxes; brown is with 0.19% annual growth ratio of plastic to urban solid waste.

2.2. Sampling treatment and core chronology

In the laboratory, PVC tubes were cut lengthwise using a core splitter (Geotek Ltd, UK), and then a thin iron-wire rinsed with ultrapure water was used to divide the sediment cores along the split line. The integrity, color change, and physical appearance of the core profiles were recorded on July 21–24, 2017. Given unsuitable chronologies (see below), only the GY core was fully processed. The integrity of the GY core was well retained. The sediment was mainly gray, argillaceous sand with coarse-grained sand between 17 and 27 cm depth. The top 1 m of the core was sliced into 1 cm samples, while the remaining core was sliced into 2 cm samples using stainless steel cutters, in which two aliquots were subsampled and preserved in aluminum foil bags preheated at 300–400 °C, with samples frozen before analysis. Note that the depths quoted in this study indicate the base of the 1 or 2 cm samples. To prevent contamination the slicing process was performed on a clean laboratory bench; the surface sediment was removed and then the undisturbed central portion of each layer was subsampled for MP analysis. We did not observe any MPs of the same white color as the PVC pipes. Additionally, the spectral fingerprint of particles derived from the PVC pipe was recorded and no matched spectra were found within the MP particles. These two results suggest that no traces of plastic particles from the PVC pipes contaminated the subsamples in undisturbed central portions of each core layer. The grain size (Fig. S4) and bulk density (Table S3) of the GY core was measured at 1 and 5 cm intervals using a laser particle analyzer (Malvern Mastersizer 2000, U.S.A.) and volumetric flask method, respectively. The procedure of MP extraction from the sediment core is detailed in SI (*MP sample treatment*).

Age models for three cores (HS, WY and GY) were determined via measurement of ^{210}Pb activity (Li et al., 2007; Wu et al., 2013; Zeng et al., 2015); HS-1 is dominated by medium and coarse sand (Table S1) and hence was unsuitable for ^{210}Pb dating. The presence of ^{137}Cs in the sediment was measured on an Ultralow-background HPGe spectrometer (0.26 cpm, 40–2700 keV) located in the China Jinping Underground Laboratory (CJPL) at a depth of 2450 m (Wu et al., 2013; Zeng et al., 2015). For details of dating, please see SI, *^{210}Pb and ^{137}Cs dating*, Fig. S2.

2.3. MP identification and quantification

The abundance, polymer composition, and size of MPs in the stratified sediment cores were characterized using FPA- μFTIR (see SI *MP identification and quantification* & Fig. S3 and Table S5). This method is robust for unbiased high-throughput detection of MPs on the filter (Löder et al., 2015), which greatly improves the accuracy of MP identification and the comparability of the results. To evaluate the background contamination during the sample treatment and the FPA- μFTIR measurement, and recovery rates of the method here, three blank samples and three spiked samples with polystyrene (PS) microspheres were randomly set up. The MPs averaged from the triplicate blank tests were used as background MP contamination (SI *Recovery rate and procedural blanks test* & Table S6), from which the identified MPs in stratified sediment samples were subtracted to yield the corrected MP numbers. The method for the calculation of MP abundance and depositional flux is provided in the SI (*MP abundance and depositional flux*).

2.4. The main sources of MPs and the Generalized Additive Model

The major indicators of regional human activities in relation to macro- and micro-plastics (SI, *The main sources of MPs*) were quantified through estimation of the registered population of Xiamen City and annual gross domestic product (GDP) (SI, *Economic and population growth in Xiamen City* Fig. S6). The land-based plastic waste fluxes into the bay from Xiamen City were estimated based upon per capita waste generation rates (PCWGR) and compared with income level changes with time (Fig. S7a). Vessel-sourced plastic debris was estimated based upon annual changes in cargo throughput to Xiamen Port, the degree of

containerization and the introduction of strengthened waste handling and disposal practices (Fig. S8). Wastewater-derived MPs were determined from estimates of urban wastewater generation volumes, linked with residential populations and increasing construction of wastewater treatment plants (WWTPs) after 1989 (Fig. S9).

A Generalized Additive Model (GAM, see SI, Fig. S10, Table S8) is applied for different scenarios to explore the coupling relationships between the temporal evolution of MPs and global, national, provincial (Fujian's), and local (Xiamen's) plastic production, land- and ship-based plastic wastes, and wastewater-sourced MPs.

3. Results

3.1. Sediment core dating

The ^{210}Pb activity of the investigated sediment cores is shown in Fig. S2. The $^{210}\text{Pb}_{\text{total}}$ of the HS and WY cores do not decrease significantly, indicating that they are greatly disturbed and cannot be accurately dated. The ^{210}Pb in the HS-1 sediment core was not measured due to the dominance of the medium and coarse sand. The sedimentation rate of GY is 0.49 ± 0.04 cm/yr in the top 50 cm (1916–2017), comparable to the published value in the same region in the 1990s of 0.43 cm/yr (Li and Li, 1991). The ^{137}Cs radioactivity in the GY core is below the detection limit, suggesting negligible ^{137}Cs deposition in this area since ^{137}Cs input from land freshwater, oceanic currents, or the atmosphere is small and unstable (Cai et al., 1988), but the inputs of ^{210}Pb from the atmosphere are more stable. The slight decadal variations in the sedimentation rate reflect a relatively stable depositional environment. Thus, the GY core is chosen as the target column for this study. Although the sedimentation rate of GY may include uncertainties, it can suggest an approximate time scale to aid the interpretation of the MP records.

3.2. Characteristics of MPs

3.2.1. The abundance of MPs

A total of 1838 suspected MPs were measured using FPA- μFTIR , of which 818 encompassing 16 polymer types (Table S5) were confirmed as MPs in the GY core, yielding a detection rate of 44.5%. The earliest record of MPs in the GY core appeared in the 32 cm layer (corresponding to ~1952), with $30,332 \pm 31,457$ items/kg-dw (Figs. 2a and S5). MP abundance declined significantly in sediments corresponding to the 1960s, with $6911 \pm 23,294$ items/kg-dw in 1968. In ~1970s sediments, MPs increased to a peak abundance of $189,241 \pm 29,495$ items/kg-dw in the mid-1980s. Subsequently, they dropped abruptly in sediments dated from the late 1980s to a mid-1990s abundance of $16,626 \pm 26,371$ items/kg-dw. MP abundance fluctuates in sediment from the 1990s to the 2010s (Fig. S5).

3.2.2. The polymer types of MPs

Sixteen types of polymers were detected, of which PES (mean 41.1%) and acrylic (23.1%) were dominant, followed by PE, PP, PE+PP, and PA (<20%, Figs. 1b and 2a,b). Phenolic was detected particularly between 40 and 32 cm depth (~1952–1936), with a mean proportion of 6.1%. Meanwhile, no phenolic compounds were detected in other layers except at 12 cm depth (~1992). At 20–14 cm depth (~1988–1976), PES accounted for 66.7–82.0% of the total MPs (72.4% on average, Fig. 3b), higher than the average proportion of PES (41.07%, Fig. 3a) throughout the entire core.

3.2.3. The particle size of MPs

The particle size of MPs spanned from 20 to 1355 μm for the whole GY core, of which 82.2% were <100 μm (Fig. 2c,d). The majority of MPs were in the size range of 20–40 μm , with sizes of 20–30 μm and 30–40 μm accounting for 20.3 and 19.7%, respectively. In particular, MPs of 20–40 μm seemed to accumulate in the 16 to 22 cm layers of the sediment core, where there was an abundance of MPs and a high fraction of

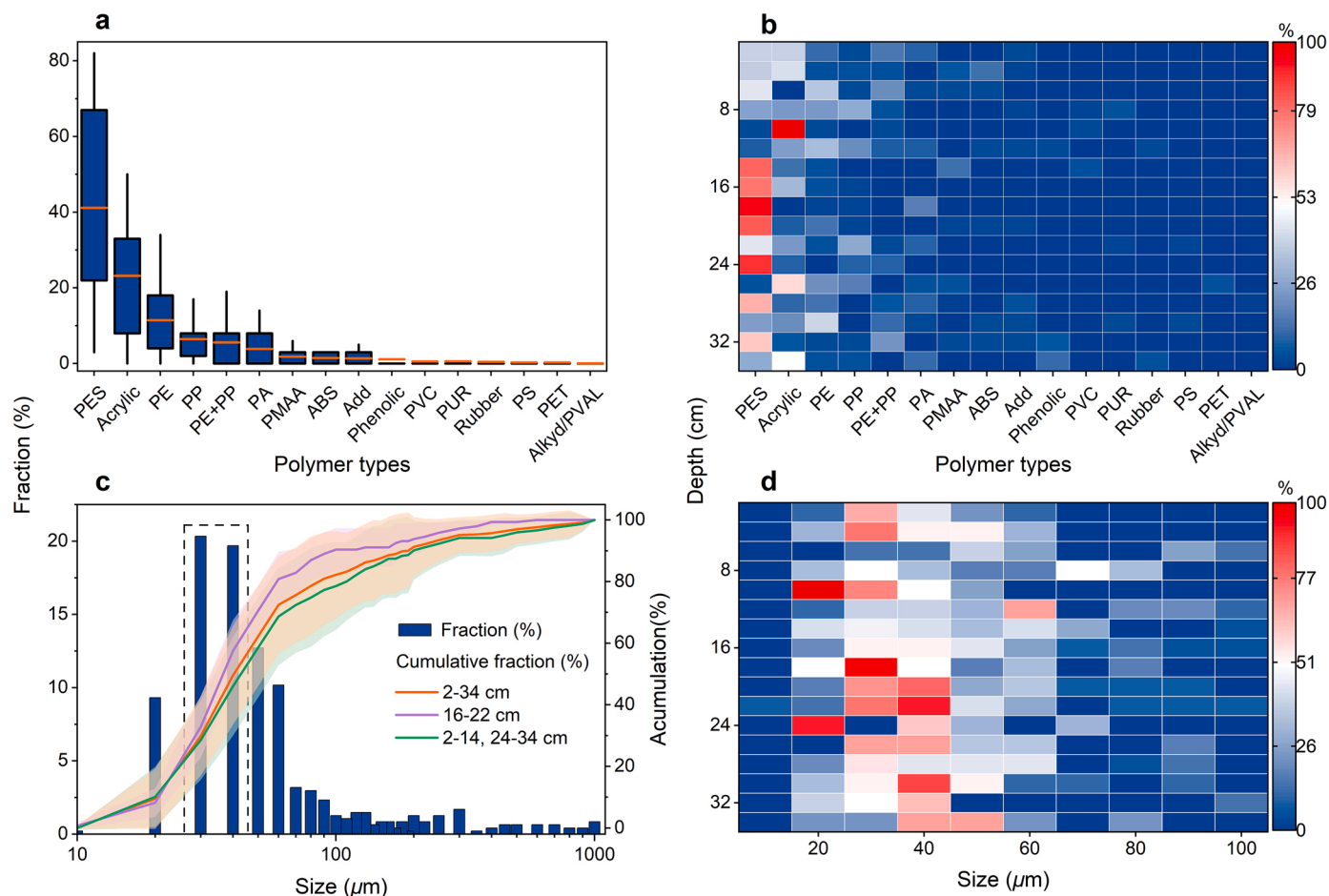


Fig. 3. Polymer composition and particle size distribution of MPs in the GY core. (a) Polymer composition, orange line is mean value. (b) Heat map of MPs composition as a function of depth. (c) Depth average MPs particle size distribution (left y-axis) and particle size cumulative fraction of MPs (right y-axis). MP size (X-axis) was displayed in logarithmic coordinates to show the size distribution of small particles. (d) Heat map of size (10–100 μm) distribution as a function of depth.

PES was also found (Figs. 1a,b and 2b). In particular, in the 16 to 22 cm interval of the GY core, the MPs are characterized by high abundance (189,241–88,276 items/kg-dw) and small particle size (30–40 μm) (Figs. 1a and 2d), which suggests a comparatively stable hydrodynamic environment with little disturbance by extreme events in this region. As in the estuarine environment, the increased bed shear stress generated by the amplified hydrodynamic forcing of waves and currents before (Zou, 2004; Zou and Hay, 2003), during and after extreme events would have transported a bulk of MPs, leaving behind only a small amount of MPs with larger particle sizes (Enders et al., 2019).

4. Discussion

4.1. The sedimentary environment in Xiamen Bay

The $^{210}\text{Pb}_{\text{ex}}$ activity profile in GY has maximum values immediately beneath the surface sediment (7–8 cm), indicating that the top layer may have been disturbed by currents or through bio- or anthro-turbation. Another contributory factor could be the labile organic matter diluting the $^{210}\text{Pb}_{\text{ex}}$ on the top layer not yet reduced to the stable residues (Appleby and Oldfield, 1978). The sediment is dominated by coarse grain sizes at depths of 16 to 27 cm, which might be assumed to be caused by changes in hydrodynamic conditions or extreme events (Fig. S4). The sediment grain size changes gradually, coarsening upwards in the depth interval of 27 to 23 cm and fining upwards between 17 and 13 cm depth (c.f. red arrows in Fig. S4a). The smooth variation in grain size indicates that the deposition is not related to a typhoon event, which

tends to generate a sharp-based bed of coarser sand. Additionally, the ^{210}Pb dating record shows that $^{210}\text{Pb}_{\text{ex}}$ at the layers of 14 to 15 cm, 25 to 26 cm, and 27 to 28 cm comply with the expected radiation decay rate (Fig. S2), suggesting the stability of sedimentary accumulation rates. The outliers of $^{210}\text{Pb}_{\text{ex}}$ (c.f. data point 16 & 22 cm in Fig. S2) at the 15 to 16 cm and 21 to 22 cm depth intervals may be attributed to the high content of coarse-grained sand (Fig. S4; Table S1), which has low adsorption capacity, thereby increasing the chances of dilution of $^{210}\text{Pb}_{\text{ex}}$ (Appleby and Oldfield, 1978).

Typhoons are one of the most devastating natural extreme events in Xiamen coastal areas, such as typhoons Meranti in 2016 (~0.5 cm depth), Dan in 1999 (~8.5 cm depth) and Pamela in 1961 (Ying et al., 2014) (~27.5 cm depth), directly hitting Xiamen and wreaking catastrophic damage. However, these extreme events, particularly Typhoon Meranti in 2016, did not cause significant changes in the (surface) sediment grain-size (Fig. S4e). This suggests the stability of the sedimentary environment in the well-sheltered study area, consistent with a prior report concluding that hydrodynamics are somewhat weak, and thus muddy deposition dominates in this area (Cai et al., 1999; Fang et al., 2010a). Collectively, the aforementioned evidence suggests little effect of hydrodynamics and extreme events (e.g., typhoons) on the observed distribution of coarse sediment grain size in the layer of 16 to 27 cm.

Given that provenance and hydrodynamics are typically the most crucial controlling factors for sediment distribution patterns (Enders et al., 2019), and having discounted the importance of hydrodynamics in the low energy study area, we hypothesize that the provenance is the

determining factor for the observed coarse sediment grain sizes at the sediment layer of 16 to 27 cm depth. The sources of the sediments in Xiamen Bay exhibited a wealth of terrestrial characteristics (Fang et al., 2010a). Fig. 1 shows the surface sediment migration pattern in the GY area, where sediments are transported westwards into the estuary north of the Jiuyu Nature Reserve, and out of the estuary to Xiamen Bay south of the Jiuyu Nature Reserve, and there is a convergence from three sediment-transport pathways (southwestward, northeastward, and eastward, *c.f.* arrows in Fig. 1c) (Cai et al., 1999; Fang et al., 2010b; Zuo et al., 2011). The MP in the study area was primarily affected by the population density and human activities around and within the bay. Before the 1960s (below ~27 cm depth), the population density was low in Xiamen Bay and economic activity was weak (Fig. S6b). Hence, the sediments within the bay were hardly affected by the coastal human activities, but were mainly influenced by the size-selective sediment transport, with a steady and long-distance transport of small grains from the upstream Jiulong River (Figs. S3 and S5). From 1960 to the early 1970s (~27 to 24 cm depth), the still underdeveloped Xiamen Bay led to limited coastline change within the bay in 1955–1970 (Fig. 1) and a gradual increase of grain size. From 1970 to 1984 (~24 to 16 cm depth), rapidly growing coastal development along Xiamen Bay (Fig. 1) and a lack of environmental protection measures and awareness collectively enhanced the coastal supply of sediment. These sediments are poorly sorted, coarse grains accumulated in the study area, causing as an abrupt change at ~24 to 16 cm depth in the GY core (Fig. S4). After the introduction of stricter environmental legislation in 1984 (Fig. 2d), the influx of nearshore sediments into the bay declined despite the growing coastal development, such as reclamation in 1984–2017 (Fig. 1). This human intervention facilitated a decrease in sediment grain size (16 to 13 cm depth) and a stable provenance (13 to 0 cm depth) (Fig. S4). Fig. 4a suggests that the correlation between high MP amounts and sand content is due to provenance, surface sediment transport pattern, and composition of terrestrial sediment near Xiamen Bay.

Salt wedge circulation is one of the controlling factors in the spatial pattern of macro- and micro-plastics on the seafloor (Acha et al., 2003; Simon-Sánchez et al., 2019), and the vertical distribution of MPs in the water column in estuarine areas (Defontaine et al., 2020). Compared to the provenance and hydrodynamics in the study area, the salt wedge is a secondary factor controlling the temporal distribution of MPs in the GY sedimentary core.

4.2. Stratigraphic record of MPs in Xiamen Bay

The stratigraphic profiles of MPs (including abundance, polymer types and sizes) in the GY core are the result of the comprehensive action of the MP provenance, physiochemical features and the sedimentary environment. Figs. S6c and S6d show the decadal variations in MP

abundance and depositional fluxes extracted from the stratigraphy of the GY core. The onset of MP appearance in the sediments coincided with that of the plastics industry in China (China Plastics Processing Industry Association, 2002) and with the proposed onset of the Anthropocene Epoch (Waters et al., 2016). Small decadal changes in sedimentary MP abundance and depositional fluxes during the 1950–1960s are observed, representing only small amounts of plastic entering the coast due to limited plastic production and an undeveloped consumer lifestyle with a low demand for plastic products (Fig. 2c). In contrast, large MP abundance and depositional fluxes are observed during the 1970–1980s due to rapid economic development, population expansion, and plastic industry growth. In the late 1980s and early 1990s, sedimentary MP abundance and depositional fluxes underwent abrupt declines due to mitigation actions to tackle plastic pollution worldwide, including the implementation of the Marine Plastic Pollution Research and Control Act (MARPOL) for the Prevention of Pollution from Ships. However, with time the decline was gradually overcome by the accelerated population and plastic industry growth, and modern consumer lifestyle that demands a myriad of plastic products. The MP abundance in the GY core has been rising slowly again since the 1990s, but at a rate less than that witnessed in the 1970s and early 1980s. It is possible that benefits from the MARPOL Annex V in 1988 and domestic environmental protection measures were counteracted by the sharp increase in domestic plastic output during the 1990s at an average annual rate of 15.34% (Anonymity, 2007).

The high fractions of PES and acrylic in the GY core are consistent with earlier studies showing that PES or acrylic-PES copolymers permeate in the subsurface ocean and deep-sea sediment, and account for a large proportion of the detected MPs (Kanhai et al., 2017; Woodall et al., 2014). The densities of pure PES and acrylic are 1.38–1.40 and 1.18–1.19 g/cm³, respectively, greater than the seawater density, so they tend to sink to the seabed. In contrast, despite considerably higher production than other polymer types, PE and PP are buoyant and typically found at the water surface or in the water column due to their low densities compared to sea water (0.91–0.97 g/cm³ and 0.89–0.91 g/cm³ for PE and PP, respectively). However, some PE, PP, and PE+PP copolymers can be found in the GY core, possibly due to increased densities and reduced re-suspension due to colonization of biofouling that facilitates their sinking and settling into the sediment (Chubarenko et al., 2016; Clark et al., 2016; Corcoran et al., 2015; Kaiser et al., 2017; Lusher et al., 2015; Semcesen and Wells, 2021; Van Cauwenbergh et al., 2013; Zhao et al., 2017).

Fig. 4 shows that the MP abundance has a significant positive correlation with the proportion of sand components in the sediments, which is consistent with the earlier results that high-density MPs revealed strong correlations with fine sand (Enders et al., 2019), and smaller MPs facilitated settling than larger ones under constant environmental

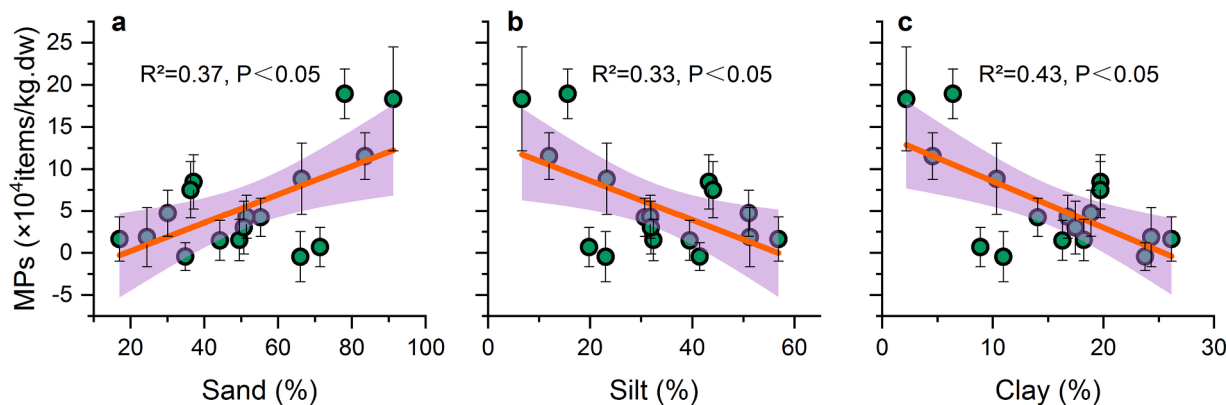


Fig. 4. Sedimentary MPs versus the grain size fraction in the GY core. (a) MPs v.s. sand, (b) MPs v.s. silt, and (c) MPs v.s. clay. The regression lines (orange) are bounded by 95% point-wise standard errors (purple area).

conditions (Semcesen and Wells, 2021). However, MP abundance was inversely correlated with silt and clay concentrations, suggesting similar sedimentation behavior of MPs and sand but different behaviors of MPs with silt and clay. The deposition of MPs has been studied in recent years (Defontaine et al., 2020; Semcesen and Wells, 2021), yet the potential MP-sediment relationship remains unclear (Enders et al., 2019; Kane and Clare, 2019). The sedimentation of MPs is different from that of natural particulates even under the same hydrodynamic conditions due to the low density of their constituent polymers, hydrophobicity, neutrality, and corrosion resistance (Browne et al., 2011; Waldschläger and Schüttrumpf, 2019). Therefore, the empirical rules of natural particulate deposition may not be applicable to MPs; for example, flocculation of clay was limited during MP deposition (Browne et al., 2010). Although the distribution and accumulation of MPs in marine sediment are influenced by various factors (Erkes-Medrano et al., 2015), sources and hydrodynamic regime have been identified as controlling factors (Enders et al., 2019). The distribution of MPs and their correlation with natural particulates in the profile of the GY core mirror the varying composition and intensity of MP sources in the different time intervals under a comparatively stable deposition environment. Considering the surface sediment migration trend in the study area and the fractionation of MPs during the depositional process due to differences in physico-chemical properties (size and density) that small-sized and high-density MPs deposited closer to their sources relative to large size and low-density ones (Defontaine et al., 2020; Semcesen and Wells, 2021), we believe that the main controlling factors come from the nearshore of Xiamen Bay rather than through variation in discharges from the Jiulong River.

4.3. MPs and plastic production

From the historical profile of MPs in the GY core, plastic pollution occurred in the early stage of plastic production and consumption in the region. Plastic waste may enter the environment coincidental with production (e.g., nurdles), but the mass of particulate plastic entering an estuarine/marine environment comes following use and disposal, with a delay caused by indirect transport pathways, involving repeated deposition and re-working before final burial. The MP concentration increased with time in parallel with increased global plastic production (Brandon et al., 2019; Dong et al., 2020; Matsuguma et al., 2017; Turner et al., 2019; Wang et al., 2018; Xue et al., 2020), but local contributions (e.g., local plastic production) have often been ignored. Fig. 5 shows the historical evolution of sedimentary MPs versus plastic production at local, regional, and global scales. Between 1950 and 1984, MPs were closely correlated with global and China's plastic production. Despite a global annual increase in plastic production, China contributed a limited share of global plastic outputs during this time (Fig. 2c). MPs in the core, therefore, appear to link more closely with global plastic production trends. The sporadic development of the plastic industry in Fujian Province and Xiamen City yields a weak association of sedimentary MPs with regional plastic production. Between 1988 and 2012, however, MPs were more related to local plastic production than to provincial or national plastic production. A sharp decline in the MPs was remarkable from 1984 to 1996 (Fig. 2a), despite growing plastic production. Hence, the observed stratigraphic record of the temporal distribution of MPs reflected exogenous sources of plastics up to 1984 and local sources from 1988 onwards. Fishing ropes are considered internationally to be an important source of MPs (Xue et al., 2020), but fishing in Xiamen Bay during recent decades has not been significant. GAM modeling results show that MPs increased nonlinearly with increasing Xiamen's plastic output, but the confidence intervals increased as well (Fig. 6a). This result suggests that local plastic production was not a major contributor to MPs in the marine sediment, and that other contributory factors (e.g., plastic waste from upstream cities and local sedimentation patterns) account for the primary temporal changes in MPs recorded in the sediment core.

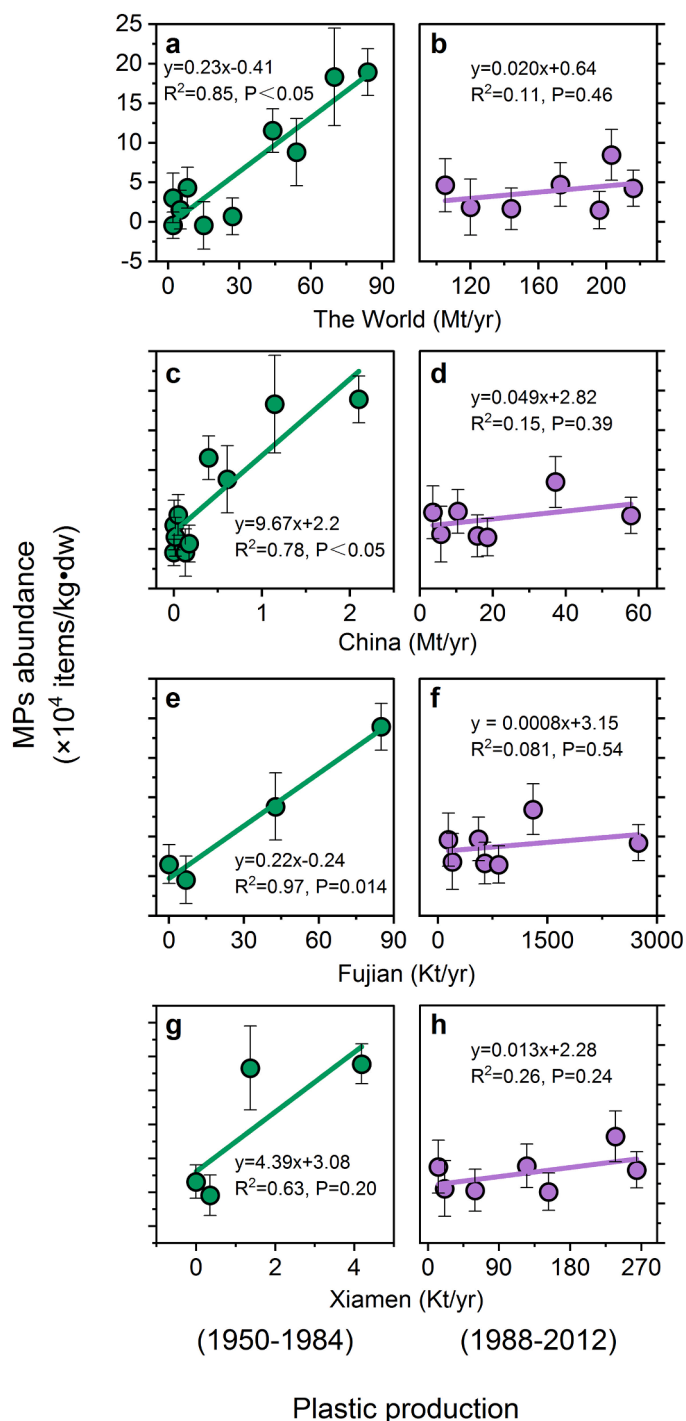


Fig. 5. Correlations between the abundance of MPs in GY core and plastics production in 1950–2016. (a, c, e and g) represent the correlation from 1950 to 1984 and (b, d, f and h) represent the correlation from 1988 to 2012, both for the World, China, Fujian, and Xiamen, respectively. Due to data availability, data on the world's and China's plastic production in 1950–1984 is available, but this is not the case for regional and local plastic production. Only a limited set of data on Fujian and Xiamen's plastic production is recorded in literature.

Comparison of MPs and production rates only provide a component of the story, a potentially significant correlation to investigate, if not least due to the abundance of production data compared with historical usage and waste management practices.

The evolution of MP composition with time (Fig. 2b) echoes the temporal variation patterns of plastic production and consumption

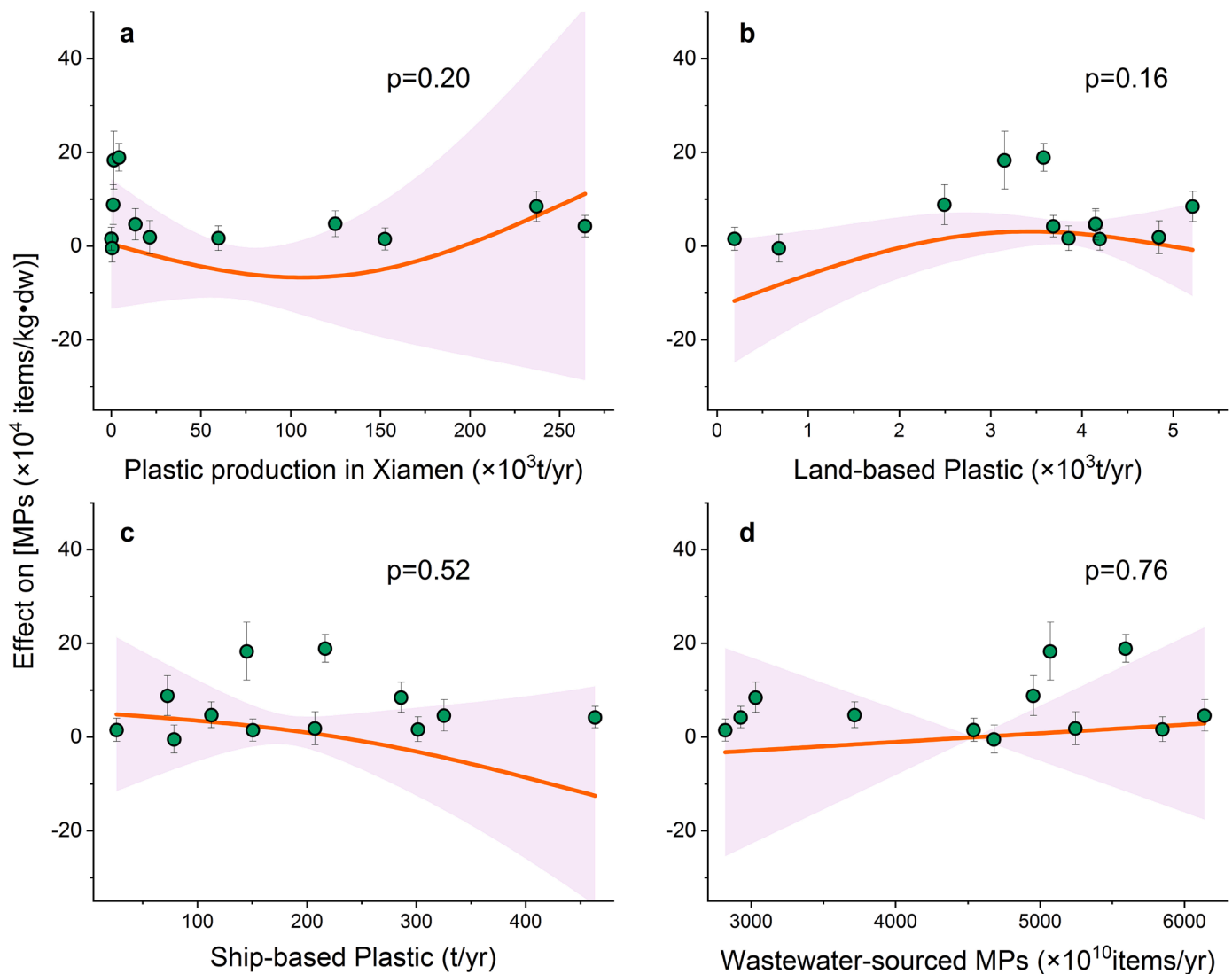


Fig. 6. GAM-derived effects on the MP abundance in the GY core. (a) of variables plastic production in Xiamen; (b) plastic debris from land; (c) plastic debris from ship littering; and (d) wastewater-sourced MPs. The orange lines are model fits based upon the GAM and the pink shading areas represent 95% confidence intervals; the green symbols are observational data.

(Fig. 2c). Although the earliest report of a phenolic production plant in China was from Shanghai in 1930 (China Plastics Processing Industry Association, 2002), details on phenolic output, scale, and distribution are still lacking. The mass product/usage of plastics in China started in the early 1950s when phenolics were the major plastic product. In 1955, China had only 703 small-sized plastic companies, 80% of which were phenolic plastic manufacturers. Thus, thermoplastics, such as PE and PP, were mainly imported (China Plastics Processing Industry Association, 2002). Phenolic, as one type of early plastic product, may provide a useful marker for the latest Holocene succession (i.e., pre-Anthropocene). However, the deposition records of phenolics pre-1952 were not discussed in this study because the available literature is insufficient to expand upon the interpretation of the observed data; it is meaningless to discuss this further without sufficient literature support. It is generally believed that the onset of the Anthropocene coincides with the "Great Acceleration" in the 1950s (Steffen et al., 2015; Waters et al., 2016), and plastic, as one of the significant stratigraphic markers of the Anthropocene, developed during this time in China. The literature and data records related to plastics have been gradually enriched from this interval. Therefore, the historical records of MPs in the Anthropocene stratigraphy and the literature after 1952 can be mutually corroborated to a certain extent. The Great Proletarian

Cultural Revolution (1966- to 1976) in China led to the stagnation of nearly all industries, including the manufacture of plastics. This may explain why MPs increased in the 1950s, followed by a downturn in the 1960s, although with the lowest concentrations in 1964 (Fig. 2a) predating this social stagnation.

Plastic pollution is affected not only by plastic production but also by the type of plastic products and technology development. China's economic reform and opening up, which started in 1978, facilitated the recovery of the plastic industry. For instance, during 1978 to 1984, China implemented >170 technological upgrading projects to replace wood with plastics (China Plastics Processing Industry Association, 2002). The explosive growth and a lack of environmental facilities and policies led to enormous emissions of plastic waste, which may have resulted in the rapid surge in the MPs in the 1970s and a peak of MPs in 1984 (Fig. 2a). The predominance of PES from 1968 to 1988 (Fig. 2b) may be explained by the thriving international imports of the plastics industry during the 1970s to 1980s, when PES was in large demand due to its popularity as clothing material and after 1976 for a growing Chinese demand for fashion. The types of plastic products are diverse, to meet the needs of not only people's daily lives but also industry requirements (China Plastics Processing Industry Association, 2002; Xiamen Special Economic Zone Yearbook Editorial Board, 1986).

From the 1990s, when there was an acceleration of plastic production nationally and locally in Xiamen (Fig. 2c), the export-oriented plastic production and extended plastic lifetime not only significantly reduced the discharge of plastics into the ocean (Fig. 2a) but also reflected changes in the nature of plastic consumption. PES decreased markedly after 1988, and the proportion of acrylic varied markedly from 86% of MPs in 1996 to absent in 2004 (Table S5), while PE, PP, ABS, and PE+PP at various times increased in concentrations, suggesting the enhanced diversity of MP polymer types due to changes in the plastic industry in Xiamen (Fig. 2b). PE, PP, and PVC were produced massively for construction, packaging, and daily life during that time. Foreign advanced plastic production technology and machinery introduced to Xiamen in 1984 included the production of PP woven bags, PVC food-grade cling film, PVC corrugated pipes, and plastic sheets (Xiamen Special Economic Zone Yearbook Editorial Board, 1986), although PVC is not a significant component in the GY core after 1984.

4.4. MPs and plastic waste regulations

Changes in plastic waste regulation also explain some of the fluctuations in historical MP records in Xiamen Bay marine sediments. Fig. 6b shows that MPs in the GY core correlate with the net flux of land-based plastic debris into the sea ($p = 0.16$), suggesting the key role of control measures of terrestrial waste to offshore sedimentary MPs. The generated municipal solid waste (MSW) in Xiamen increased from 9.0×10^4 to 1.9×10^6 metric tons (tonnes) between 1950 and 2016 (Fig. S7c), during which plastic waste increased from 360 to 2.1×10^5 tonnes (Fig. S7e). However, the volume of land-based plastic debris discarded did not follow an upwards trend (pink line in Fig. 1d; see also S8e, & S8f). This was due to the improved management of MSW after 1980, which resulted in more MSW being collected than discarded from 1985 (Fig. S7d). Counting the increased global recycling and incineration rates of plastic waste (Geyer et al., 2017), and strengthening environmental legislation in Xiamen City after the 1980s, the amount of plastic debris discarded onto the land and ultimately dumped into the sea fluctuated but remained broadly constant (shown by blue and green lines in Fig. 2d, respectively; see SI *Land-based plastic fluxes into Xiamen Bay* for details). Furthermore, since 1996 marine debris, most plastic debris, has been routinely collected along Xiamen's 52 km² coastline (Su et al., 2011). Approximately 184 tonnes of marine debris were collected each month from the sea surface of Xiamen Bay between 2009 and 2018 (Xiamen Marine and Fisheries Bureau, 2018). The mitigation efforts for plastic pollution have greatly slowed the growth rate of the net flux of land-based plastic debris into the sea. Despite an overall increase from 160 tonnes in 1950 to 4884 tonnes in 2016 (brown line in Fig. 2d), the proportion of plastic waste influx to the sea compared with the generated plastic waste has decreased from 43.7% to 2.32%. The effectiveness of regulatory efforts to reduce plastic pollution, however, has been overpowered by plastic production and consumption, leading to the massive accumulation of plastic waste in regional marine environments (Lau et al., 2020).

Ship-derived litter is a major source of plastic pollution in the ocean (Hagen, 1990; Horsman, 1982; Ryan et al., 2019); however, quantifying the volume of ship-generated plastic debris is challenging (Cantin et al., 1990). Here, we estimated the input of non-container ship-based plastic debris to Xiamen Bay, assuming three settings of reduced discharges (Fig. 2e, see SI *Vessel-source plastic debris to Xiamen Bay*). Using GAM simulation, we found that ship littering played a role in marine sediment MPs (Fig. 6c). Particularly before 1989, the MPs in the GY core showed a positive correlation between increased ship-based plastic waste littering and rising cargo throughput in Xiamen Port (Fig. S8c). Despite the rapid development of Xiamen Port, between 1989 and 2018 the cargo throughput increased by ~42 times, plastic waste from ships entering the sea has not increased concomitantly and at times has shown rapid declines. This may be due to the rapid development of containerization in Xiamen Port after 1983. The container throughput of Xiamen Port

grew by 29.5% per annum, from 3300 to 1.07×10^7 TEU (twenty-foot equivalent unit) between 1983 and 2018 (Fig. S8a), accounting for 2.1 to 83.6% of the total port throughput, respectively (Xiamen Municipal Bureau of Statistics, 2019). Nearly all garbage from container cargo is disposed onshore, and thus the rapid development of containerization resulted in a sharp decline in ship plastic littering (Cantin et al., 1990). Additionally, the laws and regulations banning ship plastic waste further reduced the vessel-source marine litter process. China started to enforce the Marine Environment Protection Law in 1983, which prevented vessel-based waste dumping (The World Bank, 1992). Subsequently, China's regulation of control over dumping wastes into the sea was enforced in 1985 (Food and Agriculture Organization of the United Nations, 2019). MARPOL Annex V prohibiting direct marine plastic debris dumping, came into force in 1988 (Ninaber, 1997; Sheavly and Register, 2007). Although the legal validity of MARPOL is still controversial, a large number of studies have documented its effectiveness in reducing plastic waste from ships into the ocean (Cantin et al., 1990; Edyvane et al., 2004; Johnson, 1994). Xiamen is one of the first six ports in China to establish onshore ship-generated waste reception and handling facilities (The World Bank, 1992). The GAM simulation results (Fig. S9, Table S8) also showed that the number of ships that achieved zero emissions of plastic waste after 1989 may be closer to the real scenario, suggesting successful implementation of MARPOL Annex V in Xiamen Port. The rigorous enforcement of Annex V, as well as national and regional environmental laws and regulations collectively bar the direct dumping of ship-generated plastic waste in Xiamen Bay, and emissions are modeled as zero by 2016 (Fig. 2e).

Fig. 6d confirms that municipal wastewater was one of the main contributors to historical marine MP pollution. Before 1989, lacking wastewater treatment plants (WWTPs) in Xiamen, untreated urban wastewater was directly discharged into the coast, providing an estimated MP influx of $4.4\text{--}6.1 \times 10^{13}$ items/yr to Xiamen Bay during 1950 to 1989 (Fig. S9b). Especially during 1980 to 1989, the mass production and use of PES collectively led to the large discharge flux of wastewater-based MPs, which may be one of the main reasons for the observed peak of MPs in marine sediments during that period. From 1989, with the establishment of WWTPs in Xiamen, the wastewater treatment rate changed from 18.2% to 96.02% between 1989 and 2018, preventing 1.38×10^{13} to 1.97×10^{14} items (20.9%–87.4%) of MPs from flowing into the sea each year (the green part in Fig. S9b). Assuming the absence of WWTPs and other sources, we estimate that the abundance of MPs in sediment would reach 2.2×10^5 and 3.2×10^5 items/kg-dw in 2012 and 2018, respectively, one order of magnitude higher than the current values. Despite the important role of WWTPs in reducing marine MP pollution, we cannot neglect the MP outputs from continuous wastewater effluent emissions (Long et al., 2019).

5. Conclusion

The evolution of plastic production, use and waste management is recorded by the temporal variations in the abundance, size distribution, and chemical composition of MPs in a marine sediment core from Xiamen Bay. The stratigraphic profile of MPs is the result of the combined influence of multiple factors, including MP provenances, physicochemical properties, and sedimentation environment. Their appearance and abundance provide a useful marker for recognizing the mid-20th century beginning of the Anthropocene Epoch and component Anthropocene strata through to the present day. Local populations started to commonly use plastic, for example, the onset of nonphenolic MPs in the sediment core (*c.f.* 1952), despite no known local production source at that time.

Despite the production of plastic at the global to national scales showing an accelerating curve since 1950, the sedimentary record of MP abundance may be used as potential chronostratigraphic horizons; a dip in 1960s values is considered to result from the effects of the Cultural Revolution in China, and a peak value in the mid-1980s following the

political drive for industrialization. MP abundance has declined rapidly since the late 1980s through improved management of municipal solid waste on land, introduction of wastewater treatment plants and regulation of plastic waste discarding by ships. Over the past two decades, MP pollution has started to slowly rise again. Combined with the evidence of stratigraphic data, targeted measures are clearly necessary, including banning nonessential and single-use plastics, efficient waste management, and plastic removal, to mitigate MP pollution and to lessen the impacts of development in the Anthropocene.

The potential uncertainty associated with the chronology of the GY core in this study may be reduced by adopting a more robust sediment core chronology to link socioeconomic patterns of plastic production, use and waste management with MP stratigraphic record in future research. It was assumed that MPs remained stable without significant geochemical changes and vertical migration once they were buried in sediments, and taphonomic processes should be considered to extract the historical MP records for future studies (Bancone et al., 2020). Although MPs have been recognized as an Anthropocene marker, the longevity and potential fragmentation of MPs in sediments are not well understood (Waters et al., 2018). The deposition of MPs from water column to seabed, the resuspension and burial and aggregation of MPs under the action of turbulence and ambient flow play an important role in MP distribution in the sediment core especially in high energy environments (Peng et al., 2018; Waldschläger and Schüttrumpf, 2019) also worth further investigation. In low-energy coastal environments close to urban and industrial areas, such as the present study area, however, sources are the most critical influencing factor on the long-term trend of MPs in marine sediment. Due to the complexity of human and natural activities in coastal areas, the sources of MPs in marine sediments are diverse and uncertain and not fully understood. The regional and global applicability of MPs as markers of the Anthropocene Epoch shall be investigated through global collaborative efforts.

CRedit authorship contribution statement

Zouxia Long: Conceptualization, Visualization, Methodology, Data curation, Formal analysis, Writing – original draft. **Zhong Pan:** Conceptualization, Visualization, Writing – original draft. **Xianglong Jin:** Validation, Writing – review & editing. **Qingping Zou:** Formal analysis, Writing – original draft. **Jianhua He:** Methodology. **Weiwen Li:** Formal analysis. **Colin N. Waters:** Validation, Writing – review & editing. **Simon D. Turner:** Validation, Writing – review & editing. **Juliana A. Ivar do Sul:** Validation, Writing – review & editing. **Xingguang Yu:** Validation, Writing – review & editing. **Jian Chen:** Resources, Formal analysis. **Hui Lin:** Supervision. **Jianye Ren:** Validation, Writing – review & editing.

Declaration of Competing Interest

The authors declare no competing interests.

Data availability

Data will be made available on request.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.watres.2022.119215.

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